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HEAVY WATER AND NONPROLIFERATION
Topical Report

by

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AND
NONPROLIFERATION

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N O T I C E

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A B S T R A C T

The following report is a study of various aspects of the relationship between heavy water and the development of the civilian and military uses of atomic energy. It begins with a historical sketch which traces the heavy water story from its discovery by Harold Urey in 1932 through its coming of age from scientific curiosity to strategic nuclear material at the eve of World War II and finally into the post-war period, where the military and civilian strands have sometimes seemed inextricably entangled. The report next assesses the nonproliferation implications of the use of heavy water-moderated power reactors; several different reactor types are discussed, but the focus is on the natural uranium, on-power fueled, pressure tube reactor developed in Canada, the CANDU. The need for and development of on-power fueling safeguards is discussed in some detail. Also considered is the use of heavy water in plutonium production reactors as well as the broader issue of the relative nuclear leverage that suppliers can bring to bear on countries with natural uranium-fueled reactors as compared to those using enriched designs. The final chapter reviews heavy water production methods and analyzes the difficulties involved in implementing these on both a large and a small scale. It concludes

with an overview of proprietary and nonproliferation constraints on heavy water technology transfer.

Our major conclusions are as follows:

1. On-power fueling of CANDU reactors leads to special, well recognized safeguarding problems. These have been addressed by a safeguards development program, encompassing both systems analysis and hardware development, jointly sponsored by Atomic Energy of Canada, Ltd. (AECL), the Canadian Atomic Energy Control Board (AECB), and the International Atomic Energy Agency (IAEA). The approach involves surveillance, containment, and item-counting of irradiated fuel bundles. Although the complete system has not as yet been tested on an operating reactor, it appears to be a good example of "proliferation resistance engineering." The major problem may be the political one of obtaining agreement to incorporate the system in operating reactors and those under construction.

2. The question of relative leverage on natural uranium vs. enriched uranium fuel cycles does not have a neat answer. At the moment, most of the countries of proliferation concern have neither large amounts of uranium ore nor the ability to enrich it. (There are, of course, some significant exceptions, the most obvious being South Africa which has both.) In the near term, the chances of achieving a consensus among current suppliers of separative work, all of whom belong to the London Club, not to supply it in the event of violations of nonproliferation agreements, also seems greater than the

prospects of reaching a similar agreement among all countries who might be able to supply uranium ore. If we assume in addition that the malefactor also can produce heavy water--no small matter--the potential leverage advantage would seem to lie with enriched reactors. On the other hand, the spread of enrichment technology--which is easier to rationalize on civilian grounds if enriched reactors are in place--could tip the scales the other way. In general, however, this weighing of enriched vs. natural uranium fuel cycles is unnecessarily restrictive. Experience has shown that there are many potential levers--nuclear and non-nuclear--which can be used to persuade countries to adhere to nonproliferation norms. The heart of the matter is the political will to use these in the face of conflicting policy objectives.

3. Unlike uranium enrichment via gaseous diffusion and the gas centrifuge, key aspects of which are closely held on nonproliferation grounds, techniques for heavy water production, particularly by hydrogen sulfide-water exchange (the GS process), have been extensively documented in the open literature. Nevertheless, construction and operation of large plants are difficult, and thus there is good reason to believe that the technology will not spread rapidly through the indigenous efforts of developing countries. Unlike uranium enrichment and fuel reprocessing, heavy water production does not provide a direct route from civilian fuel cycle to weapons-usable materials; on these grounds a logical quid pro quo for

its transfer would be adherence to the Non-Proliferation Treaty (NPT) or acceptance of the principle of full-scope safeguards by the recipient.

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"I told the President in general terms of the great progress we had made, and that our scientists were now definitely convinced that results might be reached before the end of the present war. He said his people were getting along too, but no one could tell whether anything practical would emerge until a full-scale experiment had been made. We both felt painfully the dangers of doing nothing. We knew what efforts the Germans were making to procure supplies of 'heavy water'--a sinister term, eerie, unnatural, which began to creep into our secret papers. What if the enemy should get an atomic bomb before we did! However sceptical one might feel about the assertions of scientists, much disputed among themselves and expressed in jargon incomprehensible to laymen, we could not run the mortal risk of being outstripped in this awful sphere."

Winston Churchill, recounting his conversation with Franklin Roosevelt at Hyde Park on June 20, 1942. In: The Hinge of Fate (Houghton Mifflin Co., Boston, 1950) p. 380.

1. THE HEAVY WATER CONNECTION

1.1 Discovery of Deuterium and Early History

In 1932, two fundamental discoveries in the field of nuclear physics were reported, that of the neutron by Chadwick¹ in England and a stable isotope of hydrogen of atomic weight two by Urey, Brickwedde and Murphy in the United States.² Their existence had been speculated about for many years, and the experimental confirmation initiated an intense period of scientific activity which in turn led to many important results of both a basic and applied nature. In this paper we concentrate on deuterium (D)--the name given to the hydrogen isotope by Urey and his co-workers--and, in particular, on the heavy isotope of water, heavy water (D_2O). However, the neutron is an essential element in the story since it is the interaction between the neutrons produced by the fission of uranium and heavy water which forms the basis for our interest in the latter.

In 1911 Sir J.J. Thomson, the discoverer of the electron, reported on a series of experiments in which he observed two varieties of positive ions of molecular weight three³ using an electromagnetic separation technique. In retrospect, it seems clear that these were ions of the monodeuterated hydrogen (HD) and tritium molecules (T),⁴ although an unequivocal claim was not made at the time. Besides Thomson's pioneering studies only two investigations relating to the possible existence of deuterium were reported up to 1927, when the matter seemed to be conclusively settled--in the negative--by Aston's mass

spectrographic determination of the atomic weight of hydrogen in agreement with the atomic weight determined by chemical means.⁵ However, the discovery of an oxygen isotope of atomic weight roughly equal to 18 by Giauque and Johnson in 1929⁶ reopened the question, since Aston's atomic weights were based on the mass 16 isotope of oxygen taken as exactly 16, while the chemical atomic weights are based on the natural isotopic mixture taken as exactly 16. The determination in 1931 of the relative abundance of the O^{16} and O^{18} isotopes⁷⁻⁻¹⁷ had not as yet been detected--led Birge and Menzel to suggest⁸ that the resulting discrepancy in the chemical and mass spectrographic atomic weights could be removed by postulating the existence of an isotope of hydrogen of mass 2, with a relative abundance $H/D = 4500$. They also remarked that it should be possible, although difficult, to detect such an isotope by means of band spectra.

The experimental challenge was soon taken up by Urey and his co-workers at Columbia, who predicted that considerable differences in the relative vapor pressures of H_2 , HD and HT in the solid state should exist, and hence that the fractional distillation of liquid hydrogen near its triple point should be an effective method for the concentration of any heavier isotopes. The experimental procedure was to photograph lines in the atomic spectrum of ordinary hydrogen and the residue from the evaporation of 4 liters of liquid hydrogen. In both cases spectral lines characteristic of deuterium were detected, although they were much more intense for the hydrogen enriched

by distillation. The natural abundance of deuterium was found to be one part in 4000. No evidence for tritium was found.

The discovery of deuterium was rapidly confirmed by other investigators, and over the next three years, some 200 papers dealing with its production, properties and use in research in physics, chemistry, and biology were published. Since the immediate problem was to get enough heavy water to conduct experiments, considerable effort was devoted to devising efficient separation techniques. Of these, electrolysis of water--utilizing the fact that the hydrogen developed at the cathode of an electrolytic cell contains less deuterium than the water from whence it came--was found to be the most convenient and rapid method for the production of relatively large quantities of heavy water. The concentration of deuterium could be increased to any desired level by either continually electrolyzing the water in a single cell down to an ever smaller volume, or by progressive enrichment in a cascade, each of whose stages consists of a group of cells connected in parallel, whose number decreases as the deuterium concentration increases (see Fig. 1). The former technique was first suggested by Washburn and Urey⁹ who produced water containing a few percent of deuterium; subsequently, Lewis and McDonald¹⁰ obtained 0.1 gram of practically pure D_2O as the residue of electrolysis of 20 liters of water. (Since it takes about 6.6 KWh to electrolyze one liter of water, the unit energy consumption of this product was approximately 132 KWh/gram.) The cascade technique was

used in various laboratory-scale production plants; e.g., at Columbia University, natural water pre-enriched to 0.5% in the large electrolytic cells of the Ohio Chemical Co. could be brought to a concentration of 99.9% in four stages.¹¹ Although approximately 20 universities in the US and Europe were producing small quantities of heavy water by 1934, it was an expensive and somewhat hazardous undertaking--due to the potential for explosive recombination of the hydrogen and oxygen--and to meet the increasing demand, several companies undertook commercial production. Besides Ohio Chemical, there was Imperial Chemical Industries, Ltd., in England, and Norsk Hydro in Norway.¹² Due to the high capital costs and heavy energy consumption associated with electrolysis, large scale production of heavy water is economic only if the raison d'être of the electrolysis plant is not heavy water, but hydrogen for use as a feed material in ammonia synthesis. That is, electrolytic heavy water separation is a "parasitic" process, with the amount of D₂O produced limited by the magnitude of the associated hydrogen production. In this sense, the Norsk Hydro electrolysis plant at Vermok was--literally--ideally situated, on the side of a rockface below the huge Rjukan Foss waterfall which provided--via an adjacent generating station--approximately 100 megawatts of cheap hydroelectric power for the production of about 20,000 m³ of hydrogen per hour.¹³ To effect heavy water production the electrolytic cells were arranged in a nine-stage steady-flow cascade. About 73% of the water

fed to each stage was electrolyzed; the remaining 27%, which accompanied the hydrogen and oxygen produced in that stage as steam, was condensed and fed to the next higher stage. (In cascade terminology, the cut, θ , defined as the ratio of product to feed per stage, was 0.27.) The output of the primary plant was water containing 15% D. To produce material of higher concentrations, this product was fed to a specially designed secondary plant, which also consisted of nine stages. In order to increase the amount of deuterium produced, the partially-enriched hydrogen from each stage was burned and the resulting water recycled to the earlier stages, The facility could supply product of various concentrations, up to pure ($\geq 99.5\%$) D_2O , and was the largest producer in the world. Between the start of operations in late 1934 and 1938, 40 kilograms of heavy water were produced. By late 1939 heavy water production was about ten kilograms a month, and the inventory of pure D_2O was about 185 kilograms. But this is getting a little ahead of our story.

Besides electrolysis and fractional distillation of liquid hydrogen, early experiments indicated that fractional distillation of water, gaseous diffusion, adsorption on surfaces, and isotopically selective chemical reactions might be used to separate deuterium.¹⁴ In particular, Farkas and Farkas¹⁵ pointed out that the equilibrium constant of the isotopic exchange reaction between hydrogen and water



differs sufficiently from unity at low temperatures to permit effective separation. Unfortunately, because of the low solubility of hydrogen gas in water the approach to equilibrium in this reaction is a slow process, and it was realized that catalysis would be required for its practical utilization. However, because of the availability of sufficient heavy water for laboratory studies via electrolysis, none of these alternative methods were actively pursued until the discovery of fission suddenly made the need for large amounts of pure D_2O a matter of high priority.

The discovery which led directly to the convergence between the neutron and heavy water had initially nothing to do with either. In January 1934,¹⁶ Irene Curie and Frederic Joliot observed that when the light elements beryllium, boron, and aluminium were bombarded with alpha particles, positrons were emitted, and that the emission continued after the alpha source was turned off. At the Physics Institute of the University of Rome, Enrico Fermi reasoned that this artificial radioactivity might be induced more effectively by the neutral neutron than the charged alpha particle, and in March 1934-- in collaboration with a brilliant research team which eventually included E. Amaldi, O. D'Agostino, B. Pontecono, F. Rosetti, and E. Segré--he began a systematic study of artificial radioactivity produced by neutron bombardment. The behavior of

both light and heavy elements, including uranium, was investigated, and although the discovery of uranium fission was missed,¹⁷ this work laid the foundation for the field of neutron physics, stimulated the efforts of other researchers in the US, Europe, and Japan, and won a Nobel prize for Fermi in 1938. Early in the research program, Amaldi and Pontecorvo accidentally discovered that silver irradiated on wood tables became much more active than when it was irradiated on other marble tables in the same room. To clear up the mystery, preparations were made for systematic observations involving; e.g., the effect on the activation of interposing a lead filter between the neutron source and the detector. As recounted by Segre¹⁸

That morning [October 22, 1934] Fermi suddenly decided to try filters of light elements instead of the prepared lead filter. Paraffin was tried first. Although the experiment was performed during the examination period and several of us were absent, our friends Persico and Rossi were kibitzing on our work. About noon, we were all summoned to watch the miraculous effects of the filtration by paraffin. At first I thought that some counter had gone wrong because such strong activities had seldom appeared before. But it was immediately demonstrated that this strong activation was the result of filtering the radiation producing the radioactivity through paraffin. All this happened around noon. We tried a few more substances as filters and recognized that the powerful effect occurred only with paraffin. We went to lunch extremely puzzled by our observations and came back around three o'clock, after our usual siesta, to find that Fermi in the meantime had found the explanation of the strange behavior of filtered neutrons. He hypothesized that the neutrons could be slowed down by collisions and in this way become more effective, an idea which was contrary to our expectations.

Fermi himself later described the circumstances surrounding the seminal discovery in a conversation with S.C. Chandrasekhar.¹⁹

I will tell you how I came to make the discovery which I suppose is the most important one I have made. We were working very hard on the neutron-induced radioactivity, and the results we were obtaining made no sense. One day, as I came to the laboratory, it occurred to me that I should examine the effect of placing a piece of lead before the incident neutrons. And instead of my usual custom, I took great pains to have the piece of lead precisely machined. I was clearly dissatisfied with something: I tried every "excuse" to postpone putting the piece of lead in its place. When finally, with some reluctance, I was going to put it in its place, I said to myself, 'No, I do not want this piece of lead here; what I want is a piece of paraffin.' It was just like that: with no advanced warning, no conscious, prior, reasoning. I immediately took some odd piece of paraffin . . . and placed it where the piece of lead was to have been.

The discovery of the "hydrogen effect" and Fermi's hypothesis to explain it were reported immediately in a note to the Italian journal Ricerca Scientifica;²⁰ subsequent experiments confirmed that the results could be explained on the basis that:²¹

(a) the scattering of neutrons by protons occurs as if both are elastic spheres. It immediately follows that in a collision with a proton at rest, an energetic neutron can lose a variable, but in general large fraction of its kinetic energy. Therefore, provided the cross section for elastic scattering is much larger than cross section for neutron capture by the proton, the energy of the neutron can be reduced enormously by a succession of random elastic collisions with the hydrogen nuclei of the "moderator"--as substances

which slowed down neutrons came to be known--before being captured in the target nucleus.

(b) when the velocity v of a neutron is such that its deBroglie wavelength $\lambda = h(mv)^{-1}$ (m = neutron mass) is large compared with the range of the nuclear force, the probability P that the neutron will be absorbed or captured by the nucleus should be independent of v . Expressing P as a function of the capture cross section σ_a (cm^2), the neutron density n (number per cm^3), and v

$$P = n \cdot v \cdot \sigma_a, \text{ or} \quad (2)$$

$$\sigma_a = \frac{P}{n} \frac{1}{v} \quad (3)$$

one arrives at the so-called " $\frac{1}{v}$ -law;" i.e., a capture cross section for slow neutrons inversely proportional to the velocity of the neutron. This result is equivalent to the statement that the mean life of slow neutrons against capture in a medium of m nuclei per cm^3 , $\tau = (m \cdot \sigma_a \cdot v)^{-1}$, should be independent of velocity.

Deviations from the " $\frac{1}{v}$ -law" were subsequently found by Fermi and other workers. The phenomena could be explained by postulating the existence of relatively narrow absorption bands characteristic of various elements in which neutrons of definite velocity or energy (groups) would be selectively captured. The analysis of the neutron energy spectrum into

groups which are approximately homogeneous with respect to their absorption properties proved to be very fruitful.

Heavy water entered this picture quite naturally. That is, given the billiard-ball collision model of the slowing down of neutrons in hydrogenous substances, one should expect a similar, though less pronounced effect with substances of higher atomic weight. Besides atomic weight, the magnitude of the slowing-down effect should also depend on the elastic scattering cross section since, roughly speaking, the neutron cannot be slowed down if it doesn't interact strongly with the moderating nuclei. To test this hypothesis, the relative efficiency of H_2O and D_2O in slowing down neutrons and hence enhancing the artificial radioactivity produced in silver was investigated by several experimental groups in 1935. In particular, Dunning and co-workers at Columbia²² found a relative H_2O to D_2O enhancement of 5.5, and concluding that this was too large to be explained on the basis of the difference in atomic weight between H and D, they ascribed the effect to a much smaller interaction between neutron and deuteron than between neutron and proton. The paper reported fast and slow neutron-nucleus collision cross sections measurements for practically all the elements in the periodic table; however, the experimental procedure gave only the sum of the elastic scattering and capture components of the total cross section, and hence it was not clear to what extent the

slowing down process in the lighter elements was effected by the competition between scattering and capture. This was largely resolved over the next two years by the work of Fermi and his co-workers in Italy, Frisch, Halban, and Koch in Denmark, and Kikuchi, Aoki, and Takeda in Japan. Fermi, in a theoretical treatment of the problem of slow neutron capture by a proton with the formation of a deuteron and the emission of a γ quantum,²³ showed that the assumption that the process takes place via oscillation of the magnetic moment of the neutron-proton system leads to a value for the neutron mean life against capture in agreement with the experimental result measured a few months previously by his group.²⁴ In further work with Amaldi,²⁵ he determined a value for the capture cross section of $0.31 \times 10^{-24} \text{ cm}^2$ at a neutron velocity of $2.5 \times 10^5 \text{ cm sec}^{-1}$. The important experiments of Frisch et al.²⁶ not only confirmed this result, but also gave upper bounds for the slow neutron capture cross section of deuterium, carbon, and oxygen of 0.03, 0.01, and $0.01 \times 10^{-24} \text{ cm}^2$, respectively. Kikuchi et al.²⁷ independently arrived at the same upper bound for capture by deuterium, but their work does not seem to have been as widely known as that of Frisch.

In summary, by the time that uranium fission was discovered late in 1938 by Hahn and Strassman,²⁸ heavy water could be obtained in relatively small (\sim kilogram) quantities at a high price,²⁹ and much work had been done to ascertain its physical, chemical and biological properties. Its efficacy

as a neutron moderator vis à vis ordinary water and carbon was less well known. However, it was obviously better than carbon in its slowing down power, and the evidence of low capture cross sections for deuterium and oxygen obtained by Frisch et al. indicated a clear and perhaps marked superiority in this respect as compared with H_2O . The practical significance of having better answers to these questions would soon be apparent.

1.2 Fission and the War

Despite its efficacy as a neutron moderator, the basic role that heavy water played in the historic effort to build an atomic bomb during World War II was that of an understudy, waiting in the wings should other methods for producing the required fissionable material fail. After some important early work in France and England on chain reactions in homogeneous mixtures of natural uranium and heavy water, the decision was made to centralize the Allied bomb effort in the U.S., and here a decision to pursue vigorously the possibility of graphite rather than heavy water moderation in a natural uranium reactor to produce plutonium had been made early and was ultimately successful. The two main scientific actors in this decision, Fermi and the brilliant Hungarian physicist, Leo Szilard, were not unaware of the virtues of heavy water. However, given the felt urgency to proceed, and the greater availability of large amounts of graphite of the required purity as compared with heavy water, it seemed logical to first try the former. This came after the demonstration by the Columbia University group led by Fermi and Szilard as well as by the French team of Joliot and his co-workers that ordinary water captured too many neutrons to maintain a self-sustaining chain reactor with natural uranium. Given the subsidiary role that heavy water played, it is not surprising that most accounts of the history of the bomb-- especially those written from a US perspective--concentrate on those techniques which were ultimately successful in producing

fissile materials, plutonium and highly enriched uranium. However, both the political and technical aspects of the heavy water story during this period have been treated in some detail in several books;^{13, 30, 31, 32} here we sketch briefly the key element of the story and refer to these books and the original sources for further information.

As a result of the intense scientific activity which was sparked by the discovery of uranium fission in December 1938, by the following June it was known that:

- (1) On the average, between two and three neutrons are emitted per fission
- (2) Practically all the fission is due to the interaction of slow neutrons with the uranium isotope U-235, which is present in natural uranium at an abundance relative to the isotope U-238 of roughly 1:139

- (3) Capture of neutrons by natural uranium takes place primarily via resonance absorption in the U-238 nucleus at energies above thermal to form U-239.

This immediately implied that:

- (1) A chain reaction was possible, in principle.
- (2) Since U-235 fissions primarily via interaction with slow neutrons, conditions for a chain reaction would be facilitated by mixing natural uranium with a moderator to slow the neutrons emitted during the fission process from energies on the order of 1 Mev to energies in the thermal range.

- (3) U-238, which forms the bulk of natural uranium, is a "nuisance" from the viewpoint of achieving a chain reaction

in that the resonance absorption process removes some of the neutrons which would ultimately fission in U-235 were they not captured in U-238 while being slowed down by the moderator.

(At this time it was not yet appreciated that the capture process leads to the formation of a new element, plutonium (Pu-239), which itself could undergo slow neutron fission.)⁴²

The obvious moderator to try was light water (H_2O). In parallel but independent experiments during the next several months Anderson, Fermi, and Szilard at Columbia³³ and Halban, Joliot, Kowarski, and Perrin³⁴ in Paris investigated the possibilities of a chain reaction in a uranium-water system. The nub of the problem was to determine if there existed a water-to-uranium ratio which minimized the combined effect of the two principal neutron loss mechanisms--absorption of thermal neutrons by the water, which effect increases as this ratio is increased, and resonance absorption of neutrons before they are slowed down to thermal energies, which increases as the ratio is decreased--to the extent that the neutron gain via fission exceed the loss and made a chain reaction possible. Unfortunately, the work of both groups indicated that light water absorbed too many neutrons to support a chain reaction, and their attention turned to heavy water and carbon. However, by this time (September 1939), war had broken out in Europe, and scientists in France, Germany, and Britain subsequently withheld publication on research relating to fission in general, and the efficacy of moderators other light water, in

particular. In France, Joliot and his colleagues made detailed reference to the favorable properties of heavy water for the first time in a sealed note deposited with the Paris Academy of Sciences on October 30, 1939.³⁵ They observed that the fact that D_2O was not as effective as H_2O in slowing down neutrons was not crucial, since its low absorption cross section meant that a high D_2O/U ratio could be used, and this in turn would decrease the resonance absorption in U-238. Their optimism about attaining a chain reaction in a heavy water-uranium system--especially for a heterogeneous arrangement of spheres of moderator in a mass of uranium which would decrease the resonance absorption effect as compared with a homogeneous mixture--was tempered by the realization that obtaining enough heavy water would be very expensive. This same thought was expressed by Fermi in correspondence with Szilard in July 1939. In reply to Szilard's suggestion that:

If carbon should fail, our next best guess might be heavy water, and I have therefore taken steps to find out if it is physically possible to obtain a few tons of heavy water. Heavy hydrogen is supposed to have a capture cross section below 0.03, and the scattering cross section ought to be 3 or 4 times 10^{-24} for neutrons above the 1 volt region. Since heavy hydrogen slows down about as efficiently per collision as ordinary hydrogen, and since hydrogen has a capture cross section of 0.27 and a scattering cross section of 20, heavy hydrogen is more favorable.³⁶

Fermi was cautious:

I have discarded heavy water as too expensive; but if you can easily get several tons of it, it might work very nicely.³⁷

This ambivalence concerning D_2O was based on the knowledge that even if a chain reaction were theoretically possible in a system which utilized natural uranium, the amount of moderator required might be very large; e.g., on the order of hundreds of tons. While obtaining such quantities was conceivable in the case of graphite, tons of heavy water seemed out of the question. Fermi and Szilard therefore concentrated their efforts on exploring the feasibility of the uranium-graphite system; an important milestone in this effort was a measurement of the neutron absorption of graphite by Fermi and Anderson in the spring of 1940,³⁸ using very pure graphite procured from the National Carbon Company through Szilard's efforts. The cross section proved to be very small, $3 \times 10^{-27} \text{ cm}^2$, and this reinforced the optimism at Columbia concerning the prospects for a chain reaction in the uranium-graphite system. Although reluctant at first to do so, Fermi eventually agreed with Szilard's request that this result be kept secret.³⁹ As a result, German scientists working on atomic research were not led to question an incorrect measurement of the diffusion length of slow neutrons in graphite by Bothe and Jensen in January 1941⁴⁰ which indicated that neutron absorption was too great for graphite to be used as a moderator. Thereafter German atomic pile research for the remainder of the war focused on the use of heavy water as a moderator, but the

effort was continually frustrated by their failure--thanks largely to Allied efforts --to obtain the needed quantities.

The problem of getting sufficient D_2O was also preoccupying Joliot and his colleagues in France.⁴¹ Tons were not to be had anywhere, but as previously indicated, practically the entire world's supply, approximately 185 kg, was relatively close at hand in Norway, where it comprised the D_2O inventory of the Norsk-Hydro Co. This quantity would certainly be insufficient for a self-sustaining system, but it might be enough to measure the magnitude of the infinite multiplication factor for such a system, and thus indicates just how much D_2O would needed to attain criticality. In November 1939, Joliot proposed to M. Raoul Dautry, the French Minister of Munitions, that the entire stock, worth approximately \$120,000, be purchased or borrowed. An air of urgency was added to the request when the French subsequently learned that in January 1940, a representative of the German firm I.G. Farben had also tried to persuade Norsk Hydro to sell it all its heavy water stock, and moreover, to boost production then running at 10 kg per month, to satisfy a German need for 100 kg per month. (Both I.G. Farben and the Bank of Paris and the Low Countries owned stock in the Norwegian Company; indeed, the French Bank had a majority interest.) The task of obtaining Norsk Hydro's inventory of heavy water for Joliot was entrusted to M. Jacques Allier of the French Secret Service, who was also an officer of the Bank of Paris. Travelling in great secrecy to avoid

detection by German intelligence, Allier went to Oslo in March 1940 where he succeeded in convincing Axel Aubert, the managing director of Norsk Hydro, to loan France its entire 185 kg inventory of D_2O for the duration of the war. Under cloak-and-dagger circumstances, twenty-six cans of heavy water were subsequently flown out of Norway to Scotland and then on to Paris, where Joliot and his colleagues set up an experiment to determine whether a divergent reaction is possible in a homogeneous mixture of uranium oxide and heavy water. A rough estimate of the neutron absorption cross section of heavy water was made, but in the face of the German invasion of France in May 1940, the experiment was dismantled, and the heavy water began its exit from France to England in the same dramatic fashion as it has previously entered from Norway. The first leg of the journey was from Paris to Clermont-Ferrand in central France, where it was hoped that the experimental work could be continued. However, as the military situation worsened, it was decided to move the heavy water to Bordeaux where on June 18 it left for England on a small coaler, accompanied by Halban and Kowarski but not Joliot. The French team was installed by the Maud Committee⁴² in the Cavendish Laboratory at Cambridge where they continued their work on the homogeneous uranium oxide-heavy water system begun in France. Their experimental arrangement consisted of a mixture of variable amounts of U_3O_8 powder suspended in 112 liters of D_2O inside a 60 cm diameter aluminum sphere spinning at 20 rpm

to prevent the U_3O_8 from settling at the bottom, and surrounded by a liquid hydrocarbon. A radium-beryllium neutron source was placed at the center of the system, and measurements of neutron intensity were taken at various distances from the center. The volume integral of the activity of the neutron detectors was measured in the hydrocarbon surrounding the sphere, with or without the uranium-heavy water mixture inside. In December 1940, Halban and Kowarski found an increase in the activity when the heavy water-uranium mixture was present. This implied that the infinite multiplication factor for such a system, k_∞ , was greater than one; and hence that a divergent chain reaction was possible. However, the experimental uncertainties in the calculated value for k_∞ , 1.13 ± 0.07 , left open the question of how much heavy water would be needed for a reactor of finite size. If Halban and Kowarski were correct, "only" a few tons might be enough.

In any case, the Maud Committee scientists were sufficiently encouraged to communicate the result in January 1941, to Harold Urey, a member of the Briggs Uranium Committee, and the person in the US most likely to lend a sympathetic ear to their request for an American effort to find ways of producing large quantities of heavy water. In this manner, the baton was passed to the U.S. as far as heavy water work was concerned, and remained there until the end of the war. We discuss the later developments after brief mention of the German wartime heavy water work.⁴³

Although the heavy water inventory of Norsk-Hydro had eluded them, the Germans were in a good position to capitalize on the fact that the Vemork plant had fallen into their hands with the occupation of Norway in May 1940. By this time, the efficacy of heavy water as a moderator--particularly in a heterogeneous arrangement with natural uranium--was well appreciated; the idea having occurred to several German scientists; Paul Harteck and Werner Heisenberg in particular, during the fall of 1939.

Harteck, one of the key figures in the German atomic bomb effort, had gained first-hand knowledge of heavy water production via electrolysis while working under Lord Rutherford at the Cavendish Laboratory in England in 1934. Moreover, he and his colleague Hans Suess had also investigated the hydrogen-steam isotopic exchange reaction as a means of concentrating deuterium several years before. In the development of this and other processes, they could count on the heavy water expertise of the physicist Karl Wirtz and the physical chemist Karl-Fredrich Bonhoeffer, as well as the highly sophisticated German chemical engineering industry. The German's first action was to request Norsk Hydro to increase the heavy water output at Vermork to 1.5 tons a year. However, when Bothe's erroneous measurement seemingly closed the door on the use of graphite as a moderator, increased production of heavy water became a pressing concern. Various new methods were considered: fractional distillation of water, of hydrogen, and increasing the Norwegian production by modifications

at Vemork; these included expanding the high concentration plant and using the hydrogen-steam isotope exchange process to strip a major fraction of the deuterium from the evolved hydrogen gas and cause it to be returned to the electrolytic cell. Water distillation was rejected as too expensive, but it was decided to pilot the hydrogen distillation process, and press ahead with the Vemork plant modifications. On the basis of an exponential experiment in May 1942, similar in concept to that of Halban and Kowarski but involving a heterogeneous instead of a homogeneous arrangement of uranium metal powder and heavy water, Professors Döpel and Heisenberg estimated that about five tons of heavy water would be needed for a critical reactor. By June 1942, only 800 kilograms had been delivered, but in September a set of exchange reactors containing a nickel catalyst developed by Harteck and Suess were installed at stage six of the Vemork plant. Similar modifications were planned for the fourth and fifth stages; with these improvements the Germans expected production to reach 400 kilograms a month. Unfortunately--for the German atomic effort--the Allies had been aware of the German interest in Vemork heavy water ever since their attempt to buy its inventory in January 1940. The ensuing developments were followed by British intelligence with increasing concern, and in July 1942, the War Cabinet in London authorized Combined Operations to lay plans for a ground attack to destroy the heavy water plant. The ensuing operations

involving great daring, skill and fortitude are the stuff of which popular histories and Hollywood movies are made.

Unhappily, the initial effort in November 1942 was aborted when two gliders carrying thirty-four commandos crash-landed in southern Norway. Though German intelligence was thus alerted to the mission's objective--the survivors having been interrogated before being executed--and measures taken to harden the defenses around Vemork, a second operation mounted in February 1943 by a small Norwegian sabotage team which had been trained in England succeeded in destroying all the electrolytic cells of the high concentration plant. The equivalent of 350 kilograms of heavy water was lost, and partial production could not be resumed until June. The Germans recognized the vulnerability of operations in Norway to further destruction, and intensified their efforts to produce heavy water in Germany using various methods. Their fears proved justified; in a bombing attack by the American Eighth Air Force stationed in Britain in November 1943, the hydrogen electrolysis plant and associated power station were hit, and without electric power the undamaged high concentration cells had to be shut down. Reading the handwriting on the wall, it was decided to dismantle this equipment, and ship it along with the remaining heavy water to Germany. The importance of the latter, in particular, caused the Germans to take special precautions; but the Allies were equally resolute, and the coup de gr^âce was delivered in February 1944 when

a ferry carrying the heavy water across Lake Tinnsjo in Norway was sunk by plastic explosives attached to her bows. With the severing of the Norwegian connection, German scientists were left with approximately two-and-a-half tons of heavy water for use in their ongoing efforts to construct a critical reactor, and ambitious plans to develop new sources using four alternative processes: low pressure water distillation, liquefaction and distillation of hydrogen, catalytic hydrogen-steam exchange, and a new technique which had been developed by K. Geib involving isotopic exchange between hydrogen sulphide and water, a reaction which did not require catalysis. The heavy water effort was spearheaded by Harteck, but it came to nothing because of the rapidly deteriorating war situation-and a lack of priority. According to Irving:⁴⁴

The lack of priorities behind heavy-water plant construction stemmed from a lack of resolution; and the Germans' lack of resolution stemmed from an unspoken belief that before any heavy-water plant could be build, their isotope-separation experts might have found a way to enrich the rare isotope uranium-235.

Harteck himself had ingenuously advised the authorities that

In all probability quantities of enriched 38-preparation [uranium] can be produced, which will effect a significant reduction in the SH-200 [heavy water] requirement. Whether enough enriched 38-preparation can be produced to dispense with SH-200 altogether remains to be seen.

In the US the attitude towards heavy water production was similar, although the rationale was based on the favorable outlook for graphite as a moderator in a reactor to produce plutonium rather than the use of enriched uranium as fuel.

As in Germany a variety of production processes were investigated, but the end result was different; enough heavy water was available by mid-1944 for a ~~small~~ heterogeneous reactor to reach criticality, something the Germans never achieved. As previously noted, the dormant American interest in heavy water was reawakened in January 1941 by the news of Halban and Kowarski's encouraging results.⁴⁵ Studies of large scale production methods were initiated by Urey at Columbia in collaboration with Prof. H. Taylor of Princeton, a physical chemist and one of the pioneers in American heavy water research. Taylor had done experiments on ~~the~~ electrolytic production of heavy water and the exchange reaction between deuterium and water vapor on surfaces as ~~early~~ as 1934, and so was well placed to investigate both the ~~details~~ of the latter process, and how it could be used to make heavy water efficiently by stripping the deuterium from large streams of electrolytic hydrogen and feeding it back into the electrolytic cells, the same idea that Harteck and ~~Suess~~ were planning to implement at Vemork. During 1941, Taylor and his research team studied Urey's suggestion that ~~the~~ hydrogen-water isotopic exchange process be implemented by countercurrent flow of the two substances in a tower packed with a suitable catalyst; in order to make the electrolytic-hydrogen connection, in late 1941 he went to Trail, British Columbia and convinced the management of the ammonia synthesis plant there--the largest source of electrolytic hydrogen in North America--that it

would be practical to incorporate the necessary heavy-water production equipment without disturbing plant operations. Studies of the exchange reaction at Princeton and by the Standard Oil Development Co. demonstrated that, even with the best catalysts, the overall reaction rate was slow, and hence probably governed by the diffusion of hydrogen through water to and from the catalytic surface, a very slow process compared with the actual catalysis. In support of this hypothesis, it was also shown that in contact with various catalysts, the exchange reaction between hydrogen and water vapor at atmospheric pressures occurred at much higher rates than that obtainable using liquid water. This work eventually led to the use of hydrogen-water vapor exchange at the Trail plant in an ingenious system invented by Frank Barr of the Standard Oil Co. A single stage of exchange was carried out in two steps, first equilibrating a water vapor-hydrogen mixture with water, and then passing the mixture over a catalyst where the exchange reaction between water vapor and hydrogen took place. This mixture was again equilibrated with water, and the process repeated. The water flowed countercurrent to the hydrogen-water vapor stream, so that all the effects of a countercurrent system were realized. The decision to proceed with heavy water production at Trail was made in May 1942, and construction started in September 1942. The primary exchange plant was designed to enrich the natural feed water at Trail which contained 0.0138% D to a

concentration of 2.14% D. Finishing was accomplished in a secondary electrolytic plant whose product was 99.8% D₂O. Heavy water was first produced at Trail in June 1943, but because of the long equilibrium time of this process (approximately eight months) and losses from the primary electrolytic cells, production near the design rate of 0.55 tons per month was slow in coming: only 0.25 tons was produced by the end of 1943, and 85% of design by September 1944. However, in December 1942, even before the first heavy water was produced, the leaders of the Manhattan District decided that another source should be developed and--acting on the recommendation of the DuPont Company--choose vacuum distillation of water as the production technique. This, on the basis that it afforded the most reliable technology, which could be implemented without the need for pilot plant operations. In addition, although considerably more expensive than the Trail process, this technique provides a potentially unlimited supply of product, since the source of deuterium is ordinary water, not electrolytic hydrogen as at Trail. In January 1943, DuPont started construction of three distillation plants at the site of ordnance works in Virginia, Alabama, and Indiana, where excess steam capacity for the process was available. The plants were completed on schedule by November 1943, but the average production during their two years of operation was only about 50% of design or 1.2 tons/month, primarily because of high plate leakage.

This level of activity did not satisfy all the proponents of heavy water, particularly Urey, who were dubious about the workability of the massive graphite-moderated piles at Hanford and the gaseous-diffusion method for enriching uranium. For example, as late as February 1944, Szilard was complaining that

The scientists in Urey's laboratory were of the opinion that the [water distillation] process chosen by DuPont was much more costly and inefficient from the point of view of coal consumption than another method . . . It was impossible, however, to collaborate with any firm other than duPont in the development of the alternative method. As a result of this our heavy water production does not exceed three tons a month and consumes 30,000 tons of coal per ton. The quantity of heavy water produced is not sufficient as a basis of adequate production of U²³³ or as a second line of defense for the production of plutonium. The production is expensive and we have no alternative method ready on which to fall back if a larger quantity of heavy water is needed or if economic conditions should compel us to discontinue the inefficient process used by duPont.⁴⁶

The other method alluded to by Szilard was the hydrogen sulphide-water (H₂S-H₂O) dual-temperature isotopic exchange process which was developed by a member of Urey's research group, Jerome Spevack, and dubbed the "S-Process" by the Manhattan District. The principal advantages of this process are: (1) like water distillation, it does not require catalysis, and (2) it provides a potentially unlimited supply of heavy water, but at a much cheaper price than the latter method. However, Szilard's advocacy is a bit of hindsight, since at the time the choice of a backup process to Trail was made in November

1942, only limited laboratory data on the "S-Process" was available,⁴⁷ and there were serious doubts about the feasibility of a large plant, due to the toxic and corrosive nature of wet H₂S. Similar question marks; e.g., the limited experience with the behavior of large-scale apparatus operating at 20 to 25°K, surrounded another potentially promising production technique, distillation of liquid hydrogen. Both processes, and especially H₂S-H₂O exchange, loom larger in post-war heavy water developments, and will be discussed in the next section. Szilard's remarks are symptomatic of the dissatisfaction of the physics group at the Chicago Metallurgical Laboratory with the direction of the Manhattan District, in general, and the major responsibilities given to DuPont, in particular.

As far as the wartime role of heavy water was concerned, the heart of the matter was the reliability of Halban and Kowarski's 1940 experimental result, and here the matter was effectively settled at a meeting between Urey and Fermi in March 1943,⁴⁸ three months after the first graphite pile had gone critical, and with construction of the Hanford reactors underway. The lingering attraction of heavy water was the expectation that, if the French researchers were right, only about ten tons would be required to attain criticality in a conceptually simple homogeneous device consisting of a slurry of uranium oxide in heavy water in which the latter could be circulated to serve as both neutron moderator and coolant. However,

Fermi's calculations for the homogeneous system--using Halban and Kowarski's value for the heavy water slow neutron absorption cross section of $8 \times 10^{-27} \text{ cm}^2$, and a rough estimate of the resonance absorption in a homogeneous mixture--gave a maximum value of 1.02 for k_{∞} , the infinite multiplication factor. If this value was close to the mark,⁴⁹ a critical system would require on the order of hundreds of tons of heavy water. Although the calculations for a heterogeneous system was much more promising; i.e., a k_{∞} of 1.20, the basic conclusion was that the serious disagreement between Halban and Kowarski's experimental result for the homogeneous system and Fermi's theoretical estimate called for a repetition of their experiments using improved techniques, not a crash program to make more heavy water. In June, Fermi irradiated 15 kilograms of heavy water from Trail in the CP-2 pile, and confirmed that the neutron absorption was very low. This result gave added impetus to the growing interest in heavy water among the scientists in the Chicago Metallurgical Laboratory, and in August 1943, a decision was made to utilize their talents in the design and construction of an experimental low power, heterogeneous, heavy-water pile. The pile, CP-3, went critical in May 1944 with 6.5 tons of heavy water, largely from the DuPont water distillation plants, and was the focus of the laboratory's research activities until the end of the war.

In Montreal, where an Anglo-Canadian team had been set up in 1942 under Halban's direction to pursue research on

heavy-water moderated piles, the road to a heavy water reactor was even longer. Of the many obstacles, the primary one was a strong feeling among the leaders of the American project, Bush, Conant, and Groves, that a free interchange of information and materials among US and foreign scientists would give too much away without appreciably aiding US interests.⁵⁰ It was only in April 1944, after the Quebec agreement formalizing the terms of Allied cooperation had been signed by Churchill and Roosevelt, and the English physicist John Cockcroft had replaced Halban--mistrusted by the Americans--as Director of the Montreal Laboratory, that an understanding was reached which provided for American aid--including loans of heavy water and uranium rods--in the design and construction of a heavy water pile in Canada. Subsequently it was decided to build a small Zero Energy Experimental Pile (ZEEP) before the larger National Research Experimental (NRX) Reactor. Like CP-3, both ZEEP and NRX were irrelevant to the war effort; in fact, NRX did not go critical until July 1947 and ZEEP first began operating in September 1945, a month after the war ended.

While it is interesting to speculate on what might have been if, for example, the Norsk Hydro heavy water had been available to Fermi in the spring of 1940, the work of scientists in many countries: France, the US, Canada, Germany, and the Soviet Union had demonstrated that heavy water was an effective moderator, and much insight had been gained into the physics and engineering of heavy-water

moderated reactors. Moreover, many potential methods for economic production of heavy water had been examined, and at least two promising candidates had been identified, hydrogen distillation and dual-temperature hydrogen sulphide-water isotopic exchange.

1.3 The Post War Period

The efforts to develop efficient methods for the large scale production of heavy water after the war were driven by its utility as a moderator both in reactors for the production of nuclear weapons materials and in power reactors for electricity generation. The US and Canada have been the two main actors. In the US, although the later application has had its advocates, up to the present time only the former has been implemented. In Canada, just the reverse has occurred; i.e., the wartime work in Montreal led to the evolutionary development of a commercial nuclear fuel cycle based on natural-uranium fueled, heavy-water moderated and cooled power reactors. We trace here these two strands of the post-war heavy water story, beginning with the work in the US.

In 1945, there seemed to be no pressing need for more heavy water. The Allies were victorious, with the US having sole possession of the bomb, and the cold war had not yet begun in earnest. In these circumstances, the plutonium production capability of the Hanford piles seemed adequate, and possible commercial applications requiring heavy water were distant. Accordingly, the DuPont water distillation plants were shut down in October 1945, having produced about 23 tons of heavy water until that time. (The Canadian plant at Trail produced about 9 tons up to October 1945, and was shut down in 1956 because the hasty wartime marriage between the existing electrolyte hydrogen facility and the heavy water add-on could not

produce heavy water efficiently.)

The situation changed dramatically in August 1949 with the explosion of the first Soviet atomic bomb. Interest in the possibility of thermonuclear weapons revived, and Edward Teller's optimism regarding the chances for success in achieving this goal if enough tritium was available, prompted Luis Alvarez and Ernest Lawrence to press the Atomic Energy Commission (AEC), for the construction of a heavy water reactor.⁵¹ The controversy surrounding the wisdom of a crash program to develop the "superbomb" was heated,⁵² but both sides were agreed that construction of heavy water reactors was desirable.⁵³ Not only could such reactors produce tritium via capture of neutrons in the deuterium nuclei of the heavy water moderator or, more practically, by neutron irradiation of the lithium isotope ${}^6\text{Li}$, but the copious supply of free neutrons above that required to maintain criticality also made possible more efficient plutonium production than in natural uranium graphite-moderated reactors which are only barely critical. This flexibility would be important if thermonuclear explosives should turn out not to be possible, and was a critical element in the decision of the AEC in May 1950 to pursue the heavy water reactor route to increased production of weapons materials. The work on reactor design under Walter Zinn at Argonne, where CP-3 had been constructed, and the Canadian experience with the NRX reactor at Chalk River gave confidence that efficient heavy water production reactors could be built;

the major concern was the availability of the required quantities of heavy water. In anticipation of this need, the AEC had already indicated its interest in two potential production Processes, hydrogen distillation and hydrogen sulphide-water dual temperature exchange, both of which had been investigated in both the US and Germany during the war. The Girdler Co. was asked to design and operate a pilot plant and to design a 40 ton/year production plant based on the hydrogen sulphide-water process, utilizing improvements on the wartime technology which were suggested by the US developer of the process, J. Spevack.⁴⁷ Hydrocarbon Research, Inc. was asked to make design and feasibility studies for a plant based on the hydrogen-distillation process using the synthesis gas of a large synthetic-ammonia plant as feed. The DuPont Co. entered this picture in June 1950, since its mandate as prime contractor for the project to expand weapons materials production via heavy water reactors at Savannah River, South Carolina included responsibility for selecting and providing the necessary heavy water production facilities.⁵⁴ DuPont considered three processes: hydrogen-sulfide-water (or GS for Girdler-Spevack as it became known), hydrogen distillation, and water distillation. The ultimate choice of the GS process for primary deuterium concentration was determined by the following considerations. Water distillation was too capital and energy-intensive, and the technology maturity and adequacy of feed for hydrogen distillation on a large scale were open to question. On the other hand, pilot plant experience

with the GS process indicated that the problems of corrosion, toxicity and process control inherent in dual temperature operation with wet hydrogen sulphide as the working fluid could be overcome. Subsequently, two GS plants were built; one at Dana, Indiana, the site of the Girdler pilot plant and one of the DuPont wartime water distillation plants, and the other at Savannah River. The plants differed somewhat in design and construction details, but both used the GS process for enrichment from natural concentration to approximately 15% D, with finishing to essentially pure heavy water via water distillation followed by electrolysis. The Dana Plant began operation early in 1952 and the Savannah River plant about six months later. Although each plant was designed for a capacity of 240 tons/year, each plant actually achieved a production rate of almost 500 tons/year. By 1957, the peak demand for heavy water for production reactors had passed, and the older Dana plant was shut down and later dismantled. Subsequently, two-thirds of the GS units at Savannah River were also shut down, as was the electrolytic finishing unit. Now in its 27th year, the remaining third still operates with finishing via distillation, to produce about 70 tons of heavy water per year.

The declassification of large areas of US nuclear research in 1955 in support of the Atoms for Peace Program led to the release of detailed information on the US wartime heavy water production efforts --with the exception of the work on the "S-Process"--in the book edited by Murphy et al.⁴⁵

and in a comprehensive survey paper by M. Benedict at the International Conference on the Peaceful Uses of Atomic Energy held in Geneva in August 1955.⁵⁵ In the latter, discussion of the implementation of the GS process in the US was limited to the fact that the heavy water plants of the ACE used this process for primary concentration of deuterium, and that heavy water product was available at \$28 per pound. These omissions, of course, were not accidental. Both Spevack's original patent application in 1943, which was assigned to the U.S. government since he had done the original work on the "S-Process" while working for Manhattan Project, and his subsequent application filed in 1950, which covered the improvements in the process that were subsequently incorporated into the Dana and Savannah River Plants, were held under Secrecy Order by the AEC until February 1957.⁴⁷ At that time, the AEC declassified details of the design and operation of its plants, but Spevack obtained a court order which delayed publication until his patent filed in 1950 was issued in July 1959. Thereafter, the GS genie emerged from the bottle with the publication of a comprehensive technical manual on heavy water production at the AEC plants⁵⁶ as well as more detailed studies of key aspects of the technology.

These developments were followed with much interest -- not only in the US where Spevack's battle with the AEC became something of a cause célèbre -- but in foreign countries which were considering heavy-water moderated reactors for power production. By 1959, Canada, in particular, was well

advanced towards this goal.⁵⁷ Under the inspired leadership of W. Bennett Lewis, who replaced Cockcroft as head of the Canadian nuclear program in September 1946, reactor research and development had progressed rapidly. Following ZEEP, the zero power facility built at Chalk River under Kowarski's leadership which went critical in September 1945, came the 30MW heavy-water-moderated, light-water-cooled high flux research reactor, NRX, which started up at Chalk River in July 1947. NRX represented a quantum jump in technological development, and its successful operation encouraged Lewis in 1951 to advance the feasibility of a nuclear-power effort based on natural-uranium, heavy-water-moderated reactors. To further this end, a Crown Corporation, Atomic Energy of Canada, Limited (AECL) was commissioned in 1952. The next rung up the technological ladder was the NRU reactor, commissioned in November 1957. Its innovations included the possibility of fueling while the reactor was operating, and the use of heavy water as coolant as well as moderator in the interest of minimizing neutron absorption in a reactivity-limited natural uranium system. However, the crucial step in the development of a commercially viable reactor was the design of the Nuclear Power Demonstration Reactor (NPD). The key features of the concept which finally evolved in 1956 were:

- (1) the use of a pressure tube configuration in which the high-pressure, high temperature heavy-water coolant is physically separate from the low-pressure, low-temperature, heavy-water moderator, and whose horizontal orientation facilitates

(2) bi-directional, on-power fueling utilizing short bundles of zircalloy-clad, uranium dioxide fuel for maximum burnup.

By the end of the decade, plans had been approved for the construction of both the NPD and the first CANDU (Canadian Deuterium Uranium) power plant, a 200 MWe unit at Douglas Point, Ontario. If Douglas Point was to be the forerunner of an increasing commitment to nuclear power in Canada, large amounts of heavy water would be needed, roughly one metric ton per megawatt of installed capacity. In these circumstances, it seemed prudent to AECL to implement the GS technology in Canada, rather than to rely on purchases of this strategic nuclear material from the US. It did not have far to look for a strong advocate of this plan.

A delegate to the second Canadian Nuclear Association Conference at Ottawa's Chateau Laurier in May, 1962, might have noticed a dark, powerfully built man moving purposefully from one group to the next during receptions and coffee breaks. He was often accompanied by a charming wife and an attractive daughter. His topic of conversation was always the same. He spoke with enthusiasm, with suave persistence, with absolute confidence...and always about ⁵⁸ heavy water. The man was Jerome S. Spevack.

Actually, AECL already had access to the wartime "S-Process" patent pursuant to the patent exchange provisions (Article IX) of the agreement for cooperation concerning civil uses of atomic energy which Canada and the US signed in 1955. However, no plant had ever been built to this patent alone. As noted, the Savannah River and Dana GS plants were based on Spevack's improvement patent which was issued in Canada in 1961.

This strong patent position was a major factor in dissuading

major petrochemical firms such as DuPont of Canada and Canadian General Electric from submitting formal proposals, when in February 1963 the AECL called for competitive bids for a 200 ton per year commercial heavy water plant. A year later, the contract was awarded to Deuterium of Canada, Ltd. (DCL), the wholly owned subsidiary of Spevack's US Deuterium Corporation. The plant was to be built in Glace Bay on Cape Breton, Nova Scotia, with AECL agreement to purchase its product under condition that majority ownership and control of DCL be vested in the Government of that Province.⁴⁷ Thus began Canada's attempt to create a large scale heavy water industry; unfortunately this chapter of the CANDU story did not have an auspicious beginning. Construction was delayed as a result of labor, construction and vendor supply problems, and when startup operations at the initial 200 tons per year plant began in November 1968, it was discovered that the stainless steel tubes in the seawater-cooled heat exchangers were leaking from corrosion. The original plant never operated, and was subsequently rebuilt by Canatom Mon-Max. Opinions differ as to the reasons for this fiasco. The conventional wisdom in Canadian and U.S. heavy water circles, is that the basic problem was the unworkability of some of the modifications of the GS technology as practiced at Savannah River and Dana which Spevack incorporated into the Glace Bay design. In particular, the use of sea water as feed and coolant and the use of the GS process for finishing to reactor grade product as well as

for primary concentration from natural water feed. In reply,⁵⁹ Spevack has argued that the corrosion occurred because the sea water coolant had been left in a stagnant condition in the involved heat exchanges for considerable periods of time prior to startup; a circumstance of which he had no knowledge or control. Moreover, there was no reason why the use of the GS process for the final stages of deuterium concentration, while unconventional, should not work in practice, and in any event the latter stages were of minor significance in terms of plant cost. Unfortunately, detailed analysis of the relative merits of these arguments and related matters would convey us too far afield. In any case, as a result of "the muddle at Glace Bay" as well as startup problems at a 400 ton per year GS plant built by Canadian General Electric (CGE) at Port Hawkesbury, Nova Scotia, in the early 1970s AECL was forced to seek worldwide for sufficient heavy water for the four 540 MWe reactors at the Pickering station which were being commissioned at that time. The drought finally ended with improved operation at Port Hawkesbury and the start-up of an 800 ton per year GS plant, Bruce A, at the Bruce Nuclear Power Development near Douglas Point. Improvements in the GS process suggested by Proctor and Thayer in 1962⁶⁰ were incorporated at both Port Hawkesbury and Bruce. The prime contractor at both plants, the Lummus Company, had also functioned as subcontractor at Savannah River. Presently, the Bruce A, Port Hawkesbury, and the rehabilitated Glace Bay plant are all operating, with a cumulative output at 100% capacity of about 1600 tons per year, and another 800

tons a year of capacity will soon be available from the Bruce B plant, now being commissioned. With these developments in mind, one is tempted to summarize the story of heavy water production in Canada as "all's well that ends well." However, the uphill battle to reach this goal supports the proposition that efficient operation of transferred high technology is not exclusively a problem for the "underdeveloped." As Dyne, et al., have cautioned with regard to the prospects for regional fuel reprocessing,

Our experience with other nominally developed technologies, heavy water production being an example, is that they cannot be operated efficiently in another country without the host country having detailed technical expertise in that technology.⁶¹

Besides Canada, many other countries have built and operated heavy water-moderated research and demonstration power reactors during the post-war period. Of these, the country with the longest and most steadfast commitment to a commercial fuel cycle based on natural uranium and heavy water has been Canada's Commonwealth partner and nuclear collaborator, India. In pursuit of this goal, and further evolutionary developments including fast breeder reactors utilizing plutonium cores and thorium blankets, a high priority was placed on developing a self-sufficient indigenous capability in science, technology, and industry. Along the way, personnel would be trained and technology acquired abroad as necessary, but always with the aim of advancing along the learning curve as rapidly as possible. The history

of heavy water production in India is a textbook example of how this policy has been pursued.⁶² When in 1954 the Indian Department of Atomic Energy (DAE) began heavy water feasibility studies, information on only two processes was available in the open literature, water distillation and electrolysis. The former was rejected as too expensive, while expert opinion regarding the latter was obtained by hiring as a consultant, Dr. Jomar Brun, chief engineer of the Vemork heavy water plant, which had resumed operations after the war. Brun's study indicated that, even when produced as a by-product of a fertilizer plant based on electrolytic hydrogen, heavy water in India would be expensive, about \$100 per pound.⁶³ However, with the disclosure of information on various production processes at the 1955 Geneva Conference,⁵⁵ it became feasible to consider a wider range of possibilities. The method finally chosen was distillation of pre-enriched electrolytic hydrogen produced for ammonia synthesis. Besides its inherent advantages; e.g., high deuterium recovery from feed and low energy consumption, the choice of this process was favored because of the experience which would be gained by prior operation in France and Germany of two plants using the same technology. In 1959, an agreement was signed with the German company Linde to build a 14 ton per year plant at Nangal, and the plant started up in 1962. However, by this time it was clear that much more heavy water would be needed to meet the needs of India's rapidly developing nuclear program. Given the access to US GS technology

available since 1959 via the technical literature and visits to Savannah River, the logical step for a country wishing to develop this technology indigenously would be to build a pilot plant incorporating the improvements suggested by Proctor and Thayer, while keeping a keen eye on Canadian efforts in this area. In a paper presented at the 1964 Geneva Conference,⁶⁴ it was announced that a GS pilot plant had been constructed by DAE at Trombay, and design considerations were given for a 200 ton per year plant using the GS process for primary enrichment with either hydrogen or water distillation contemplated for final concentration. By 1970 these plans had changed somewhat; only 100 tons per year was to be produced by the GS process; the difference to be made up by other plants where heavy water production would be effected by another isotopic exchange process, that between hydrogen and ammonia. The hydrogen-ammonia exchange reaction had been studied by Urey's research group during the war, but was not considered promising because, like the hydrogen-water system, no catalyst was available which permitted counter-current operations with the ammonia in the liquid phase. The discovery by Claeys, Dayton and Wilmarth in 1950,⁶⁵ that amide ion in liquid ammonia serves as an efficient homogeneous catalyst for the reaction stimulated research in this process by groups at the Brookhaven National Laboratory in the US and the Commissariat a L'Energie Atomique (CEA) in France. The French work led to the operation of a pilot plant at Mazingarbe in 1957 and a decision in 1964 to build a production plant at this site

using this exchange reaction for primary enrichment with final concentration by distillation of liquid ammonia.⁶⁶ The feed was synthesis gas (75% H₂, 25% N₂) from an associated ammonia plant whose 500 ton per day capacity limited heavy water production to about 26 tons per year. The plant was brought into production in December 1967, and its operation encouraged India to order in 1969 and in 1971 two similar units from the French consortium, GBLPRA, one to be built at Baroda and the other at Tuticorin, with annual design capacities of 67 and 71 metric tons, respectively. Another plant based on the ammonia-hydrogen exchange process using a dual-temperature flowsheet rather than the mono-thermal mode favored by the French,⁶⁷ was subsequently ordered from the Uhde Co. of West Germany, where research and development, including pilot plant operations starting in the early 1960s, had demonstrated the feasibility of dual-temperature ammonia-hydrogen exchange for primary deuterium enrichment. The Indian plant employing the Uhde process to be built at Talcher, has a design capacity of 63 metric tons per year.⁶⁸

Of the four Indian heavy water plants in various stages of construction, the only one directly affected by the Indian explosion of a nuclear device in May 1974 and the subsequent break in Indo-Canadian nuclear collaboration was the GS plant at Kota next to the Rajasthan Atomic Power Station. Although considerable know-how concerning the GS-process had been acquired by the DAE, certain key pieces of equipment as well as the design engineering services

were to be imported. With the sudden unavailability of contracted assistance, the design engineering job was given to an Indian firm, and needed equipment sought from other suppliers. The progress of the Kota plant, in particular, is being watched with great interest, since its successful operation would add weight to the proposition that, beyond a certain stage of technological development, a cutoff in outside assistance can at most lead to delays, added costs and troubled relations.

As of the Spring of 1979, however, all of the plants under construction were experiencing difficulties. According to the then Prime Minister Moraji Desai ⁶⁹

The technology of production is new and complex, and the processes adopted were being used for the first time in India, so the original schedules could not be adhered to. This factor and delays in the supply of equipment from indigenous and foreign sources, problems of transport of certain heavy equipment, failure of some of this equipment, interruptions in power supply, and events like strikes, are the main reasons for the delay in the commissioning of the heavy water plants.

According to more recent estimates, ⁷⁰ all four plants should be producing heavy water by the end of 1981, and plans for the construction of the next generations of plants are underway.

In summary, the GS process is today the dominant process for primary enrichment in large-scale (hundreds of tons per year) heavy water production. However, processes which are based on extracting deuterium from ammonia synthesis gas, such as hydrogen-ammonia and hydrogen-amine exchange ⁷¹

could be competitive, especially if the ammonia synthesis and heavy water plants are designed together, and the capacity of the former is large. For example, if the deuterium recovery from a 1500 ton per day synthesis plant is 80%, more than 100 tons per year of heavy water could be produced. In this manner, depending on local conditions -- including the relative availability of the technologies -- several parasitic plants may be more advantageous than a single large GS plant. Alternatively, it is possible to combine the advantages of hydrogen-ammonia gas or hydrogen-amine exchange with the unlimited capacity possible with a water feed by adding either an ammonia-water or hydrogen-water step to these non-aqueous systems. We discuss these so-called transfer processes briefly in Chapter 3, Section 3.2.4.

Of course, the future large-scale use of heavy water⁷² in countries which have already made a commitment to the natural uranium, heavy water fuel cycle; e.g., Canada, India, and Argentina, will depend on the future growth of nuclear energy in these countries, while its use in other countries will depend on both this factor and the relative attractiveness of the light and heavy water fuel cycles.

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Although extensive development work has been done, the amine process has not as yet been reduced to commercial practice. For more details, see Chapter 3 , Section 3.2.1.

72. The consumption of heavy water in controlled thermonuclear fusion reactors is modest; e.g., to supply a 1000MW_e plant utilizing the D-T reaction with a thermal efficiency of 40%, only about 0.4 tons per year of D_2O would be required.

2. Non-Proliferation Considerations

Thermal reactors which use heavy water as a neutron moderator allow a variety of configurations depending on the choice of:

- (1) the fissile material (U-235, U-233, or Pu239)
- (2) the fertile material (U-238 or Th-232)
- (3) a pressure tube or pressure vessel design
- (4) on-power or off-power fuelling
- (5) the coolant
- (6) a direct or an indirect steam cycle.

Reactors embodying various combinations of these options have been built. However, the one of most current interest is the pressure tube, natural-uranium, on-power-fueled, heavy-water-cooled reactor which has been developed and commercialized in Canada, the CANDU. Here, we focus on the proliferation considerations associated with this reactor and its associated fuel cycle, with a brief discussion of some of the other possible reactor-types, in particular the pressure vessel, natural-uranium, heavy-water-cooled reactor, which has been commercialized in Argentina, and the pressure tube, mixed-oxide, light-water-cooled reactor, a prototype of which is operating in Japan.

2.1 The CANDU

Broadly speaking, the question of proliferation and the CANDU can be divided into two parts: (1) how serious is the threat of diversion of irradiated fuel, and (2) what are the implications of the widespread utilization of a fuel cycle which requires isotopic enrichment of the moderator but not the fuel?

With regard to the former, the following issues have been raised in the non proliferation literature:

(1) As compared with a light water reactor (LWR), on a once-through fuel cycle normal operation of a CANDU produces more plutonium in the spent fuel per kilowatt of electricity generated, and CANDU plutonium emits fewer spontaneous fission (SF) neutrons, which are widely cited¹ as the cause of "pre-initiation" and consequent nuclear explosive yield uncertainty and degradation.

(2) On-load fuelling implies enhanced capability for covert diversion of fuel; in particular, for diversion of spent fuel which has had a lower than design burnup in order to achieve better weapons quality plutonium.

With regard to the latter, specific contentions have been that:

(3) Since the sources of natural uranium are widespread, the possessor of CANDUs can proliferate with relative impunity, as compared with a nation that depends on supplies of enriched uranium to operate LWRs.

(4) The CANDU fuel cycle provides a rationale for the acquisition of large-scale heavy water production facilities, and diversion of relatively small quantities of their product would suffice for the construction of efficient, compact heavy-water moderated reactors for the production of weapons materials.

We discuss each of these points in turn:

(1) At typical design burnups: 7,500 MWD/T in a CANDU, 33,000 MWD/T in a pressurized water reactor (PWR) and 27,500

MWD/T in a boiling water reactor (BWR), the total amount of plutonium in the spent fuel of a CANDU is greater than that in either LWR by about a factor of two. However, the quantities in all cases are large compared to that needed for a weapon: (~500 kg in a CANDU vs ~260 kg in either LWR per gigawatt-year at 75% capacity;² in addition, the plutonium concentration in the CANDU spent fuel is lower (0.4% vs 0.8-1% in an LWR),² and this means that more fuel must be reprocessed to obtain the same quantity of plutonium. Moreover, the neutron background due to spontaneous fission of the even plutonium isotopes, Pu-238, Pu-240, and Pu-242, is of the same order of magnitude (Table 1).²

Table 1
Plutonium Composition of Spent Fuel at Discharge (%)

Isotope	7,500 MWD/T CANDU	27,500 MWD/T BWR	33,000 MWD/T PWR
Pu-238	0.1	1.0	1.5
Pu-239	68.4	57.2	55.7
Pu-240	25.6	25.7	24.5
Pu-241	4.6	11.6	13.4
Pu-242	1.4	4.5	4.9
Pu-238+240+242	27.1	31.2	30.9
SF Neutron background (n/s-g)	287	363	371

(2) Specifically, as regards production and diversion of high weapons-quality; i.e., low-burnup plutonium, the conventional wisdom is that:

In contrast to the LWR, production of low Pu-240 in the CANDU does not involve a significant loss of power output.³

To put this question in perspective, we note that a 600MWe CANDU has 380 fuel channels each containing 12 fuel bundles,

and each bundle contains 18.7 kg of uranium. On-load fuelling is accomplished via two remotely controlled fuelling machines, one at each side of the core. It is estimated that 300 fresh bundles will be discharged from and a like number charged to the reactor per month in a 600 MWe reactor. Although the frequency and programming of these bundle changes has not yet been firmly established --none of these reactors is yet in operation-- it is anticipated that the loading pattern will be similar to the frequency and method of operation of the Pickering power station. At Pickering, about 250 fuel bundles per month are removed from and added to each of the 540 MWe reactors; these bundles are changed on the day shift, Monday to Friday. Eight bundles are removed at a time, and the total refuelling operation takes about six hours.⁴

At the design burnup, the total plutonium content of the irradiated fuel is about 3.8 gm Pu/kg U, so each irradiated bundle contains approximately 70 gm Pu. Thus to obtain 8 kg of reactor-grade plutonium -- which is considered to be a quantity of safeguards significance by the IAEA⁵ -- approximately 112 bundles must be diverted. In order to get weapons-grade quality material, i.e., an isotopic mixture containing on the order of 95% Pu-239, the fuel burnup must be equal to or less than 1000 MWD/T, at which the plutonium content is about 0.87 gm Pu/KgU. Thus to obtain 8 kg as before, about 8 kg Pu \setminus $(0.87 \text{ gm Pu/kg U} \times \frac{18.7 \text{ kg U}}{\text{bundle}}) \approx 490$ bundles would be required. To what extent can either of these operations be carried out covertly? The answer, quite obviously, depends on the efficacy of the safeguards system. If there are

no safeguards, or equivalently, a system which is not tamper-proof and does not have built-in redundancy protection in case of normal equipment failure, then either operation is, in principle, straightforward; e.g., without shutting down the reactor, the required number of irradiated bundles would be remotely removed from the reactor area, and shipped off site in casks.

Nonetheless, it has been suggested⁶ that the production of low-burnup material in a CANDU might give a potential proliferator pause even if there were no safeguards because moving fuel bundles more rapidly than normal through some sub-set of the fuel channels:

- (1) might overtax the capacity of the fuelling machines and lead to a loss of reactivity in the rest of the core.

- (2) might produce hot spots in the core with a potential danger of fuel melting due to the higher reactivity of the fuel in the low-burnup channels.

Unfortunately, neither of these problems is insurmountable. In the first place, as we have noted, normal fuelling of a CANDU is accomplished during one six-hour shift, five days a week; this suggests that machine availability for illicit operations would not be a problem. Secondly, there are various ways of avoiding potential hot-spot problems which will be obvious to anyone with some knowledge of nuclear reactor physics and engineering.

An "obvious" and radical solution to the on-load fuelling diversion problem is to eliminate on-load fuelling. While this is feasible in enriched fuel or mixed-oxide designs; e.g., the SGHWR or the FUGEN BLW, on-load fuelling is essential with natural uranium which is highly reactivity limited. Moreover, off-load fuelling does not obviate the need for effective safeguards since "unscheduled" reactor shutdowns to replace defective fuel or some "fuel mismanagement" at the time of scheduled refuellings create opportunities for the diversion of irradiated fuel containing plutonium of high Pu-239 content. A more rational approach to the on-load fuelling vulnerability is to devise safeguards procedures tailored to the problem. This has been done over the past several years by the Safeguards Division of AECL, the Canadian Atomic Energy Control Board (AECB), and the IAEA Department of Safeguards and Inspection. A short description of their work follows; further details can be found in references 7-10.

The basic premise underlying the CANDU safeguards scheme is that the major concern is the plutonium contained in the irradiated fuel, and that an approach which utilizes item counting of irradiated fuel bundles and containment/surveillance measures can insure timely detection of the unreported removal of significant quantities of irradiated fuel:

- (1) from the core by abnormal means, and
- (2) from the spent fuel storage area, with the possible

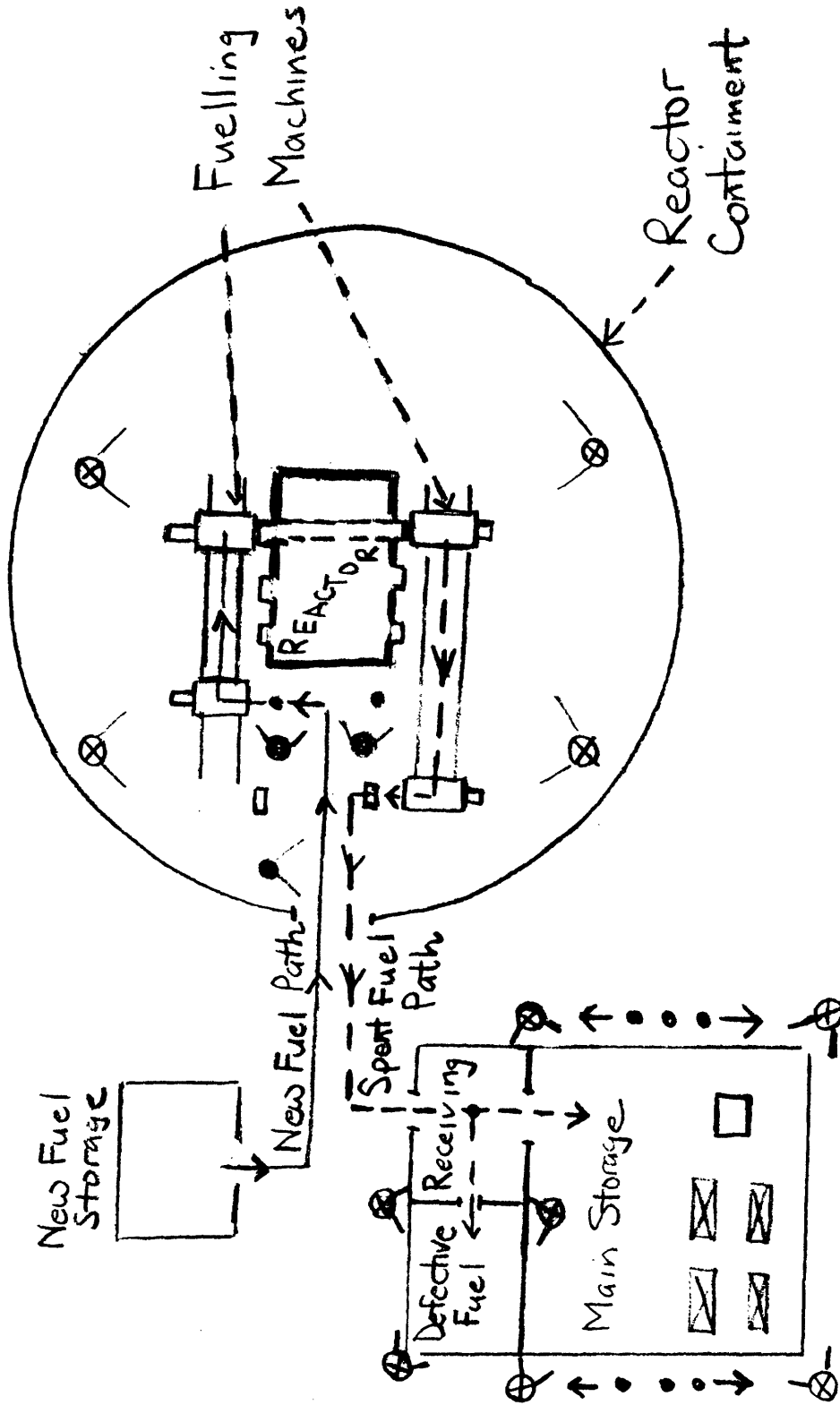
substitution of dummy or unirradiated fuel.

To achieve this end, the safeguards scheme employs a variety of instrumentation: TV surveillance systems, photo-surveillance cameras, radiation yes-no monitors, bundle counters for spent fuel, a "bundle verifier" for spent fuel trays, and security seals and cages in a redundant fashion to minimize the effect of equipment failure. Briefly, the function of these components is as follows (see Fig. 1).

(1) The surveillance cameras and the radiation yes-no monitors in the reactor containment building verify that irradiated bundles have not been removed from the core except via the normal route which carries them past the bundle counters and into the spent fuel storage bays. (There are three bays: a receiving bay where the irradiated bundles are remotely loaded onto the storage trays, and transferred into the main storage bay. Defective fuel is first canned, and then transferred via the receiving bay to the defective fuel bay.) In particular, four TV cameras observe all movements of the fuelling machines and two film cameras and yes-no radiation monitors are positioned to insure that irradiated fuel is not diverted along the route normally taken by fresh fuel being charged to the reactor. An additional film camera is used to monitor various openings in the containment shell such as the main airlock.

(2) The two bundle counters sense and record the number of bundles passing from the reactor to the storage bays as well as their direction of motion and the time of transfer.

(3) Seven TV cameras are positioned to observe the water



- Bundle Counter
- ⊗ TV Camera
- Film Camera
- Yes/No Monitor
- ⊠ Sealed Cage
- Bundle Verifier

Figure 1 CANDU Safeguard System (Schematic)

surface, access doors, and other above-surface openings in the spent fuel bays. Possible movement of irradiated material through the water purification piping system is detected and recorded by yes-no radiation monitors.

(4) In the main storage bay the following equipment is utilized to secure irradiated bundles and to verify that dummies have not been substituted.

(a) An array of gamma-sensitive diodes mounted on a frame on which a loaded tray can be placed in a manner which allows the activity of each bundle on the tray to be verified.

(b) Stainless steel mesh cages, with covers, to enclose a stack of spent fuel trays, and

(c) Ultrasonic seals, verifiable in situ under water, to seal the cage covers.

At each routine IAEA inspection, the system could be used in the following manner to insure that irradiated bundles have not been diverted.

(a) The bundle counters would indicate the number of irradiated bundles which have taken the normal route from the reactor to the spent fuel bay since the last inspection. The inspector would count the number of bundles in the bay which have not been sealed into cages, and confirm that it checks with the bundle counter tallies.

(b) The gamma verifier would be used to verify that the bundles in (a) are not dummies. All trays might be checked individually, or a sampling technique used, depending on the degree of confidence considered necessary. For example, if routine inspections took place every two months, approximately 720 bundles would normally have been discharged from the reactor and loaded onto 30 trays between inspections. Assuming a proliferation scenario which involves diversion of a significant amount of plutonium during the inspection "dead-time," at least 112 bundles would have been taken from the pool and replaced by dummies. Thus, in the worst case, only five out of thirty trays would contain dummies, and it would be prudent to check each individual tray since a random sampling of a considerable fraction of the total would be needed to insure that the bundle substitution scheme was detected with a high level of confidence. For example, if only ten trays are checked, the confidence level is less than 85%.

(c) The inspector would supervise the sealing of the fuel verified in (b) into cages, and the sealing of these cages. The seals on previously secured cages would be checked on a random basis.

(d) Film and videotapes from the surveillance cameras would be reviewed either on-site or at IAEA Headquarters.

(e) The seals on safeguard equipment would be verified and the equipment serviced as required to insure tamper-proof, reliable operation.

Equipment redundancy is provided in the following manner:

(a) A diversion of irradiated bundles via a route which bypasses the bundle counters and then leads off-site through an opening in the containment shell would be detected by the cameras which observe the fuelling machines, the cameras and radiation monitors which watch for the presence of irradiated material along the fresh fuel path, and finally the camera which monitors the openings in the containment.

(b) A diversion of irradiated bundles from the spent fuel bay after they have been counted would show up as a discrepancy between the bundle counter tally and the number in storage, and would also be observed by the cameras which monitor the bay. These cameras, as well as the bundle verifier, would also detect an attempted substitution of dummy bundles.

Since a working demonstration of the entire system at an operating reactor has not yet taken place--one is planned at Gentilly 2 in early 1980 when the reactor is scheduled to start-up--a judgement as to its efficacy would be premature. We confine ourselves to the following preliminary observations:

(a) It is apparent that much thought has gone into the system design, and considerable effort into developing the required equipment; e.g., tamper-indicating, highly reliable bundle counters. Moreover, there is an awareness that further improvements which would reduce the residual vulnerabilities of the present scheme could be implemented. For example,

fresh fuel bundle counters would provide additional confidence that a diversion of undeclared fresh fuel after irradiation would be detected, even in the event of a "common-mode" failure in which all surveillance cameras stopped working and the irradiated fuel bundle counters were bypassed. The question of the degree of safeguard protection is obviously a matter of costs vs. benefits which may be most keenly felt in the case of retrofitting existing reactors.¹¹

(b) The question of whether this safeguards scheme provides timely warning of the diversion of irradiated bundles depends on one's perception of the time required to extract a significant amount of plutonium from spent fuel. The IAEA had estimated this time to be between one and three months.¹² Accordingly, a routine inspection every two months and an annual physical inventory have been suggested as a reasonable procedure.⁸ In the light of some estimates that 10 kg of plutonium might be extracted from spent fuel in a matter of weeks,¹³ a more intensive routine may be prudent. However, as in (1), this is a matter of added costs vs. expected benefits, where resolution requires the accumulation of some operating experience. In this regard, some non-proliferation analysts have suggested that timely detection of the diversion of spent fuel can only be assured via an instrumented near-real-time surveillance system involving monitors at the reactor site which can be remotely interrogated via a secure communication link from IAEA Headquarters. Other observers feel that this is an example of proliferation-resistance engineering

"overkill" which may be appropriate for weapon-grade material, but not spent fuel.¹⁴ Here, without entering this debate, we would only point out that a remote monitoring system which only checks the seals on secured assemblies of light water reactor fuel between periodic refuellings is not sufficient in the case of on-load fuelling where irradiated bundles are constantly accumulating in the fuel bay.

(3) The thesis that dependence on external supplies of enriched uranium fuel for its light water reactors would be a constraining influence for a nation contemplating a move towards nuclear weapons while the use of the CANDU national uranium fuel cycle removes this inhibition presupposes that:¹⁵

(a) the supply of natural uranium and heavy water for normal makeup either in the world market or from indigenous sources is relatively secure.

(b) the enrichment control leverage will be applied uniformly by all suppliers, including perhaps an international fuel bank, and uranium enrichment technology will not spread.

The indigenous resources of uranium ore of various non-nuclear weapons states who have expressed interest in or already made a commitment to the natural uranium fuel cycle are given in Table 2.¹⁶ Although only Argentina has large, reasonably assured resources, the general availability of sufficient indigenous resources to operate a modest nuclear program based on the CANDU fuel cycle (~ 100 mg.U/GWe-yr. at 75% capacity) should be noted.

Table 2

Uranium Resources Recoverable at Costs up to \$130/kg U (megagrams U)

Country	Reasonably Assured Resources	Estimated Additional Resources
Argentina	41,800	---
South Korea	3,000	---
Rumania	Not available	
Mexico	4,700	2,400
Yugoslavia	6,500	20,500
Philippines	---	400
Turkey	4,100	---
Pakistan	Small deposits exist, but magnitude unknown	
Taiwan	No deposits of commercial value found	

The nuclear export policies of three of the five major suppliers of ore outside the Communist Bloc: the United States, Canada, and Australia are quite stringent with regard to satisfaction of non-proliferation and safeguards criteria. Thus, for countries which need to import ore, the mere existence of these resources would provide little cause for comfort to potential nth countries or even nations whose non-proliferation bona fides are rarely questioned. Indeed, Canadian insistence on the right of prior consent to reprocessing of Canadian origin material, control over all material of Canadian origin even if others attached their own safeguards requirements to the same material, and control on retransfers of nuclear technology, led to an embargo on uranium shipments to the European Economic Community (ECC) Japan, and Switzerland in January 1977.¹⁷ The case of South Africa and Niger, the other major non-Communist Bloc uranium suppliers.

is more complex. Because of its non-adherence to the NPT, its pariah status in the international community, and its indigenous nuclear expertise--symbolized by the non-safeguarded uranium enrichment facility at Valindaba --South Africa has been a prime focus of non proliferation concern. Its official policy on uranium exports has been described as follows:¹⁸

The export of uranium is governed by the provisions of the Atomic Energy Act, No. 90 of 1967. In terms of that Act, uranium may be exported only on the authority of the Minister of Mines, acting after consultation with the Atomic Energy Board. When considering applications for permission to export, account is taken of the status of the end-user, the country of destination, applicable safeguards, price and other proposed conditions of sale.

Obviously, it would be naive to expect that South Africa will attach the same non-proliferation criteria to its uranium exports as do the United States, Canada, and Australia, all of whom require; e.g., safeguards on all peaceful nuclear activities as a condition for nuclear commerce of any kind. However, whether it would agree to either continue to export to a country which had clearly violated its non-proliferation undertakings or step in as a supplier of last resort in the same situation is problematic. Given its diplomatic situation, South Africa would probably find it prudent to maintain a low profile in nuclear matters unless its own vital interests were clearly at stake.

The other important African supplier is Niger, with reasonably assured resources of 160,000 megagrams U recoverable at costs up to \$130/kg U.¹⁶ A wide range of companies from foreign countries--most notably France, but also including Japan, West Germany, Canada, Italy, the FRG, and the UK--are carrying out uranium prospecting in the country in accordance with the wish of the Niger Government to have a diversity of partners in developing the mining industry.¹⁹

In sum, supplies of natural uranium ore in quantities large enough to support a modest nuclear program based on the CANDU fuel cycle may be generally available either from indigenous resources or from suppliers who might not insist on satisfaction of the same non-proliferation criteria as do the U.S., Canada, and Australia.

As regards the availability of heavy water, the feedstocks for the production processes which have been implemented commercially, ordinary water, electrolytic hydrogen and ammonia synthesis gas, are more readily available than natural uranium. However, in the event of a total embargo on heavy water which took effect after the amount necessary for reactor inventory had been supplied (approximately 0.8 MT/MWe, or about 500 MT for the standard 600 MWe CANDU export model), a nation would either need to have heavy water in reserve or the technology required to produce it in amounts required for either normal makeup or accidental loss.²⁰ Presently, the countries which can supply large amounts of heavy water; i.e., on the order of a hundred tons per year, are the U.S. and perhaps the U.S.S.R., Canada, while the U.S., Canada, France, Switzerland, and the FRG have

demonstrated a capacity to build large-scale production facilities. All of the above are members of the Nuclear Suppliers Group whose safeguards trigger list includes both heavy water product and production technology. Moreover, current U.S. and Canadian policy goes significantly beyond the guidelines with regard to heavy water, requiring, at the minimum, either adherence to the NPT or acceptance of the principle of full-scope safe-guards for the transfer of heavy water technology. Whether the heavy water genie can be kept in the bottle in the face of: (1) the reluctance of France, Switzerland and the FRG to go beyond the Nuclear Suppliers guidelines as far as the transfer of this technology is concerned, and (2) the ability of countries to master it on their own, is problematical. (These questions are discussed in more detail in Chapter III.)

(2) In the 1980's the major suppliers of separative work will be the US, the USSR, and the Eurodif and Urenco consortia. The US and the USSR, the Urenco partners: the UK, the FRG, and the Netherlands, and France, the major actor in Eurodif, are all adherents to the Nuclear Suppliers guidelines which call for "restraint" in the transfer of uranium enrichment technology, and also raise the specter of supplier sanctions in the event of a violation of supplier/recipient understandings. Regarding the latter, the guidelines do not require the application of sanctions.²¹ This would not be in the spirit of the document which is essentially a non-binding gentleman's agreement rather than a treaty; indeed the word "sanctions" is not mentioned. However, there is an attempt to address the potential problem of non-uniform perceptions of the seriousness of violations of

non-proliferation undertakings. That is, it is agreed that:²²

Pending the early outcome of such consultations [between suppliers on possible violations of supplier/recipient understandings resulting from these guidelines], suppliers will not act in a manner that could prejudice any measure that may be adopted by other suppliers concerning their current contacts with that recipient.

A crucial test of this policy is likely if the U.S. does not continue supplying India with enriched uranium for the light water reactors at Tarapur. In this case, it is conceivable that Eurodif or the USSR might become the supplier of "last resort". There seems to be little disposition on the part of the US government, particularly in the Congress or the Nuclear Regulatory Commission, to exempt India from adherence to full-scope safeguards, mandated by the Nuclear Nonproliferation Act of 1978, as a (retroactive) condition for the continuation of nuclear commerce with the US, while on the Indian side there is great reluctance to accede to US pressure. The dangers inherent in this situation are obvious, since the temptation to allow commercial and/or geo-political considerations relating to nuclear supply to dominate nonproliferation concerns is ever present. The same generic problem is also at the heart of the issue of the spread of enrichment technologies. That is, the conventional wisdom embodied in such maxims as: "technological barriers inevitably erode in time" and "the secrets of nature are an open book" overlook the historical fact that the spread of nuclear technology in general, and expertise relating to heavy water production, reprocessing,

and to a lesser extent, uranium enrichment, in particular, have largely been the result of conscious decisions by various actors that the political/commercial benefits of transfer outweighed the proliferation risks, where this factor has been considered at all.²³ If a serious commitment is made towards restraining the future transfer of enrichment technology, while making credible assurances of supplies of low-enriched uranium--how the US "plays the Tarapur card" may be crucial in this respect--there are grounds for optimism that this technology will not spread rapidly.²⁴ In summary, the question of whether the cause of non proliferation would be better served from the viewpoint of relative leverage on the supply of strategic nuclear materials --by the adoption of the light or heavy water reactor fuel cycles has no clear-cut answer. Moreover, specifically nuclear-related sanctions may not always be the most appropriate response to violations. The overriding issue is the degree of consensus among suppliers and recipients on the importance of adherence to mutually agreed-upon non proliferation norms.

(4) Whether a nation which decides to obtain nuclear weapons via a path involving irradiation of natural uranium to produce plutonium in a so-called "production" reactor will choose to use graphite or heavy-water as a moderator depends on various factors; e.g., relative availability of these materials,²⁵ technical capability, and the proliferation scenario. As far as the historical record is concerned,

a case can be made for either material. That is, the US, USSR, UK and France all initially used graphite-moderated reactors to produce weapons material, while India and perhaps others have taken the heavy water route. For the former group of nations, the greater availability of graphite at the time the initial weapons decision was made, the relative simplicity of the graphite reactor design, and the fact that there was no perceived need to fuz the weapons-nature of the device by calling it a research reactor, outweighed the superior moderating properties of heavy water, which lead to higher neutron fluxes and hence greater neutron production per megawatt of reactor power in a physically more compact device. The implication one may draw from this as far as the possible future course of proliferation via the production reactor route can be summarized as follows. Nations without a high degree of nuclear expertise who wish a relatively cheap and simple route to weapons, and are not overly concerned by the signal that detection of a clandestine graphite-moderated reactor would send to the international community, might find this an attractive method. On the other hand, nations who nurture more sophisticated weapons objectives and/or those who wish to develop a civilian nuclear program in a manner which fosters ambiguity as to weapons intentions would find the heavy water route more to their taste. With regard to the latter, a series of articles published in, 1970 by the Indian strategic analyst K. Subrahmanyam are particularly interesting, and we quote from them

in some detail.

The other countries of the world took their decisions to become nuclear weapon powers (China the last one in 1957) when civil nuclear technology had not developed. Consequently, in all these countries, military technology preceded civil nuclear technology. In India it is possible to reverse this process and move from a developed civil nuclear technology to weapons technology.²⁶

The proposals made by Dr. Sarabhai (then chairman of the Indian Department of Atomic Energy) on May 25 constitute a well-thought out and balanced programme for the development of nuclear option to the point when the Government will be in a position to decide to become an operational nuclear power. However, it may be emphasized that the programme as formulated now is still a programme of development of infrastructure only, and not a weapons development programme. It stops just short of that. This, however, need not discourage those who would like to see India become a nuclear weapon power. If we proceed on the lines proposed by Dr. Sarabhai, the point of decision will come three to four years from now. It is, therefore, not necessary for the government, at this stage to announce a decision. It can continue to proclaim honestly for the next three years that it has taken no decision to become a nuclear weapon power.²⁷

Rana Pratap Sagar and Kalpakkam (sites of the first two heavy water power projects in India, the former built with Canadian assistance) can be made to produce weapon grade plutonium but at uneconomic costs. Therefore if India decides to produce plutonium for weapons purposes it is necessary to set up new reactors the sole purpose of which will be to produce plutonium for weapons and not attempt to get it from power reactors. It will take at least three years from the date of decision to set up such reactors. Further such reactors will need heavy water and this heavy water will have to be produced in addition to our requirements for Rana Pratap Sagar and Kalpakkam. Therefore our heavy water capacity will also have to be augmented. This again will take time of the order of three to four years from the date of decision.²⁸

The costs of a complex designed for integrated production of some eight kilogrammes (95%) plutonium-239 per year and concentrated around a 40-50 MWth heavy water cooled and moderated reactor with a throughput of 20 tons metallic natural fuel per year can be estimated as follows...The reactor capital and operating costs are based on information concerning the Canadian reactor NRX.²⁹

Although this "latent proliferation" philosophy was adopted by the Indian Government, it is interesting that Subramanyam's realpolitik advice regarding the inadvisability of using plutonium from spent fuel irradiated in either the Rana Pratap Sagar power reactor or the CIRUS research reactor in a crash weapons programme was not followed.

Such a crash programme...will involve India breaking her treaty obligations to Canada. It has been argued that in the supreme national interest such an action would be justifiable. It is not so much the ethics of the action as the advisability of it that is to be considered. If we contravene our treaty obligations to Canada, there will be immense pressure on France to repudiate the technical collaboration agreement with India on space and on the fast breeder reactor. France even though not a signatory to the Non-Proliferation Treaty, has declared that she will behave as though she is a signatory in her relationship with other nations...

As pointed out earlier, the fast breeder reactor programme is the core of future nuclear energy development in India. In addition, our repudiation of treaty obligations with Canada is also likely to endanger the continued supply of enriched uranium for the Tarapar plant for which we depend on the United States. For these reasons, it is obvious that breaking of our treaty obligations with Canada will not be a prudent step and will not be advantageous to India.²⁷

In retrospect, Subramanyam was unnecessarily concerned about France's reaction, remarkably prescient regarding the U.S., but completely overlooked the impact on India's nuclear

program stemming from the adverse reaction of the country directly involved, Canada. In this connection, a recent critique of India's nuclear policy by G.S. Bhargava seems closer to the mark,

Considering the crucial importance of the Rajasthan reactors to the whole scheme of things, and in view of the need for continued collaboration with Canada, nothing worse could have been done than to have provoked the disruption of the arrangement with Canada...Against this background, embarking on a peaceful underground explosion which was not either directly or indirectly relevant to the power generation programme, and the objective of speedy self-sufficiency in nuclear technology and equipment, could only be described as an essay in adventurism. ³⁰

2.2 Other Heavy Water Reactors

Besides the natural uranium CANDU, there are four other heavy water moderated reactors of current interest. These are:

- (1) The natural uranium, on-power-fuelled, heavy-water cooled, pressure vessel reactor.
- (2) The mixed-oxide, light-water cooled, pressure tube reactor.
- (3) The slightly-enriched CANDU
- (4) The CANDU on the Thorium cycle.

Of the above, representatives of the first two are currently on-line in Argentina (ATUCHA I) and Japan (FUGEN), respectively. The latter two are, for the moment at least, "paper" reactors; (3) is characterized by uranium ore requirements approximately 20% smaller than the standard CANDU and about 40% smaller than the light water reactor on a once-through cycle,³¹ while the latter, operating in a recycle

mode, is seen by some as an economic alternative to the gas breeder reactor.³² Although the slightly enriched and thorium cycle CANDUs are interesting concepts, there are technical, economic, and political impediments to the commercialization of either, and for this reason we confine our discussion of these reactors to the following brief comments regarding their proliferation resistance as compared to the standard CANDU:

(3) The use of slightly-enriched fuel (typically 1.2% U-235) would make this reactor dependent on supplies of both enrichment and heavy water and reduce the amount of plutonium in the spent fuel to the level of current LWRs.³¹ This combination of features, taken together with its superior uranium resource utilization and the possibility of off-load fuelling, makes it a favorite among those who see a role for nuclear power, but wish to defer the plutonium economy as long as possible. Its penetration in developing countries on these grounds is dubious.

(4) The version of the thorium cycle which has received most attention is the so-called "denatured-uranium thorium cycle." The non-proliferation issues raised by this cycle are largely independent of whether the reactor utilizes light water, heavy water, or graphite moderation, and are concerned with the resistance of the isotopic barrier to misuse at the front-end of the cycle, the decreased plutonium at the back-end, and how it would be deployed. We refer the reader to reference³³ for a discussion of this concept by one of its chief proponents.

2.2.1 Heavy-Water-Cooled, Pressure Vessel Reactors

Reactor Physics and Engineering³⁴

Both pressure tube and pressure vessel designs utilize tubes in which the fuel resides and through which the coolant flows. The essential difference is that in pressure vessel reactors these tubes do not have to withstand a pressure differential. Accordingly, they may be made from thinner material, thus minimizing the amount of structural material, and hence neutron absorption in the core. As a compensating feature, the entire moderator at full primary system and at an elevated temperature must be contained by the pressure vessel; the latter must be much larger and thicker than an LWR pressure vessel for the same rated output because of the inferior slowing-down power of heavy water. Although the individual coolant feeder pipes characteristic of the pressure tube design are not necessary in pressure vessel reactors, the use of natural uranium fuel necessitates on-power fuelling in both, which means that external operable pressure connections to each coolant channel are a common feature.

Capsule History of ATUCHA I

The prototype of the 319 MWe ATUCHA I reactor built in Argentina by Siemens AG (first commercial operation June 1974) was the 50 MWe MZR reactor, (operational 1965), also built by Siemens at Karlsruhe in the F.R.G. In the consideration of the bids received for the ATUCHA station by the Argentine Atomic Energy Commission (CNEA) in 1967, there was a strong inclination on technological grounds in

favor of the CANDU; this because in addition to using natural uranium, it also had the added advantage that its design and engineering made it particularly suitable for important participation by the Argentine semi-heavy electro-mechanical industry. In particular, while the manufacture of a large pressure vessel was beyond the competence of the latter, there was confidence that it could master the technology involved in making pressure tubes and the calandria. Nevertheless, the Germans -- with their greater negotiating flexibility as compared to the Canadians -- carried the day.³⁵ As one participant in the negotiations has remarked: "Ideally, we would have liked a CANDU sold by Siemens. All the same, we paid for a Chevrolet and got a Cadillac."³⁶ For a comprehensive discussion of ATUCHA I, see reference 37.

Nonproliferation Considerations

As compared to the CANDU, where the fuel is contained in short bundles (0.5m), fuel in ATUCHA I is contained in long assemblies (5.3m). The fuelling pattern requires replacement of one assembly per day or 30 per month in contrast to the 300 bundles per month estimated for a 600MWe CANDU. The smaller number of larger fuel elements should make covert diversion of spent fuel more difficult. In this regard, we note that each fresh fuel assembly contains approximately 150 kg of uranium. At a discharge burnup of 6000 MWD/T,³⁸ CANDU fuel would contain about 0.32% Pu.³⁹

Neglecting the small differences in reactor physics between ATUCHA and CANDU,⁴⁰ this gives approximately 0.5 kg Pu per assembly with a Pu-239 isotope content of about 70%. Thus, 16 assemblies would contain about 8 kg of Pu. Aside from this difference in fuelling, the broader nonproliferation implications of CANDU and ATUCHA are similar. In particular, both utilize natural uranium at the "price" of a heavy water requirement of about 0.8 MT D₂O/MWe for initial inventory, or 265 MT for ATUCHA I.

2.2.2 Mixed Oxide (MOX), Boiling-Light-Water-Cooled (BLW) Pressure

Tube Reactors

Reactor Physics and Engineering

As compared with reactors such as CANDU and ATUCHA I, those which use heavy water for moderator only have the obvious advantage of saving the cost of the heavy water coolant, which amounts to approximately 0.3 MT/MWe. Moreover, in those BLW designs such as FUGEN and the British Steam Generating Heavy Water Reactor (SGHWR) which use enriched fuel, the heavy water moderator requirement is also reduced by about 0.1-0.2 MT/MWe. In particular, the total heavy water inventory in a MOX, BLW heavy water reactor is roughly half-0.4 MT vs. 0.8 MT per MWe - of that required for CANDU or ATUCHA. At a cost of \$250,000/MT D₂O this represents a considerable saving, e.g., \$60 million for a 600 MWE reactor. The implications of the use of boiling light water instead of pressurized heavy water as coolant on the reactor physics and engineering can be briefly summarized as follows.

If natural uranium is chosen as fuel, as in the Canadian Gentilly-1 reactor, the excess reactivity is so small that on-power fueling is essential. Even with this feature, the ratio of heavy to light water must be large in order to minimize the loss of neutrons via absorption in the latter, and hence the light water contributes little to neutron moderation. A loss of coolant, then, has a positive feedback effect on the reactor power; i.e., less coolant implies less neutron absorption, which leads to greater reactor power, which causes a further decrease in coolant density, and so on. Thus, such a reactor has a positive void coefficient of reactivity. (Since heavy water also absorbs neutrons, this effect is also present in a CANDU, but is much smaller because of the much smaller neutron absorption cross-section of heavy water, and hence is easily manageable.) There are basically two ways to alleviate this positive feedback effect. Both require the use of enriched fuel, which also affords the option of using off-power fueling. In the enriched uranium SGHWR, the ratio of heavy to light water is much smaller; in this situation the light water coolant contributes significantly to the moderation of neutrons; hence a loss of coolant implies both a decreased neutron absorption in the water and an increased resonance absorption of neutrons in the fuel, because of the hardened neutron spectrum. These effects tend to cancel; i.e., the increase in the thermal utilization is counterbalanced by a decrease in the resonance escape probability, and the void coefficient of reactivity is practically

zero. Another way to achieve this goal without paying the price of substantial neutron absorption in the coolant is to use plutonium MOX instead of enriched uranium as fuel. Since the neutron absorption in plutonium is greater than in uranium, the relative increase in thermal utilization due to loss of coolant is smaller in the former case. Hence, the heavy-to-light-water ratio can be higher than in the SGHWR. In addition, the use of Pu MOX means that the operating characteristics of the fuel remain essentially unchanged during irradiation, which helps to simplify the reactor system control design, this in contrast to the use of slightly enriched uranium fuel with a concomitant ingrowth of plutonium isotopes during irradiation. In the case of enriched designs, on-power fueling is an option having the advantage of a possible gain in fuel burnup at the price of increased fueling complexity and increased reactor "hardening" in areas of seismic activity.

Capsule History of FUGEN

Although an English graphite-moderated, gas-cooled reactor (GCR) was the first reactor to supply commercial power in Japan (1966), LWRs imported from the U.S. (roughly equal numbers of PWRs and BWRs) quickly became the dominant reactor type. Today, 18 LWRs generate approximately 14,000 MWe of power. However, even before the operation of the first GCR, the Japanese Atomic Energy Commission (JAEC) decided, in 1966, to establish a national policy for the development of advanced thermal reactors (ATR) of the heavy-water-moderated,

boiling light water-cooled type and fast breeder reactors (FBR). The rationale for this policy was a feeling of uncertainty about the future availability of enriched uranium fuel, and a desire to establish an independent fuel cycle capability. In particular, the ATR was seen as insurance in the event that the commercialization of the FBR was delayed. To implement advanced reactor development, the Power Reactor and Nuclear Fuel Development Corporation (PNC) was established in 1967.

A prototype ATR named FUGEN with a capacity of 165 MWe was designed by PNC, and its construction began in 1970. The original design called for the predominant use of 1.5% U-235 fuel in the core, as plutonium was difficult to obtain commercially. However, in the early 1970s, with the greater availability of plutonium, the design was changed to incorporate 96 (0.55-0.8% Pu) MOX fuel assemblies in the center of the core, with 128 (1.5% U-235) UO_2 assemblies around them. Using this configuration FUGEN went critical in March, 1978. However, the exclusive use of Pu MOX fuel is a basic philosophy of the FUGEN-ATR development, and the reactor will switch to this operating mode after the enriched uranium fuel in the initial core has achieved its design burnup. The conceptual design of a 600 MWE demonstration reactor based on FUGEN has already been completed, but the decision to begin construction has not yet been made by the JAEC. For further technical and historical details, see reference 41.

Nonproliferation Considerations

From the nonproliferation perspective the most vexing feature of the FUGEN system is that - unlike CANDU or ATUCHA I - it has been designed to operate using plutonium fuel. This opens the door to reprocessing and plutonium recycle in advanced thermal reactors, and thus undercuts the proposition that the plutonium economy can be postponed until it is clear that the continued viability of nuclear power dictates the need for fast breeder reactors.⁴² In this regard, it is interesting that a combined LWR-FUGEN system - with make-up plutonium for the latter derived from the spent fuel of the former - has a net uranium resource requirement essentially equivalent to a slightly enriched CANDU on a once-through cycle, and decidedly inferior to a CANDU operating on self-generated plutonium recycle.⁴³ This indicates that (1) uranium resource savings on the order of those achieved by FUGEN may also be achieved on once-through cycles, and that (2) there are more efficient thermal converters for operation on the recycle mode. Unfortunately, from the Japanese (and U.S.) perspective, both CANDU cycles have a fatal flaw: they suffer from "the NIH (not invented here) disease" which, given current political and economic realities, is probably incurable. Outside Japan, successful implementation of FUGEN may have its greatest influence on the nuclear establishments of its neighbors, Taiwan and South Korea, both of whom have a long-standing interest in heavy water-moderated, advanced converter reactors.

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2. IAEA Regional Fuel Cycle Centers, Vol. II, Vienna, 1977, Table II-I, p. 134. The SF neutron background rates have been calculated using the following SF isotopic rates cited in Table 1 of Ref. 1; i.e., Pu-238 (2,600 n/s-g), Pu-240 (1020 n/s-g), and Pu-242 (1,670 n/s-g). According to Lovins, the probability of avoiding pre-initiation varies as $1/\exp$ (neutron background rate x weapons assembly time). Besides S.F., an additional source of background neutrons are (α, n) reactions with light-element impurities in the plutonium.
3. Nuclear Proliferation and Safeguards, Congress of the United States, Office of Technology Assessment, Praeger, 1977, p. 159.
4. Letter from R.M. Smith, AECL Safeguards Coordinator, to Marvin Miller, March 1, 1979.
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9. A.J. Stirling and V.H. Allen, "The Application of Safeguards Design Principles to the Spent Fuel Bundle Counters for 600 MW CANDU Reactors," ref. 7, Paper IAEA SM-231/31.
10. S.J. Crutzen and R. G. Dennys, "Use of Ultrasonically Identified Security Seals in the 600 MW CANDU Safeguards System," ref. 7, Paper IAEA/SM/231-124.
11. An agreement has been reached by Canada and Argentina to incorporate on-power fuelling safeguards in the CANDU now under construction at Cordoba. Private communication, J. Stanton, External Affairs, Canada, July 9, 1979.
12. Ref. 5, Table 2.
13. See ; e.g., Nuclear Proliferation and Civilian Nuclear Power: Report of the Nonproliferation Alternative Systems Assessment Program (NASAP), Volume II: Proliferation Resistance, Table 3.3-7, p. 3-64, U.S. Department of Energy, July 1979.
14. Research towards this end in the U.S. is sponsored by the U.S. Arms Control and Disarmament Agency (ACDA). A description of a prototype system -- called RECOVER, for Remote Continual Verification -- is contained in a draft report submitted to ACDA by the Atlantic Research Corp., Alexandria, Virginia in December 1976, pursuant to Contract No. AC6AC734. Implementation of such intensive safeguard schemes probably depends more on their political acceptability than technical feasibility; in any case, ACDA has not adopted as an agency position the view that such safeguards are necessary.

15. These contentions are also discussed by R. Morrison, in "Is Canada peddling nuclear bombs world-wide in the guise of nuclear reactors?", Science Forum, December 1977, p. 3.
16. Nuclear Resources, Study Group Report produced for the Conservation Commission of the World Energy Conference by J.S. Foster et al., IPC Science and Technology Press, 1978, Tables III-1 and III-2, p. 80-81.
17. See the discussion by J.J. Noble in "Canada's continuing search for acceptable nuclear safeguards," International Perspectives, July/August 1978, p. 42.
18. Uranium: Resources, Production and Demand, Joint Report by the OECD Nuclear Energy Agency (NEA) and the IAEA, OECD, Paris, December 1977, p. 100.
19. Ref. 18, p. 87.
20. Canadian experience indicates that careful management can reduce losses to 0.3% of system inventory per year, equivalent to about 1.5 MT for a 600 MWe CANDU. See ref. 16, p. 145.
21. Article III of the Statute of the IAEA does provide for the Board of Governors to report noncompliance with safeguards undertakings to all members and to the Security Council and General Assembly of the United Nations. The Board may also curtail or suspend assistance, call for the return of materials and equipment, or in accordance with Article XIX suspend any noncomplying member from the exercise of the rights and privileges of membership.

22. Nuclear Suppliers Group: Guidelines for Nuclear Transfers, IAEA Document INFCIRC/254, February 1978, Appendix, Item 14(c).
23. See ; e.g., the discussion by S.J. Baker in "Commercial Nuclear Power and Nuclear Proliferation," reprinted in Export Reorganization Act of 1978, Hearing before the Committee on Government Operations of the U.S. Senate, 94th Congress, in S. 1439, U.S. Government Printing Office, Washington, D.C., 1976, p. 1878.
24. Although South Africa has a well-advertised ability to enrich uranium, its current capacity to produce low enriched fuel for reactors is limited.
25. In this regard we note that graphite production technology is not on the London Club trigger list, and that most artificial graphite, even material which is not nuclear-grade in terms of a specified maximum boron composition, usually has a low boron composition nonetheless. Private communication, A.E. Goldman, Union Carbide Corporation, 11/29/78.
26. K. Subramanyam, "The Path to Nuclear Capability," The Institute for Defence Studies and Analyses Journal, Vol. 3, No. 1, July 1970, P. 97.
27. Ibid., p. 95.
28. Ibid., p. 93
29. Extract from Nuclear Costs, U.N. Secretary General's Report A/6558, 1968, quoted in ref. 25, p. 59.

30. G. S. Bhargava, "India's Nuclear Policy," India Quarterly, Vol. 34, no. 2, April-June 1978, p. 131.
31. V. I. Chang et al., Alternative Fuel Cycle Options: Performance Characteristics and Impact on Nuclear Growth Potential, RSS Technical Memorandum No. 4, Applied Physics Division, Argonne National Laboratory, July 1977, Table I, p. 6 and Table III, p. 18.
32. A. J. Mooradian and O. J. C. Runnals, CANDU--Economic Alternative to the Fast Breeders, AECL-4916, September 1974.
33. H. A. Feiveson, "Proliferation Resistant Nuclear Fuel Cycles," in Annual Review of Energy, Volume 3, 1978, Annual Reviews, Inc., p. 357.
34. J. L. Gray and C. L. Moon, Heavy Water Moderated Nuclear Power Reactors, AECL-3660, June 1970.
35. For an interesting account of the ATUCHA I negotiations and related matters, see J. A. Sabato, "Atomic Energy in Argentina: A Case History," World Development, Vol. 1, No. 8, August 1973, p. 73. Sabato does not address the issue of the extent to which the German position on reactor safeguards as compared to that of Canada influenced the final decision.
36. J. A. Sabato, private communication, June 1979.
37. G. B. Herzog and K. J. Sauerwald, "ATUCHA Nuclear Power Station," Nuclear Engineering International, June 1969, p. 485.
38. This was the average discharge burnup in 1976. Operating Experience with Nuclear Power Stations in Member States in 1976, IAEA, Vienna, 1977, p. 15.

39. M. J. Halsell, Graphs and Tables of the Isotopic Composition of Plutonium Produced in Canadian D₂O Moderated Reactors, AECL-2631, January 1967.
40. In particular, the initial conversion ratios for CANDU (Pickering station) and ATUCHA are 0.82 and 0.814, respectively.
41. "FUGEN HWR reaches commercial operation", Nuclear Energy International, August, 1979, p. 33.
42. In this regard we note that the recent report of a working group on Future U.S.-Japanese Nuclear Energy Relations sponsored by the National Institute of Research Advancement, Tokyo, and the Rockefeller Foundation, New York, K. Oshima and M. Willrich, Co-Chairmen, November 1979, has recommended (p. 9) both that: "A decision on commercial plutonium recycle *in light water reactors* would be deferred until present uncertainties are reduced regarding the timing and scale of breeder reactor commercialization and the optimum manner of exploiting the fuel value of plutonium" and that "U.S. supply of enrichment services to Japan should provide in advance for the reprocessing of spent fuel containing U.S. enriched uranium and the use of recovered plutonium for the Japanese breeder *and other advanced reactor development and demonstration activities*" (italics mine). See also the interesting analysis by A. Kramish, "FUGEN: A Mirror of Japan's Nuclear Policy--The Significance for the U.S. and INFCE," Report No. RDA-TR-194203 (REV), R&D Associates, Arlington, Virginia, July 1979.

43. An LWR-FUGEN system with a ratio of 1.3 LWR to 1 FUGEN requires 112 tons of uranium per GWe year (reference 41, Fig. 15), while the slightly enriched and plutonium recycle CANDUs require 120 and 80 tons, respectively (reference 30).

3. The Heavy Water Genie

Since its discovery in 1932, and particularly in the 1950s and 1960s when interest in heavy water moderated reactors for civilian and military applications was at its zenith, many potential processes for large scale production of heavy water have been investigated¹ (For an extensive bibliography, see reference 2). Of these, very few have been pursued beyond the laboratory stage, and still fewer have been used for production on the order of tons of product per year. As of 1975, cumulative world production of heavy water - excluding China and the U.S.S.R.³ - was between eight and nine thousand metric tons, with just over 90 per cent having been made by the GS process. The remainder has been produced by hydrogen - water vapor exchange plus electrolysis (6%), hydrogen distillation (2%), ammonia - hydrogen exchange (1%) and water distillation (0.3%)⁴ (Table 1). As these figures indicate, the GS process holds the dominant position. It is the only proven process for large-scale production, and in this respect its position is similar to that of the gaseous diffusion process for uranium enrichment. Besides the GS process, there are five other techniques which merit attention. Of these, hydrogen-ammonia exchange and hydrogen distillation have been implemented commercially. For the other three; hydrogen-water exchange, hydrogen-amine exchange, and laser isotope separation, both theoretical and laboratory studies - and for the amine system, pilot plant experience - have demonstrated their potential promise. In sections A and B we review heavy water production fundamentals and process comparisons,

Table 1. HEAVY WATER PRODUCTION PLANTS*

<u>Site, Country</u>	<u>Primary Process</u>	<u>Most Recent (Design) Capacity, MT/yr.</u>	<u>Status</u>	<u>Startup</u>
1. Port Hawkesbury, Canada	GS	400	Producing	1970
2. Bruce A, Canada	GS	800	Producing	1973
3. Glace Bay, Canada	GS	400	Producing	1976
4. Bruce B, Canada	GS	(800)	Commissioning	1980
5. Bruce D, Canada	GS	(800)	Mothballed	?
6. Savannah River, S.C., U.S.A.	GS	69 (480)	Producing	1952
7. Dana, Ind., U.S.A.	GS	(490)	Shutdown (1958)	1952
8. Marzingarbe, France	NH ₃ -H ₂ Exchange	26	Shutdown (1972)	1968
9. Rjukan & Glomfjord, Norway	H ₂ O Electrolysis & H ₂ O-H ₂ Exchange	12	Producing	1934
10. Nangal, India	H ₂ O Electrolysis & H ₂ Distillation	14	Producing	1962
11. Kota, India	GS	(100)	Construction	1981
12. Baroda, India	NH ₃ -H ₂ Exchange	(67)	Commissioning	1980
13. Tuticorin, India	NH ₃ -H ₂ Exchange	(71)	Commissioning	1980
14. Talcher, India	NH ₃ -H ₂ Exchange	(63)	Commissioning	1980
15. Arroyita, Argentina	H ₂ O-NH ₃ -H ₂ Exchange	(250)	Planned	1983

*Capacity greater than 10 MT/yr.

respectively. Our intention here is not to write a primer on heavy water production - several excellent reviews are already available^{5,6} - but to provide enough background for an appreciation of the problems involved in attempting to implement this technology on both a large and small scale, sections C and D. In the latter, we remark briefly on some considerations involved in safeguarding large heavy water plants. Finally in Section E, we discuss some of the proprietary and political considerations which bear on the question of the spread of heavy water technology.

3.1 Heavy Water Fundamentals

The basic technical facts regarding heavy water production are as follows:

(1) Deuterium source materials which are in sufficient supply to be used as feed for large-scale heavy water production are: water, petroleum, methane, hydrogen and ammonia. Since water is by far the most abundant of these materials, heavy water production processes which utilize it as the feed material, the GS process in particular, have essentially unlimited plant capacity, whereas the capacity of those processes which utilize one of the other feedstocks such as ammonia synthesis gas are limited by the availability of the latter, and must be sited in proximity to its source; e.g., the ammonia synthesis plant.

(2) The natural deuterium abundance in potential feedstocks is very small, varying from roughly 90 to 160 ppm atom %.⁴ Since the cost of heavy water production is approximately inversely proportional to the deuterium content of the feed - basically because the more dilute the feed, the greater the amount of material that has to be processed to extract a given amount of deuterium - there is a strong economic incentive in using feed of the highest possible concentration. At the same time, the feed should not corrode standard construction materials - unfortunately this makes the use of sea water a questionable proposition - and should also be reasonably free of impurities, since even traces of the latter can cause severe operating problems. For example,

(a) Various impurities in the feed water to the GS process can cause foaming of the gas-liquid mixture on the contacting trays leading to large decreases in the production rate.

(b) Solidification of the nitrogen impurity in the synthesis gas feed to a hydrogen distillation plant causes eventual blockage in key process components.

(c) Oxygen impurity in the synthesis gas feed to a hydrogen-ammonia exchange plant can react explosively with the potassium amide catalyst used in this process.

The stringency of the purification task stems from the large amount of feed material which must be processed per unit of product. For example, at a 800 ton per year GS plant, approximately one ton of freshwater is processed per second. If the efficiency of feedwater treatment is such as to reduce the impurity level in the feed to one ppm, about one ton of impurity would still build up in the system every two weeks.

(3) The low deuterium abundance in the feed, typically 150 ppm in fresh water, coupled with the high concentration of deuterium required for reactor-grade product implies that the equipment must effect an increase in abundance ratio of about 3.3×10^6 . The incremental enrichment provided by an individual separating element in all known heavy water production processes is not sufficient to span this concentration range. Thus, many elements must be connected in series to multiply the elementary effect. For example, in the GS process, the individual separating elements are sieve trays on which the upflowing hydrogen sulfide gas and downflowing

water are successively mixed. While it is possible in principle to obtain the required overall enrichment in a single tower or stage, in practice it is advantageous instead to use several towers of progressively smaller size in series; i.e., an enrichment cascade. This procedure takes advantage of the smaller flows of enriched material in the higher stages, and reduces the total tower volume and heavy water inventory.

(4) In all processes, the lower stages dominate the plant capital and energy costs. An equivalent statement is that most of the separative work is required at the bottom of the cascade. In heavy water production, it is common practice to use one process for extraction of deuterium from feed to an abundance on the order of 1-10% and another process such as water distillation, which would be too costly in terms of separative work per unit product for extraction, for concentration to reactor grade product. Besides its inherent simplicity, the basic advantage of water distillation is that it can be operated at sub-atmospheric pressure, thus minimizing the potential for leakage of the enriched water.

3.2 Process Comparisons

The two most useful indices of the economic attractiveness of an isotope separation process are the increment of enrichment possible in a single separating element, α ,⁷ and the energy consumption per unit of the product. The importance of the former stems from the fact that the size and hence the initial cost of the plant as well as the thermodynamic minimum energy consumption for irreversible processes such as gaseous diffusion and electrolysis are proportional to ϵ^{-2} , $\epsilon \equiv (\alpha-1)$. For reversible processes such as distillation, chemical exchange, and centrifugation, the thermodynamic minimum energy consumption is proportional to the entropy of mixing which is independent of ϵ . In practice, the energy consumption of reversible processes is much greater than the mixing entropy, and depends on ϵ . In particular, for chemical exchange and distillation, most of the energy is consumed in providing the reflow of the working substance (technically, the reflux) at the ends of the tower rather than in the contacting equipment where the isotope separation takes place. These refluxing costs are proportional to ϵ^{-1} . Generally, for reversible processes, the actual energy consumption depends on the details of the plant design, and represents a tradeoff between the added capital cost of equipment—such as additional heat exchangers in dual-temperature chemical exchange processes and sophisticated rotor bearings in centrifugation—which can reduce the energy consumption. A comparison of five separation processes with

respect to separation factor and energy consumption is given in Table 2. It is clear that the mass dependence of ϵ favors electrolysis, distillation and chemical exchange for heavy water production, provided the working substance in distillation is hydrogen. The latter two are equilibrium processes and their utility for isotope separation; i.e., the fact that α differs from unity, is a purely quantum phenomenon stemming from the differences in the vibrational energy levels of the isotopic molecules.⁸ The energy requirements for heavy water production of both chemical exchange and hydrogen distillation are moderate. The same cannot be said of either electrolysis or gaseous diffusion. Indeed, if heavy water had to be made by these processes, the cost of the heavy water inventory for a CANDU reactor would make the system uncompetitive with LWRs. As is well known, the situation is reversed for heavy elements such as uranium. Here, the mass dependence of α favors gaseous diffusion and the centrifuge, with the latter also enjoying a considerable advantage compared to the former with respect to energy consumption.

In sum, the chemical exchange processes and hydrogen distillation are the only realistic non-laser possibilities for economic production of heavy water. Of these, the standard of comparison is, of course, the GS process. In the following we briefly discuss these processes as well as the prospects for laser isotope separation of deuterium.

Table 2. COMPARISON OF SELECTED ISOTOPE SEPARATION PROCESSES

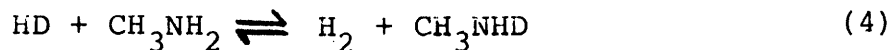
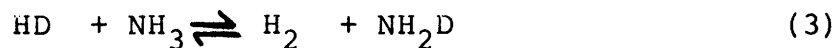
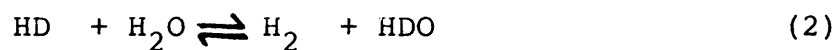
Process	Isotopic Mixture:	Mass Dependence for large m	H ₂ -HD		235UF ₆ - 238UF ₆	
			Separation Factor, α	Energy Consumption	Separation Factor, α	Energy Consumption
Distillation	ε ~ Δm/m ²	1.5	22 GJ/kg D ₂ O	1.00009	620 W/kg SWU/yr	
Chemical Exchange	ε ~ Δm/m ²	5.5	20 GJ/kg D ₂ O	1.0016	---	
Electrolysis	ε ~ Δm/m	10	800 GJ/kg D ₂ O	---	---	
Gaseous Diffusion	ε ~ Δm/m	1.225	~10 ⁵ GJ/kg D ₂ O	1.00429	250 W/kg SWU/yr	
Gas Centrifuge (peripheral speed: 500 m/sec)	ε ~ Δm	1.056	---	1.162	20 W/kg SWU/yr	

NOTES:

1. Values for separation factors and energy consumption in H₂-HD distillation, (monothermal H₂-NH₃) chemical exchange, and electrolysis of water, from Ref. 5, Fig. 14, p. 21. Energy estimated by converting electrical or mechanical energy to equivalent thermal energy using 40% efficiency, and adding to actual thermal energy to give total equivalent thermal energy.
2. Energy consumption for gaseous diffusion of H₂-HD is thermodynamic minimum compressive work in ideal cascade enriching to 0.1 atom fraction D from feed containing 1.5 x 10⁻⁴ atom fraction D and tails stripped to 5 x 10⁻⁵ atom fraction; conversion to equivalent thermal energy as above.
3. Values of separation factor for ²³⁵UF₆-²³⁸UF₆ distillation and chemical exchange and energy consumption for former from M. Benedict et al., Report of Uranium Isotope Separation Ad Hoc Review Committee, USAEC Report OR0-694, June 1974, Table 2.2.1., p. 2.3.
4. Centrifuge separation factors are maximum radial values at 300°K. In a counterflow centrifuge of sufficient length, the separation factor between the top and the bottom may be much greater.

3.2.1 Chemical Exchange Processes

● The reactions of interest are those between hydrogen sulfide gas and water (GS process), and between hydrogen gas and water, ammonia, and methylamine. At low deuterium concentrations the heavy isotope is present primarily in the monodeuterated form, and hence these reactions can be represented respectively as,



The separation factor for these processes is directly proportional to the reaction equilibrium constant, K. For example, for the GS process it is easily seen that

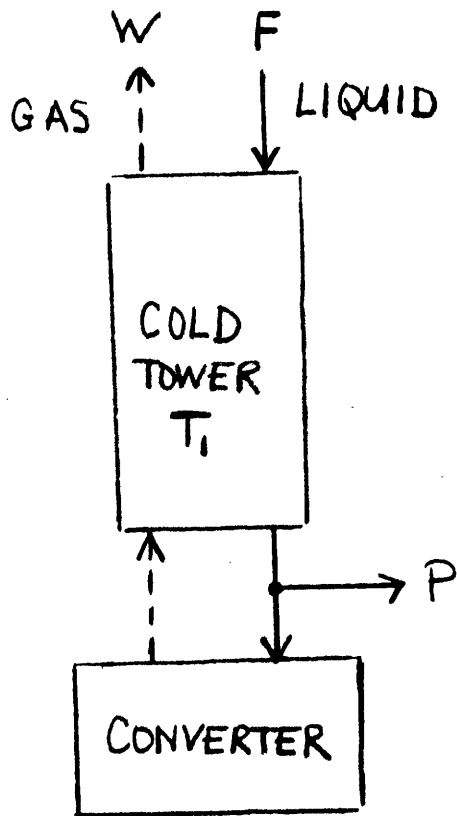
$$\alpha = \frac{(\text{D}/\text{H})\text{H}_2\text{O}}{(\text{D}/\text{H})\text{H}_2\text{S}} = \frac{[\text{HDO}]/[\text{H}_2\text{O}]}{[\text{HDS}]/[\text{H}_2\text{S}]}$$

is just equal to K.⁹

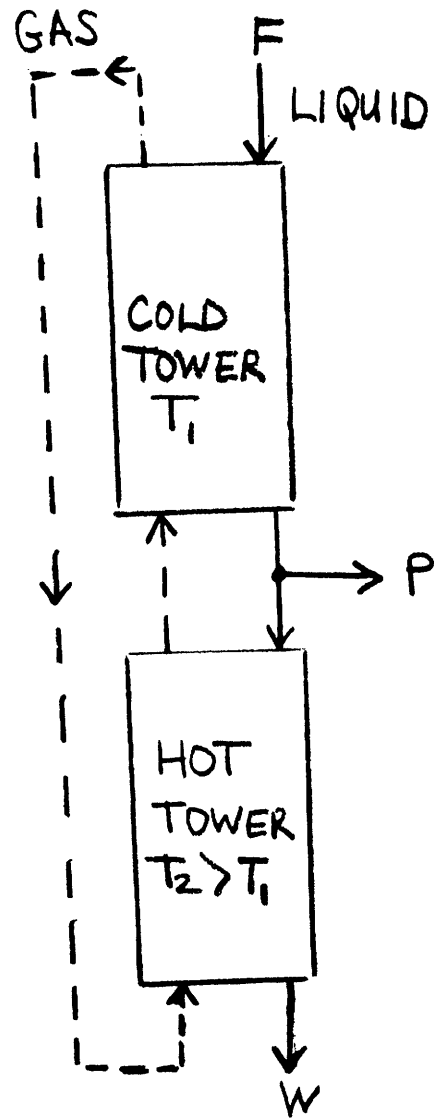
As previously noted, the fact that K and hence α for this reaction differs from unity, or more generally, that the equilibrium constant of chemical exchange reactions differs from the value corresponding to classical equipartition of the isotopes between the two chemical species, is a strictly quantum-mechanical effect. It arises because of differences

in the internal vibrational frequencies of the isotopic molecules. The deviation from classical behavior and hence the separation effect is greatest at low temperatures and for exchanging molecules in which the chemical binding of the proton and deuteron differs most widely. In this regard, the hydrogen-based systems, (2)-(4), particularly the non-aqueous ones, have a marked advantage compared with the GS process where the similarity between the hydrogen bonds with oxygen and with sulfur limits the separation factor. The "other side of the coin" is that this similarity facilitates the reaction kinetics; by comparison the rate of exchange in the hydrogen-based systems is very slow, especially at the low cold tower temperatures at which they must be operated in order to realize their potential advantage in separation factor. While sufficient solubility of hydrogen sulfide in water and rapid isotopic exchange can be achieved at moderate pressures on simple sieve trays in the GS process, high pressure, sophisticated contacting equipment, and catalysis are required in hydrogen-based process to dissolve enough hydrogen in the liquid solution and promote the exchange reaction.

● In principle, gas reflux for process (1)-(4) can be produced in either of two ways leading to two basic flowsheets: mono-thermal or bithermal (dual-temperature). In the former, the enriched liquid is chemically converted to the gas at the bottom of the tower, Fig. 1a. In the latter, Fig. 1b, the depleted gas and enriched liquid from the exchange tower are fed to another tower operating at a higher temperature. In



(a)



(b)

Figure 1. Monothermal and Bithermal Flowsheets for Chemical Exchange.
($F \equiv$ Feed ; $P \equiv$ Product ; $W \equiv$ Waste)

this "hot" tower the reaction equilibrium constant is smaller than in the "cold" tower, and the gas is reenriched at the expense of the liquid. Formally, the hot tower replaces the chemical conversion step as a source of enriched gas reflux. However, there are important differences between the two methods. These stem from the fact that while the deuterium abundance of the reflux in monothermal operation is equal to that of the liquid input to the phase converter, the abundance of the reflux provided by the hot tower in the bithermal method is much lower. This implies a larger gas flow, more contacting equipment, and a larger cold tower volume to recover the same amount of deuterium from the feed stream in a bithermal vs. a monothermal realization of the same exchange process. Moreover, production of enriched product between the two towers in bithermal operation demands precise control of the gas-to-liquid flow ratio; this is not required in monothermal operation. Unfortunately, chemical reflux is only practical in the hydrogen-water and hydrogen-ammonia systems. In the former, economic heavy water recovery is possible because the cost of the chemical conversion step, water electrolysis, is charged to hydrogen production for ammonia synthesis. In the latter, the feed is ammonia synthesis gas, and the ammonia which is cracked at the bottom of the exchange tower for gas reflux must also be synthesized for liquid reflux at the top of the tower. Although the added costs of monothermal operation - primarily associated with the energy needed for ammonia cracking - lead to an energy requirement larger than that

possible in the dual-temperature realization of this process, a priori, this does not make a monothermal operation economically uncompetitive. Indeed, up to this time, only the latter process has had commercial operating experience. Unfortunately, the same is not true of hydrogen sulphide - water exchange where the cost of chemical conversion would be prohibitive,¹⁰ and only bithermal operation, the GS process, is economic.

- Both the GS process and hydrogen-water exchange have the advantage that their process heat requirements fall within the capability of nuclear steam, by far the cheapest heat source available today. The other exchange processes require either heat at much higher temperature; e.g., for ammonia cracking in monothermal hydrogen-ammonia exchange, or expensive refrigeration; e.g., the cold tower in hydrogen-ammonia and methylamine exchange as well as hydrogen distillation.

- While the GS process requires stringent quality control because of the toxicity and corrosiveness of hydrogen sulphide, it does not require catalysis, and utilizes a water feed allowing essentially unlimited production. Moreover, many man-years have been spent in process optimization, particularly in Canada, and a new process would have to offer substantial incentives in terms of cost or safety to offset the accumulated experience with GS. Of the hydrogen-based processes, hydrogen-water exchange combined with electrolysis for hydrogen production would be especially attractive since the process fluids are not toxic and only mildly corrosive,

and the relative tower volume is only about one-third that required in the GS process.⁵ Historically the basic technical problem with this system has been the lack of a catalyst which retains its activity in contact with liquid water. Development efforts in this direction at AECL appear very promising.¹¹ However, at this time, there is no economic incentive to produce large quantities of electrolytic hydrogen, and thus this method - as well as hydrogen distillation - will find application only in special situations. Of the two non-aqueous hydrogen exchange parasitic processes, the greater solubility and higher exchange rate in the amine system as compared with ammonia permits operation at a lower temperature and leads to a smaller plant volume in bithermal operation. Unfortunately, a full scale plant based on the amine process has not been built, although extensive pilot plant experience has accumulated. The choice between a monothermal or a bithermal flowsheet for the hydrogen-ammonia system revolves mainly on the relative importance of a larger plant volume for the latter but also a smaller energy consumption. A considered judgement awaits the commissioning of the Indian plants and the accumulation of significant operating experience.

3.2.2 Hydrogen Distillation

Because of its high deuterium separation factor, distillation of liquid hydrogen is a potentially attractive method for producing heavy water. Several relatively small plants (3-14 tons per year capacity) built and operated over the past

20 years in Western Europe, India and the U.S.S.R. have demonstrated the technological feasibility of this process; e.g., the ability to handle large quantities of liquid hydrogen at cryogenic temperatures of 20° K. Its economic competitiveness hinges on the nature and quantity of the available hydrogen feed material. In common with all hydrogen-based deuterium separation methods, distillation must be operated as a parasitic process; otherwise the cost of the feed alone would be prohibitive. (For example, assuming a 90% recovery of hydrogen from feed and a hydrogen cost from methane reforming of \$0.03 per STP cubic meter, the feed cost alone would be \$250 per kg. D_2O .) There are basically two sources for feed: ammonia synthesis gas and electrolytic hydrogen. The former contains about 75% H_2 , 25% N_2 , and small amounts of CH_4 , A , CO_2 , O_2 and H_2O , while the latter contains only H_2O and trace amounts of N_2 and O_2 . As previously noted, the distillation process requires extremely thorough purification of the feed gas to eliminate condensed impurities which would cause fouling of the equipment. This makes the flowsheet of a plant using synthesis gas as feed more complicated, and its energy consumption higher than one using electrolytic hydrogen. Unfortunately, sources of electrolytic hydrogen are far less common than synthesis gas, and if only the latter is available, the hydrogen-based chemical exchange systems, especially amine, may be more attractive. The prospects for heavy water production via hydrogen distillation would be considerably brighter if the industrialized

countries decided to produce large quantities of liquid hydrogen as a synthetic fuel to replace hydrocarbons. In this case, the cost of refrigeration could be charged to the liquid hydrogen plant instead of the heavy water extraction. However, at present the hydrogen economy seems a distant prospect. The key aspects of the chemical exchange processes and hydrogen distillation are summarized in Table 3 .

3.2.3 Laser Isotope Separation (LIS)

The physical basis for photochemical separation of isotopes is the fact that atomic and molecular vapors absorb radiation only at well-defined (discrete) frequencies, specific to every atomic or molecular species. Isotopic substitution in an atom or molecule can change the absorption spectrum both qualitatively and quantitatively, by changing the number and position of the spectral lines. These (usually) small differences in the spectra of different isotopes can be exploited by tuning a narrow-band laser to excite only one species in an isotopic mixture, thus bringing about large changes in the internal state of the isotope; e.g., stripping it of an electron, changes which are then used to effect macroscopic separations. The possibility of photochemical isotope separation is not new; it was recognized in 1920,¹² but before the discovery of the laser, it could not be exploited commercially because the required isotopic selectivity and spatial coherence of the light source could only be obtained by

Table 3. SELECTED HEAVY WATER SEPARATION PROCESSES

<u>Method</u>	<u>Advantages</u>	<u>Disadvantages</u>
Hydrogen Distillation	Moderate separation factor (~ 1.5); small tower volume; moderate energy consumption; extensive small plant experience	Parasitic process (limited capacity); only competitive with GS if cold, pure hydrogen source available; operation at 20°K
GS	Water feed (unlimited capacity); moderate separation factor (~ 1.3), tower volume and energy consumption; well developed technology	Process fluids toxic and corrosive; sensitive process control
H ₂ O-H ₂ Exchange (monothermal)	Process fluids not toxic and only mildly corrosive; high separation factor (~ 3.9); small tower volume; low energy consumption	Hydrophobic platinum catalyst still under development; parasitic process (limited capacity)
NH ₃ -H ₂ Exchange (monothermal)	High separation factor (~ 5.5), moderate tower volume and energy consumption; simple to control; demonstrated technology	Requires catalysis, operation at high pressures, stringent gas purification, sophisticated contacting equipment, NH ₃ cracking and synthesis; parasitic process (limited capacity)
NH ₃ -H ₂ Exchange (bithermal)	Moderate separation factor (~ 1.7); low energy consumption; as compared with monothermal mode, chemical conversions and catalyst stripping not required	Large tower volume; requires catalysis, operation at high pressures, stringent gas purification, sophisticated contacting equipment; parasitic process (limited capacity)
Amine-H ₂ Exchange (bithermal)	Moderate separation factor (~ 2.2); small tower volume; low energy consumption	Requires catalysis; sophisticated contacting equipment; catalyst unstable above 70°C; parasitic process (limited capacity)

spectral and spatial filtering of a conventional light source to the extent that most of its output energy is wasted. By contrast, the large light intensities within a narrow spectral bandwidth characteristic of lasers allows processing of a significant amount of isotopic material, while the high degree of spatial coherence allows efficient use of large f-number optical systems to keep the unilluminated volume to a minimum. The potential of LIS for uranium enrichment was recognized quite soon after the discovery of the laser by a number of investigators in different countries. However, because of the sensitive nature of this application, the historical development of LIS of uranium has been made quite unintelligible by secrecy; this is still largely true. By contrast, much of the research on LIS of isotopes other than uranium has been reported in the open literature. In particular, over the past few years work has been in progress in the U.S., Canada and several other countries on the application of LIS to heavy water production. In terms of research expenditures, articles in scientific journals and the popular press, and concern in the nonproliferation community, LIS of deuterium is a poor relation of uranium, except perhaps in Canada. This is, to a certain extent, justified by the economic facts of life. In the first place, the current demand for enriched uranium is much greater than that for heavy water. Moreover, using current technologies, enriching uranium is much more expensive than heavy water production,¹³ and hence considerable ingenuity is required to invent a laser separation technique for the latter which has a chance

of displacing the GS process. However, if this could be done; e.g., if LIS could produce heavy water at 1/5 to 1/2 of its present cost, it would enhance the market for the CANDU reactor, and the process would be very attractive, especially to countries which had not already made a substantial commitment to other methods. Here, we briefly review the recent research which applies the technique of isotopically selective multiple photon dissociation (MPD) to deuterium separation. General discussions of the prospects for heavy water via LIS are given in references 14 and 15, while results of research in techniques other than MPD can be found in references 16-18. The basic idea in MPD is quite simple: an infrared laser is tuned to one of the characteristic molecular vibrational frequencies of the deuterated species in a natural mixture, and dissociation takes place when the number of photons absorbed from the beam is sufficient to reach the threshold energy for dissociation via a process of "climbing the vibrational ladder". However, the details of MPD, e.g., its dependence on: the energy and length of the laser pulse, the rotational structure of the molecule, and molecular collisions, makes it a very complicated subject. Most attention has been focused on the molecule uranium hexafluoride, UF_6 , and its unclassified surrogate, SF_6 . However, recent work has demonstrated that MPD has promise for deuterium separation as well. In this regard, Marling, Herman and Thomas have summarized some of the requirements for the use of this technique in a commercially viable process as follows:

- (1) The single-step deuterium enrichment factor must exceed 1000 to minimize make-up costs, i.e., the replacement cost of optically destroyed working molecules. In addition, a ratio of greater than 1000-to-1 and preferably 10^4 -to-1 should exist in the optical absorption coefficients of the mono-deuterated and normal molecule at the laser wavelength and energy fluence where reaction occurs. This is essential for gas-phase medium penetration and efficient photon utilization.²⁰
- (2) The selective absorption must be accessible by an efficient, high-peak and high-average-power laser--essentially restricting consideration to only the CO₂ laser.
- (3) The protonated molecule must undergo rapid deuterium exchange tied to an aqueous or natural gas feedstream for redeuteration of the working material, if water or methane is not used directly.
- (4) Hydrolysis, solvolysis or other irreversible reactions of the working material with the exchange medium must be at least 10^3 - 10^4 times slower than the rate of redeuteration. This insures that the major loss of the working material per cycle (amounting to 0.01-0.1%) is dominated by the photochemical removal of deuterium

and not losses during the redeuteration step.

- (5) The deuterium-bearing photoproduct must be resistant to D/H exchange with the working molecule.
- (6) The yield of deuterium-bearing photoproduct should approach unity ($\gg 0.5$) under MPD fluence conditions consistent with high single-step deuterium enrichment.
- (7) The spectroscopy and photochemistry of the candidate molecule must allow high pressure operation in the range of $\gg 0.1$ atm. This is essential for realistic gas pumping costs and manageable photochemical reactor size.
- (8) The working material should be of low cost ($\ll \$2/\text{kg}$), adequately volatile and (ideally) non-flammable, non-carcinogenic and non-toxic.

In the light of the above, it is instructive to make a "back-of-the envelope" calculation of the laser costs for a process based on MPD of the trifluoromethane molecule, CHF_3 .

The laser energy required per separated D atom, E_D , for deuterium separation by CO_2 laser MPD of CHF_3 is given by the expression

$$E_D = \frac{nhc}{\epsilon_0 \eta \lambda} \left[1 + \frac{1}{S(D/H)} \right], \quad (5)$$

where we use the measured values²¹ of optical selectivity,

$S = 1500$ at a photolysis wavelength λ of $10.3\mu\text{m}$, and dissociation probability ϕ of one at a laser fluence of $30\text{J}/\text{cm}^2$ with sufficient buffer gas, and assume: the dissociation barrier $n = 25$ photons, the laser efficiency $\epsilon = 14\%$, the molecular excitation efficiency $\eta = 0.5$, and the deuterium abundance $(\text{D}/\text{H}) = 150$ ppm. Equation (5) then gives a laser electrical energy $E_D = 0.25$ Kev per separated D atom, or about 0.6 Kev of equivalent thermal energy as compared with approximately 2.7 Kev per D atom for the GS process. Despite the fact that the former estimate neglects non-laser related energy costs associated with pumping the gas and redeuterating the depleted CHF_3 , it is still rather encouraging. Unfortunately, the same cannot be said of the laser capital costs for this process. It is straightforward to show that this is about 75 watts of installed capacity/kg $\text{D}_2\text{O}/\text{yr}$. If TEA laser power is available at $\frac{\$100}{\text{watt}}^{22}$ then at a capital charge rate of 0.15/yr. the laser capital cost component for this process would be $\$7,500/\text{kg D}_2\text{O}/\text{yr} \times 0.15/\text{yr} = \$1125/\text{kg D}_2\text{O}$. This again illustrates the fact that high quality (low entropy) laser energy is expensive, and unless order of magnitude reductions in capital cost can be achieved, lasers may be uneconomic for this application.

3.2.4 Transfer Processes

As noted in Chapter I, transfer processes are those which take advantage of the high separation factors of hydrogen-based processes without the capacity limitations inherent in parasitic operation. The generic means to this end is to contact one of the depleted exchange partners in a hydrogen-based process with

a natural water feed, thereby transferring deuterium from the latter to the former. The price one pays for this flexibility is the capital and operating costs of the transfer unit and the associated equipment required to make the transfer operation compatible with the flowsheet of the hydrogen-based process. The potential for linking the amine-hydrogen process to a water feed via either hydrogen-water or amine-water exchange is discussed by Rae²³ and by Sulzer²⁴, respectively. Their basic conclusion is that the need for catalysis in hydrogen-water exchange and the fact that the exchange tendency is in the "wrong" direction for this application--i.e., it favors transfer of deuterium from hydrogen to water, thus necessitating operation at high temperatures--makes this route less attractive than amine-water exchange. Since the amine-hydrogen process itself has not been implemented commercially, it is perhaps premature to consider a transfer process incorporating it in further detail; of more immediate interest is the transfer process combining monothermal ammonia-hydrogen exchange with redeuteration of the ammonia via a water contact.²⁵ Since a detailed flowsheet of this process is not available at the present time, we content ourselves here with some educated guesses as to what it might entail.

A simplified flowsheet is shown in Figure 2. As indicated, liquid ammonia, depleted in deuterium, from the ammonia synthesis unit of the hydrogen-ammonia process (1), is depressurized and introduced as ammonia vapor at the bottom of the transfer section of the extraction tower (2). Here the upflowing gas

becomes reinriched with deuterium via countercurrent chemical exchange with downflowing natural water introduced at the top of this section (3). The distillation sections at the bottom (4) and top (5) of the extraction tower strip dissolved ammonia from the waste water before discharge, and strip water vapor from the redeuterated ammonia gas before it is reintroduced into the hydrogen-ammonia process. The latter step is particularly important since, as previously noted, the potassium amide catalyst used to promote hydrogen-ammonia exchange forms explosive mixtures with oxygen compounds. In addition, this anhydrous ammonia must be condensed (6) and pumped (7) to the high operating pressure of the hydrogen-ammonia exchange tower which consists of stripping and enriching sections (8). Also indicated are the ammonia cracking unit for gaseous reflux and the final treatment step to produce reactor-grade heavy water. Not shown is the equipment for catalyst deuterium stripping and separation.

The operating parameters of the extraction tower; e.g., temperature, pressure, number of theoretical plates would, of course, be chosen so as to minimize the incremental cost of heavy water produced by hydrogen-ammonia exchange due to contact with water. This implicitly assumes that the latter would be built around a standard monothermal hydrogen-ammonia flowsheet; e.g., as practiced at Marzingarbe. Since transfer operation frees one from the constraint of designing for much the same pressure and temperature range as the associated ammonia plant, this is not necessarily the case. Thus, the

values shown in Figure 2 may be far from optimum, and are given for illustrative purposes only.

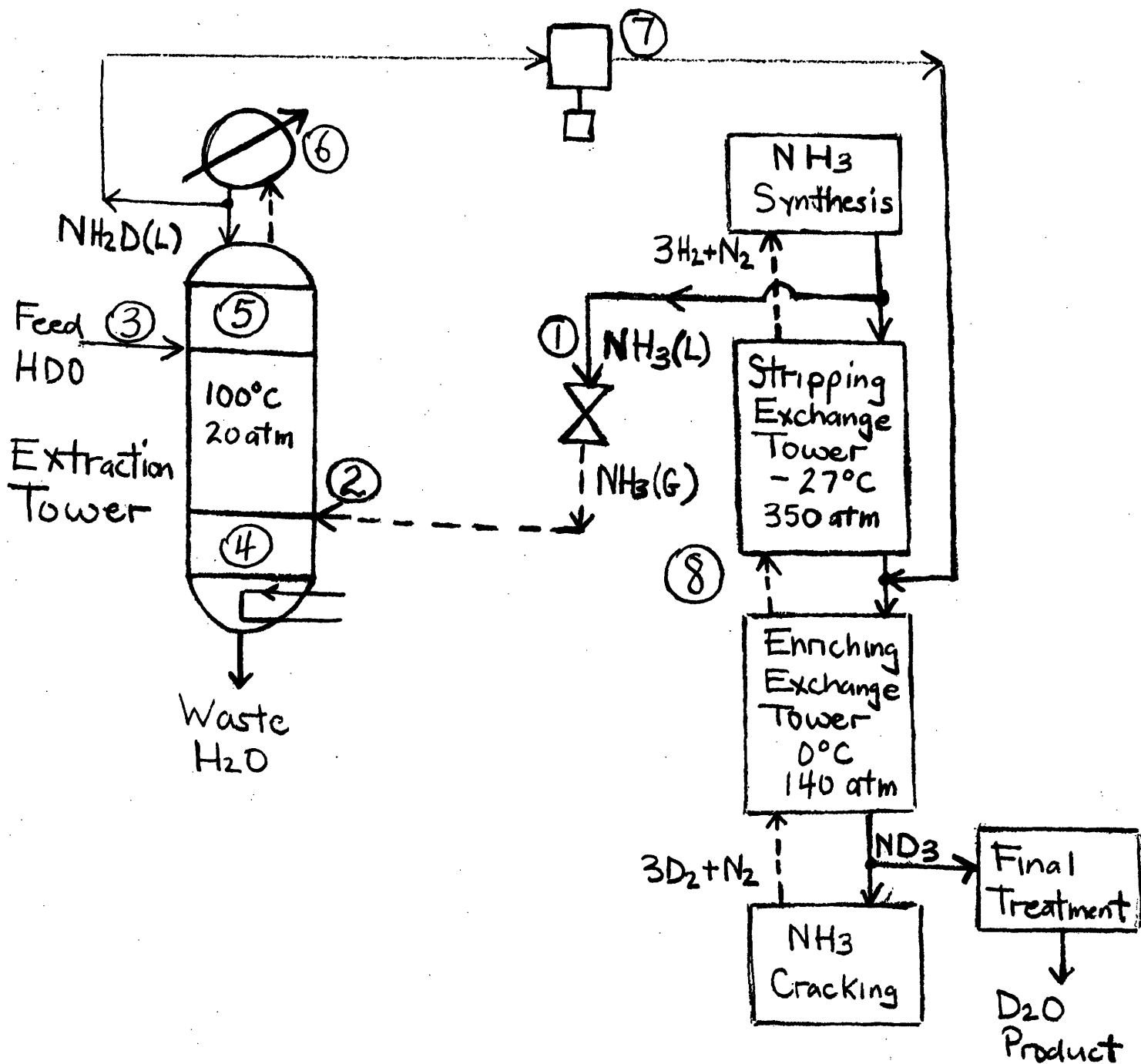


Figure 2. Monothermal Ammonia-Hydrogen Exchange Process with Water Feed

3.3 Implementing the Technology on a Large Scale

The basic question we wish to consider here is the following: to what extent can various heavy water production processes be implemented in a particular country at a given time? In particular, we assume that the processes have been developed elsewhere, so that what we are considering is a specific example of the general issue of conditions for the successful transfer of technology from developed to developing countries. While we do not rule out the possibility of innovation in heavy water technology in a developing country, it seems reasonable in the light of historical experience to expect that such innovation will take the form of variations on a technique developed elsewhere, rather than pioneering research and development.

An additional caveat concerns how we define successful implementation of technology. Of late, that aspect of the problem which is conjured up by the phrase "appropriate technology" has received increasing attention.²⁶ These dimensions of technology transfer are certainly interesting, and in some sense lie at the heart of the matter. Unfortunately, an adequate discussion would carry us too far afield. Here we adopt a narrow, operational view of success: can the plants be built in a time, at a cost and operated at a capacity factor characteristic of plants of the same type in developed countries? And if not, why?

A key factor here is the availability of and the extent of reliance on outside assistance. At one extreme is the classic turnkey operation wherein the basic design of a facility, as well as the key personnel, hardware and software needed to implement the design are all imported. At the other end of the spectrum is the essentially indigenous undertaking. Here, while it occasionally proves profitable to take advantage of the availability on a timely basis of selected design data and particular items of equipment from outside suppliers, there is often an official policy of maximizing the indigenous component, even at the price of a significant time/cost penalty. In practice, the design, construction and operation of plants utilizing what might loosely be called "high technology" in most countries, developed and developing, involves degrees of outside assistance which fall between these extremes. In the particular case of heavy water processes, it would seem at first glance that there are a number of factors which would tend to enhance the prospects for the successful implementation of this technology in those developing countries with a significant industrial sector; e.g., India, Argentina and Taiwan.

(1) the existence in the open literature of a wealth of relevant information; specifically, data relating to process fundamentals, flowsheets, materials and construction techniques, and operating history, particularly for the GS process. In fact, using such information, pilot GS plants have been con-

structed in a number of countries, including India and Romania.

(2) as compared with either nuclear reactors and spent fuel reprocessing plants on the one hand or facilities for uranium enrichment via gaseous diffusion or the gas centrifuge on the other, heavy water production involves neither containment of intensely radioactive nuclear materials nor sophisticated technologies which, moreover, have been closely held on nonproliferation grounds.

(3) the existence of other industrial processes which involve operations such as countercurrent chemical separations, catalysis, and handling of some of the same materials used in heavy water processes. Of particular relevance are hydrocarbon fuel refinery operations such as fractional distillation, catalytic cracking, and desulphurization, especially of sour natural gas.

Despite the above, the historical record indicates that construction and successful operation of heavy water production plants is difficult, even in developed countries such as Canada. The countervailing factors include the following:

(1) Although much has been published about heavy water production processes, there are important pieces of information which are closely held, for both nonproliferation and proprietary reasons. For example, for large GS plants, the details of such matters as: feedwater treatment, sieve tray design, process control, behavior of impurities in the system, and materials performance and corrosion are not readily available.

A similar statement could be made about the other process of current commercial interest: hydrogen-ammonia exchange.

This is not academic, since insight in these areas can mean the difference between a plant which works haphazardly at best, and one that operates reliably at design capacity. In this connection, the point is often made, especially with regard to clandestine facilities for reprocessing irradiated nuclear fuel, that an important difference might exist between a commercial plant and a "quick-and-dirty", weapons-dedicated minimal facility. The distinction might be an important one, but should not be overemphasized. For example, production of any significance in the GS process requires that trays be level, foaming on trays be minimized, process temperatures and flow ratios be carefully controlled, there be good maintenance of piping insulation and steam tracing, power supply be reliable, etc.

(2) Related to the above is the question of hardware availability. That is, even with knowledge sufficient to write detailed specifications for all required equipment, it must still be built according to these - often stringent - specifications, and, in many cases, this poses a severe technological challenge. For example, in the GS process, the dilute nature of the feed, limited recovery from feed, and a design philosophy of minimizing the number of parallel units in a stage leads to the use of first stage high pressure towers and sieve trays which are amongst the largest in the

chemical industry. Moreover, high standards of quality control in tower construction is not only a matter of economics; the use of such large vessels containing correspondingly large inventories of toxic hydrogen sulphide gas makes the consequences of a major leak a matter of grave safety concern. Due to transportation limitations, these towers must be constructed on site by welding individual ring sections together, and then stress relieving the entire completed tower.²⁷ Another piece of equipment whose construction is highly non-trivial are the gas blowers which circulate the hydrogen sulfide through the towers. These are very large machines; e.g., typical specifications are: speed: 1860 rpm, motor horsepower: 7000, and capacity: 96×10^4 SCFM.²⁸ Moreover, they must be corrosion-resistant to wet H_2S and leakproof. In the case of both the monothermal and bithermal realizations of the hydrogen-ammonia exchange processes, careful attention must be paid to the integration of the heavy water plant with the syngas feed, so that ammonia production is not affected. As compared to the GS process, the problems posed by the corrosion and toxicity of the process fluids are not as great; neither are they entirely absent, however. For example, the catalyst needed to promote the exchange reaction, potassium amide, corrodes organic materials, and the use of high-purity hydrogen - to prevent explosive reaction with the amide - reduces the notch impact strength of high-grade steels. Considerations such as these, in the context of

the low temperature and high pressure characteristic of this process, imply that careful attention must be paid to materials and construction. In particular, the contacting equipment must be of special design, and the large, heavy towers must be constructed at the factory in one piece, and transported to the site.²⁹

(3) While pilot plants are useful in gaining familiarity with a process, there is a quantum jump involved in building a large production facility on the basis of pilot plant experience. Basically, this gap involves the technical and managerial expertise required to build and operate large industrial plants. For example, while a 100 ton per year GS plant may have three first-stage towers, each 4.4 m in diameter and weighing 285 tons which must be fabricated in the field, the first stage for a one-ton per year pilot plant could be a single tower approximately 0.6 m in diameter and correspondingly thinner and lighter, which could easily be built in one piece at the factory and transported to the site. The smaller plant volume also implies a smaller holdup of hydrogen sulfide, smaller and/or fewer trays, blowers, pumps, heat exchangers and other equipment, no requirement for siting near a nuclear power plant in order to utilize low-cost nuclear steam, less need for reliable back-up power and precise process control, easier maintenance, etc. Similar considerations would apply in scaling up from pilot plant realizations of other heavy water processes such as hydrogen-ammonia exchange

to large capacity commercial facilities. The former are certainly useful for gaining experience with the process - particularly in the area of materials testing - and also as a source of heavy water for: scientific research, inventory makeup in existing power reactors, and possibly the initial inventory of small heavy-water-moderated plutonium production reactors (The last issue is discussed in the next section.). However, the great difference in the number and variety of technical and managerial skills required makes construction and operation of large plants significantly more difficult. The manpower estimates for a 400 ton per year GS plant enumerated in Table 4 give an indication of the magnitude of the latter task.³⁰ In addition, a special initial operations group of approximately 15 professionals would be required for plant commissioning, and a staff of approximately 400 - including resident engineers, maintenance personnel, and control room operators - is needed for normal operations.³¹

In sum, the construction and operation of a hundred ton per year heavy water plant is a large industrial enterprise with demanding quality control requirements. Thus, it is not surprising that successful implementation of heavy water technology require the resolution of similar problems as arise in the introduction of nuclear power technology in developing countries. The following statement from a recent IAEA publication is particularly germane:

Table 4
Manpower Requirements for
the Construction of a 400 ton per year
GS Heavy Water Plant

Home Office

Engineering Design Group - ~ 150
including chemical, civil,
mechanical, electrical,
industrial engineers and
draftsmen

Project Management Group - ~ 50
including scheduling, cost
control, expediting and
procurement

Site

Management Group - ~ 100
including contractor
supervision and quality
assurance inspection

Labor Force - ~ 1500
including pipefitters,
welders, concrete pourers,
crane operators, insulators
and electricians

Experience has shown that the most serious problem which the developing countries have had to date have not been of the type which require additional manpower with advanced academic training in nuclear science and technology, but are rather of the type which require (a) manpower with practical experience and training in the technical, organization, contractual and administrative aspects of large projects and (b) technicians and skilled craft labour. ³²

(4) Regarding technology transfer from other industrial processes, experience in such areas as oil refining and hydrogen production via coal gassification for synthetic ammonia production would certainly be helpful, but there are also significant differences. For example, in fractional distillation of crude oil, the equipment is of much smaller scale since basically all the feed is utilized as product. Moreover, as compared with the GS process, tray correlations are different, and process control is less stringent. On the other hand, refinery desulphurization via catalytic extraction of organosulphur values from the oil as hydrogen sulphide does provide useful experience in working with H_2S . In general, a country which can build and operate a complete oil refinery or synthesis gas plant for ammonia production has a solid foundation which might make the construction of a large heavy water plant a feasible undertaking.

Of late, the realities of technology transfer are in danger of being lost in a cloud of North-South rhetoric. In particular, the North is being warned that it is in its best

interests to share its technological expertise on more favorable terms than hitherto since, at best, it is only a matter of time before the advanced developing countries catch up and then forge ahead. This view is widely shared, even by many astute "Northern" observers. For example, according to Harvey Brooks,

The most important fact about a new technology is the knowledge that it can work. With that knowledge any nation with a reasonably sophisticated technical capability of its own can duplicate a technological development more cheaply than its originator, even without technology transfer.³³

With regard to nuclear technologies such as heavy water production, we would reply: Yes, but. The heart of the matter is the phrase, "reasonably sophisticated technical capability." Does it include engineering "know-why" as well as "know-how"?--i.e., not only how to reproduce a known design, but also the ability to understand the rationale behind the original designer's choices so as to be able to modify that in the light of new operating conditions. Does it include an appreciation of the importance of quality assurance and quality consciousness among both craft labor and supervisory personnel? Does it include a management team with the skills needed to co-ordinate and direct the construction of a large project? Does it include an overall *esprit de corps* and dedication to getting the job done right?

Rhetoric aside, the heart of the matter is a keen understanding of the technical fundamentals and the availability of the requisite skills/attitudes, materials and facilities. Without these elements--some of which are technology-specific --the cost and time penalties involved in reproducing a foreign high technology will be prohibitive.

(continued)

3.4 Heavy Water for Production Reactors

To meet the moderator requirements of a production reactor which produces roughly 10 kg. of plutonium per year requires on the order of 15-20 tons of heavy water. This amount could be supplied by several years of operation of a small plant, or much more quickly via diversion from a large facility.

3.4.1 Small plants

As with the initial choice between graphite or heavy water moderation for the production reactor, the decision as to which process to use in a small plant would depend on the proliferation scenario. That is, for a country which has chosen natural uranium, heavy water reactors for power generation, construction of a pilot plant utilizing the GS process for primary extraction - and water distillation for finishing to reactor grade concentration - would be a logical way to gain familiarity with the GS process. As we have seen, working with the H_2S/H_2O system requires that careful attention be paid to toxicity and corrosion, but at this date, 27 years after the initial operation of the first GS plants in the U.S. and with GS pilot plants having been built in France, Sweden, Romania, India, the Soviet Union, and probably the People's Republic of China, this task is probably within the capability of those developing countries with a moderate industrial base and some expertise in chemical processing. On the other hand, if indigenous skills are modest, and there is no desire to pursue heavy

water production commercially on a large scale, it would be logical to use water distillation for both extraction and finishing. As compared with the Manhattan District water distillation plants, plants built today would use tower internals with greater capacity per unit volume to minimize total tower volume, and more efficient means of providing the required low temperature process heat. While the tower volume would still be large in comparison with that required for primary extraction via the GS process, this factor alone need not be decisive in ruling it out even if a desire for minimal "visibility" is an important criterion in the choice of a process. A recent publication devoted to the subject of clandestine production of small amounts of heavy water concluded that the likely combination to be used would be primary enrichment by electrolysis followed by ammonia distillation.³⁴ While it is true that modification of an existing electrolytic hydrogen plant can provide primary enrichment of heavy water at practically zero cost, the obvious caveat is that electrolytic hydrogen plants are rare, essentially restricted to those sites where cheap hydropower is available. Hydrogen is more commonly available as ammonia synthesis gas; unfortunately, this does not provide a similar opportunity for preinrichment; in fact - due to the water-gas reaction used to make hydrogen - the deuterium concentration is lower than normal, typically about 100 ppm. Also, if electrolytic hydrogen is available, it would be much more straightforward to bring the preenriched product up to reactor grade by water distillation than the

recommended technique of first converting it to ammonia, distilling the ammonia, cracking the product to obtain enriched deuterium, and finally burning this in pure oxygen to produce heavy water product.³⁵ It is interesting to note that the idea of using ammonia distillation with enriched feed from an electrolytic cascade was considered by India for its first heavy water plant at Nangal.³⁶ However, this scheme was rejected in favor of hydrogen distillation, and the only commercial application of the ammonia distillation process was at Marzingarbe where it was used from 1968-1972 to bring ammonia enriched to 2% deuterium via monothermal hydrogen - ammonia exchange to reactor grade product. However, in the two hydrogen-ammonia plants presently nearing completion in India which are technically derived from Marzingarbe, final enrichment is made in a second monothermal stage rather than by ammonia distillation. In the Indian plant utilizing bithermal hydrogen-ammonia exchange for primary extraction, the product is brought to reactor grade by first transferring the deuterium to water and then distilling it. In practice, of course, the process actually used in a clandestine facility would depend strongly on the "targets of opportunity". That is, the opportunity to secretly acquire key hardware and/or technical data might make attractive a process which would not be feasible in terms of indigenous capabilities alone. In this sense, the attitude towards export controls of the advanced industrialized countries will obviously

be a key factor in the spread of this technology. Of particular importance are the interpretations given the phrase "or major critical components thereof" in the definition of exported technology which triggers safeguards according to the London Suppliers Guidelines.

'Technology' means technical data in physical form designated by the supplying country as important to the design, construction, operation, or maintenance of enrichment, reprocessing, or heavy water production facilities or major critical components thereof, but excluding data available to the public, for example, in published books and periodicals, or that which has been made available internationally without restrictions upon its further dissemination.³⁷

3.4.2 Diversion from a large plant

Here the key concern is the presence and efficacy of safeguards. Although the London Club Guidelines include heavy water technology on the trigger list, heavy water plants have heretofore not been safeguarded by the IAEA. An obvious starting point would be to define the safeguards goal as that amount of heavy water which could moderate a reactor whose yearly plutonium production is on the order of 10 kg. This would be on the order of 15-20 tons of reactor-grade product; or correspondingly larger amounts of material of lower concentration. Safeguards for a heavy water plant would, of course, have to be tailored to the particular process involved. However, all would involve the safeguards triad: containment, surveillance, and materials accounting. In lieu of detailed analyses of particular processes,³⁸ we make the following general observations:

(1) Heavy water is a valuable material, process materials are often toxic, and plant shutdowns are costly in terms of lost production. (For example, even a short interruption of power, lasting minutes for the steam and only milliseconds for the electricity in a large GS plant, will result in complete shutdown and five days are required to regain equilibrium: equivalent to two to three days lost production.) This indicates that standard industrial practice would mandate stringent containment, surveillance and product accounting procedures, as well as periodic testing of equipment integrity, and sampling of process fluids as part of the normal operating procedure. Safeguard procedures could naturally build on this base.

(2) Virgin heavy water product contains only minute activity due to natural tritium, 5×10^{-7} Ci/Kg,³⁹ and in any case, the beta emission from this isotope would not penetrate the steel drums in which the product is stored. Thus, surveillance and containment of product could not take advantage of an inherent radiation signature. However, determination of deuterium abundance by active means; e.g., neutron interrogation, would seem feasible.

(3) A logical sub-block of the entire plant for materials accountancy would be the finishing unit, which as noted, is usually accomplished by water distillation. It is at this point; i.e., when the deuterium content is on the order of 1% or higher, that the heavy water becomes valuable since only a small amount of additional separative work (~1%) is required to bring it up to reactor grade. Fortunately, it is

also at this point that the concentrations are high enough and the flows low enough so that it should be feasible to take measurements accurate enough to insure that the limit of error in the materials unaccounted for (LEMUF) is within the safeguards goal.

(4) All safeguard schemes will have to take into account, to a varying degree, the proprietary nature of all of the production processes of commercial interest. As previously noted, basic information on all of these is available in the open literature, but detailed know-how is protected by patents and otherwise restricted. This problem, of course, is not unique to heavy water plants, but exists with other "sensitive technologies;" e.g., uranium enrichment.

3.5 Proprietary and Nonproliferation Constraints

There are two types of constraints on the transfer of heavy water production technology: proprietary and nonproliferation; we discuss each briefly in turn.

3.5.1 Proprietary

The situation with regard to patents on heavy water production processes has been aptly summarized by Silberring:

Most early patents for different processes or process steps are expired. A number of later patent applications are weak, since what is claimed has already been published earlier. Consequently, no serious legal obstacles exist to prevent the use of a number of processes and/or equipment by anybody.⁴⁰

Indeed, one comes away from even a short exposure to the relevant technical literature with the strong impression that the control of such companies as Lummus, Sulzer and Uhde over their heavy water technologies is much less a matter of patent protection than a mastery of the details of the particular processes. Moreover, because of the strong temptation for the competition to utilize an invention without paying the price--by grasping its essence and then modifying and improving or designing around it--high technology companies today tend to omit important details from their patent applications or take the risk of not filing at all. In the heavy water field, the "modifying and improving" method has been the basis for the (thus far) successful effort of AECL to establish a heavy water industry in Canada utilizing the GS process without further compensation to Jerome Spevack and the Deuterium Corporation beyond the \$3 million payment made

by the Province of Nova Scotia in 1966 for Spevack's interest in his Canadian subsidiary, Deuterium of Canada, Ltd., together with an exclusive license for use in the Province of Nova Scotia of Deuterium Corporation's Canadian patents and proprietary information relevant to the GS process. This agreement thus covers the Glace Bay and Port Hawkesbury GS plants in Nova Scotia, but not the Bruce heavy water plants in Ontario. The position of AECL with regard to the latter is that Canadian modifications and improvements to Spevack's original wartime patent, which was transferred from the U.S. AEC to AECL in 1959 pursuant to the patent exchange provisions (Article IX) of the 1955 U.S.-Canadian agreement for co-operation in the civil uses in atomic energy,⁴¹ establishes its own patent position on GS technology, in particular in Canada, but also in the U.S. and other countries. Spevack's control of GS technology in the U.S. is based on his numerous U.S. patents and a license agreement between Deuterium Corporation and the U.S. AEC which was part of a settlement reached in 1974 between the two parties of litigation initiated by Spevack for compensation for the use of his GS inventions by the AEC at its Savannah River and Dana plants⁴². The agreement allows the U.S. Government to continue to use Spevack's inventions for production of up to 250 tons of heavy water per year at the Savannah River plant. In addition, the Government has the option to convert its license to one for use of the invention in new government-owned plants, but limited

to the production of heavy water solely for use by the Government itself. Deuterium Corp. and Spevack receive the irrevocable royalty-free right to unlimited use of the inventions of all patents and patent applications owned or controlled by AEC for concentration of any isotopes by the dual-temperature method and/or for the production of heavy water. Although the battle between Spevack and AECL over heavy water patent rights in the U.S. and Canada continues⁴³, one has the impression that it is largely academic both for the reasons cited by Silberring and because the prospects for additional GS plants in these countries, particularly the U.S., are remote. The situation in other countries is more interesting. Spevack has taken out patents on his inventions in numerous foreign countries⁴⁴; many of these have now expired. Unfortunately (for him), India, the only other country besides Canada which has attempted to implement the GS process commercially, adopted quite early in the development of its nuclear program a highly protectionist policy towards the import of foreign nuclear technology which, in particular, precludes the issuance of patents for heavy water production processes. To the extent that other advanced developing countries become interested in heavy water production, it is quite probable that they would follow the Indian example. In this situation, the existence of patents on GS and other processes⁴⁵ will not be the limiting factor in their ability to implement them.

3.5.2 Nonproliferation

3.5.2.1 U.S. Controls

There are three types of controls on heavy water technology mandated by U.S. legislation. These are administered by the Department of Energy (DOE), the Nuclear Regulatory Commission (NRC), and the Commerce Department, and are described briefly below. Statutory authority for these controls derives from the Atomic Energy Act of 1954, as amended (most recently by the Nuclear Nonproliferation Act of 1978 (NNPA)), and the Export Administration Act.

3.5.2.1.1 Department of Energy

The authority for DOE controls stems from Section 57b of the Atomic Energy Act of 1954, as amended, which prohibits "persons" (i.e., individuals, corporations or other organizations) under U.S. jurisdiction from directly or indirectly engaging in the production of any special nuclear material outside the U.S. except pursuant to an intergovernmental agreement for cooperation or by a determination of the Secretary of Energy, with the concurrence of the Secretary of State, that such activity will not be inimical to the interests of the U.S. (The heavy water connection obviously stems from the indirect production of special nuclear materials; e.g., via heavy water-moderated production reactors.)

The implementing regulation for Section 57b is 10 CFR 810, Unclassified Activities in Foreign Atomic Energy Programs. Specifically, pursuant to para. 810.7(2) and para. 810.8,

specific authorization from the Department of Energy is required before engaging in any of the following activities outside of the United States:

(i) Designing or assisting in the design of facilities for...the production of heavy water ... or especially designed equipment or components ...; or

(ii) Constructing, fabricating, or operating such facilities; or

(iii) Constructing, fabricating, or furnishing equipment or components especially designed for use in such facilities; or

(iv) Training foreign personnel in the design, construction, fabrication, or operation of such facilities or equipment or components especially designed therefore [sic]; or

(v) Furnishing information not available to the public in published form for use in the design, construction, fabrication or operation of such facilities or equipment or components especially designed therefore; ...

Indeed, according to paras. 810.7(b)(3) and (4), the only activities involving heavy water production technology transfer which do not require prior authorization by the Department of Energy are limited to: participation in meetings or conferences sponsored by educational institutions, laboratories, scientific or technical organizations; international conferences held under the auspices of a nation or a group of nations; or exchange programs approved by the Secretary of State; or the furnishing of information which is available to the public in published form.

In the NNPA, a new category of information is defined: "sensitive nuclear technology." According to Sec. 4(a)(6) of

the Act, this means

any information (including information incorporated in a production or utilization facility or important component part thereof) which is not available to the public and which is important to the design, construction, fabrication, operation or maintenance of a uranium enrichment or nuclear fuel reprocessing facility or a facility for the production of heavy water, but shall not include Restricted Data controlled pursuant to chapter 12 of the 1954 Act; ...

According to the legislative history of the Act, the task of deciding what information falls under this definition was viewed by the drafters of this legislation as a natural extension of the duties of the DOE Office of Classification; i.e.: "The normal processes of classification and declassification by the Department of Energy [DOE] will determine what information will be controlled by this definition."

Although this does not constitute a statutory requirement, the task of explicitly defining "sensitive nuclear technology" has been undertaken by the DOE Division of International Security Affairs (ISA), and will be published as amendments to the relevant portions of paragraph 810.7(2).

3.5.2.1.2 Nuclear Regulatory Commission

The NRC is responsible for licensing the export of nuclear facilities and materials, and specially designed parts and components therefor. The implementing regulation for this requirement is 10 CFR 110, Export and Import of Nuclear Equipment and Materials. In particular, paragraph (3) of appendix A of this regulation lists as items subject to NRC licensing authority:

Plants for the production of heavy water, deuterium and deuterium compounds, and specially designed or prepared parts and components therefor (export only).

With regard to the above, it is important to note that the scope of export control under NRC's export licensing jurisdiction which is mandated by the NNPA goes beyond "specially designed or prepared parts and components therefor." According to Sec. 402(b):

In addition to other requirements of law, no major critical components of any uranium enrichment, nuclear fuel reprocessing, or heavy water production facility shall be exported under any agreement for cooperation ... unless such agreement for cooperation specifically designates such components as items to be exported pursuant to the agreement for cooperation. For purposes of this subsection, the term "major critical component" means any component part or group of component parts which the President determines to be essential to the operation of a complete uranium enrichment, nuclear fuel reprocessing, or heavy water production facility.

3.5.2.1.3 Department of Commerce

According to Department of Commerce Regulation para. 378.4(b), a validated license is required for export to all destinations, including Canada, of any commodity where the exporter knows, or has reason to know that the export will be used in heavy water production. This regulation reinforces the NRC and DOE regulations outlined above, which are the primary U.S. mechanisms for control of heavy water production technology and equipment.

The sum and substance of the above is that the U.S. Government is now authorized by domestic law to take into account

what might be called "the big picture" when considering the export of heavy water production equipment and technology. That is, not only are complete plants covered, but also, in decreasing order of specificity, specially designed or prepared parts and components therefor, major critical components, or indeed any commodity. However, the latter two categories are obviously grey areas, and a strong commitment to the importance of export controls in U.S. government agencies and among U.S. "persons" will be needed to attain compliance with both the spirit and the letter of the law.

3.5.2.1.4 Criteria and Administration

According to the NNPA, heavy water production equipment and technology cannot be exported unless certain criteria are satisfied. In particular, Sec. 306 of the NNPA mandates that no export of sensitive nuclear technology to non-nuclear weapons states shall be made "unless IAEA safeguards are maintained with respect to all peaceful nuclear activities in, under the jurisdiction of, or carried out under the control of such state at the time of the export."

Export license requests relevant to heavy water production are reviewed by the administering agencies, in consultation with the Arms Control and Disarmament Agency (ACDA) and the Departments of State and Defense. To facilitate this review and to comply with Sec. 304 of the NNPA, a formal interagency Subgroup on Nuclear Export Coordination has been established. The Subgroup reviews any license application for the export or

transfer of nuclear materials, equipment or technology which any of its member agencies consider to be sensitive or to raise significant nonproliferation policy questions. The Subgroup consists of representatives from the Departments of State, Energy, Defense, Commerce, the NRC and ACDA.

The procedure for the disposition of the export license applications depends on the agency with which they originate. Both NRC and DOE applications are reviewed by the other agencies of the Subgroup who forward their recommendations to the originating agency. In the former case the Commissioners of the NRC, and in the latter the Secretary of Energy, then makes an independent determination in approving or denying the license. If either the NRC or the DOE fail to issue a license because of their determination that the export criteria set forth in the NNPA have not been met, the President is empowered under Sec. 306 of this law to authorize the proposed export if he determines that failure to approve it "would be seriously prejudiced to the achievement of U.S. nonproliferation objectives or otherwise jeopardize the common defense and security."

The Department of Commerce refers to the DOE, for technical and policy review, proposed exports where the exporter knows or has reason to know that the commodities or technology to be exported are for use in heavy water production. The vast majority of these cases are cleared by the DOE and a license issued by Commerce without further interagency consideration. A small minority of cases are referred to the

Subgroup on Nuclear Export Coordination where they are analyzed and a decision is made either to approve, disapprove, return without action, seek additional information or obtain governmental assurances.

From a historical perspective, the lumping of heavy water production with uranium enrichment and fuel reprocessing as sensitive technologies in the NNPA represents a swing in the pendulum of concern towards a less benign view of this technology in U.S. government bureaucracies and the Congress. However, it would be misleading to characterize the preceding Atoms for Peace era, from roughly 1955 to the explosion of the Indian PNE in May 1974, as one of complete naivete in the U.S. concerning the proliferation implications of heavy water and heavy water production technology. The U.S. reacted with growing concern in the 1960s to evidence of Indian ambitions to obtain an independent natural uranium fuel cycle capability without safeguards. For example, while heavy water was supplied for the CIRUS research reactor, and via Canada, for the RAPP I power reactor, the attempt by India in 1967 to obtain GS production technology from a U.S. company without safeguards was refused.⁴⁶ In a similar vein, Indian opportunities to gain "hands-on" experience with this technology by sending technical personnel to the Savannah River plant were increasingly curtailed.⁴⁷ We do not argue that more could not have been done in this regard; only that nonproliferation was not completely ignored in policy decisions. Thus, the decision in 1972 to allow a U.S. company to negotiate

with Romania for the construction of a GS plant in that country⁴⁶ had obvious geopolitical overtones. However, the fact that Romania is a party to the Non-Proliferation Treaty (NPT) and evidenced no weapons ambitions was also an important consideration.

According to the NNPA, adherence to the NPT or acceptance of the principle of full-scope safeguards is a necessary condition for nuclear commerce with the U.S. Regarding the specific question of the transfer of heavy water technology, the current policy of the Carter Administration is to regard these conditions as necessary but not sufficient. This policy is viewed with misgivings in Canada because it lumps a technology which does not supply a direct route to weapons with uranium enrichment and fuel reprocessing which do,⁴⁸ hence it will be much more difficult to obtain a consensus among other suppliers of this technology on any transfer requirements which go beyond full-scope safeguards. Indeed, as recent events in Argentina bear witness²⁵, even this may prove elusive. This brings us to the subject of

(2) International Controls.

In contrast to the situation with other energy sources such as coal, the U.S. government has always played an integral role in the development and control of atomic energy. Not surprisingly, the same is true in all other countries where civilian and/or military applications of atomic energy have been developed. However, the domestic statutory requirements and implementing regulations regarding criteria and procedures for nuclear transfers in many countries have until

recently been rather vague, and past policy--not necessarily practice--is in most cases better gleaned from a scrutiny of published bilateral agreements for co-operation in the peaceful uses of atomic energy.⁴⁹

The first public attempt to introduce a degree of international uniformity in the safeguard requirements for nuclear transfers was the guidelines published under the auspices of the IAEA as INFCIRC/209 in September 1974. This document was the work of a group of nuclear exporting countries, the 20-member Zangger Committee, which was set up to interpret Section III of the NPT. In particular, Memorandum B of INFCIRC/209 contains a list of materials and equipment which should trigger the application of IAEA safeguards to the nuclear material produced, processed, or used in the facility for which the items are supplied. From a nonproliferation perspective, the most significant deficiencies of this "trigger" list with regard to heavy water were the failure to:

- (1) require that the transfer of sensitive technology, as well as materials and equipment, trigger safeguards, including safeguards on replications and retransfers of the technology;
- (2) require safeguards not only on heavy water itself, but also on heavy water production plants and equipment.

Moreover, France, not being a member of the NPT, was not a party to INFCIRC/209. Following the Indian PNE in May 1974,

and subsequent disclosures that France and West Germany planned to sell sensitive facilities to Pakistan and Brazil, respectively, the U.S., prodded by Canada, initiated a diplomatic campaign to raise the world's proliferation consciousness, and in particular to repair the deficiencies of INFCIRC/209. (India, for example, had used the omission of heavy water plant equipment from the trigger list to obtain, in Western Europe, items for their GS heavy water plant at Kota which were embargoed by Canada after the Indian test.) To this end, the "London Club" of nuclear exporters was convened in April 1975 amid great secrecy. Their deliberations have been much commented on;⁵⁰ where we only note that their final document, published by the IAEA as INFCIRC/254 in February 1978, only corrects the above-noted deficiencies in the Zanger trigger list, but also calls on the suppliers to "exercise restraint" on the transfer of sensitive facilities and technology (including heavy water production equipment and technology (including heavy water production equipment and technology) and, with regard to the latter, specifically lists three generic technologies--exchange, electrolysis, and hydrogen distillation--transfer of which would trigger safeguards. Although the London Club guidelines are a significant improvement on INFCIRC/209,⁵¹ they are the fruit of a fragile consensus among states with varying political and commercial perspectives, and leave open various loose ends.

In particular,

(1) The suppliers have agreed only to exercise restraint on the transfer of sensitive technology and facilities rather than to prohibit them completely. Especially with regard to heavy water production, recent events indicate that Switzerland, and perhaps other countries, are not prepared to go beyond the guideline provisions for facility rather than full-scope safeguards as a condition for transfer, much less embargo these facilities entirely.

(2) The membership of the Club does not extend to countries which may in the future be in a position to engage in significant transfer of nuclear technology and equipment. In the heavy water field, India in particular may attain such a capability within the coming decade. We also note that Argentina is going ahead with construction of an indigenous GS pilot plant in spite of its recent agreement with the Swiss firm Sulzer for the construction of a 250 ton per year hydrogen-ammonia exchange transfer process plant in Argentina.⁵¹ Apparently, the Argentine strategy, like that of India, is "not to put all its heavy water eggs in one basket."

(3) Part B, para. (2) of the guidelines seems to close the "specially designed and prepared" equipment loophole.

That is, it states that

if a supplier nation should transfer in the aggregate a significant fraction of the items essential to the operation of such a facility, together with the knowhow for construction and operation of that facility, that transfer should be deemed to be a transfer of "facilities of major critical components thereof."

However, like the similar provisions in the U.S. NNPA, the word "essential" is obviously open to varying interpretations which may be tailored to accommodate different perceptions of the importance of adherence to nonproliferation norms.

In sum, INFCIRC/254 may well represent the high-water mark in international attempts to control the spread of sensitive technologies. Specifically, with regard to heavy water production, it is problematic whether a consensus can be reached on a requirement of full-scope safeguards as a condition for transfer. Given the problems involved in implementing this technology, for the foreseeable future this represents the major cause for concern, rather than the prospect of spread via the efforts of countries outside the London Club.

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