A Model for the Evolution of Gas Pressure and Composition During Sealed Storage of Metallic Uranium Fuel

by

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Lisa Stiles Shell

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Requirements for the Degree of
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Abstract

A model has been developed to predict the pressure generation and concentrations of oxygen and hydrogen gas in a Multi-Canister Overpack (MCO) containing metallic uranium N Reactor fuel during staging, drying, conditioning, and storage at the Hanford Site. Associated uncertainties with each parameter were also calculated.

The mechanisms that were included in the analysis were:

- **Uranium Corrosion** in which oxygen is consumed, or water is consumed and hydrogen is produced,
- **Uranium Hydriding** in which hydrogen is consumed, and
- **Radiolysis** in which water is consumed and hydrogen and oxygen are produced.

The model characterizes the evolution of gases inside the MCO for the three regimes in which the fuel will be staged or stored. Prior to treatment the fuel will be immersed in water in the MCO. Proceeding the first treatment step, the MCO will contain no free water, but water vapor will contribute to a moist atmosphere. After the last treatment step, the inside of the MCO will be dry with only tightly-bound chemisorbed water remaining associated with sludge that will be present with the fuel.

The model shows that for likely conditions inside the MCO, the container will not pressurize during its expected service life. The model also shows the effects that varying parameters have on the final pressure.

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1. Introduction

The Hanford Site is located in south central Washington state and is part of the Department of Energy (DOE) weapons complex. It was here that the first plutonium (Pu) was produced in a reactor.

The history of N Reactor and the K Basins, where spent fuel from the N Reactor is stored, is summarized briefly in the report "Integrated Process Strategy for K Basins Spent Nuclear Fuel" (1995). N Reactor operated from the 1960's to 1987 at the Hanford Site in Washington State. Its mission was to produce plutonium for the U.S. weapons program as well as to provide power to the Washington Public Power Supply System. Prior to 1978, spent nuclear fuel (SNF) from N Reactor was processed to separate actinides and fission products and to extract plutonium at the PUREX (Plutonium-Uranium Extraction) plant. At this time, SNF was stored in K Basins (water-filled pools located in the K area of the site) while PUREX was refurbished and when the modifications were complete, much of the N Reactor fuel was reprocessed. However, due to the end of the Cold War and greater attention given to proliferation concerns, PUREX was deactivated in 1992, leaving approximately 2100 metric tons of N Reactor SNF stored under water in the K Basins ("Integrated Process Strategy for K Basins Spent Nuclear Fuel," 1995) (Abrefah, 1995).

As a result, a significant portion of the more than 100,000 individual assemblies are degraded and corrosion will continue as long as the fuel is stored under water ("Dry Storage of N Reactor Fuel," 1994). In the K East Basin, the fuel assemblies are stored in open aluminum or stainless steel canisters. Visual inspections reveal that a significant portion of the fuel is corroded and a layer of sludge, up to 30 cm deep, has been deposited

on the floor of the basin and in the canisters. The canisters in K West Basin are closed so visual inspection of the fuel is impossible. Also, it is not known if the water inside the canisters in K West is oxygen depleted. Both basins were constructed in the 1950's, do not meet current construction requirements, and are located less than ¼ mile from the Columbia river ("Integrated Process Strategy for K Basins Spent Nuclear Fuel", 1995) (Abrefah, 1995).

The majority of the fuel has now been in storage in excess of 10 years. Some of the fuel has been stored in this configuration for more than 20 years. The Department of Energy is pursuing a path forward to remove the fuel canisters from the K Basins into sealed containers called Multi-Canister Overpacks (MCOs), treat it at a facility to be constructed on site, and store the MCO's containing the dry fuel in tubes inside the Canister Storage Building (CSB) until a permanent repository is available ("Integrated Process Strategy for K Basins Spent Nuclear Fuel," 1995). To accomplish this goal, the fuel must be treated in a way that renders it stable for at least fifty years. To eliminate pressure generation and ignition hazards, a process is being developed to thoroughly dry the fuel by removing free, bound, and chemisorbed water to the maximum extent possible. In addition, oxygen may be admitted in the final treatment step to condition the surface of the fuel by deposition of an oxide film.

The process being developed is based on a French process for similar fuel ("Dry Storage of N Reactor Fuel," 1994). First, fuel will be loaded into baskets and placed inside a Multi-Canister Overpack (MCO). After transport, the MCO will be drained of free water, inerted, and the fuel will be dried under vacuum. Next, the MCO will be transferred to the Canister Storage Building (CSB) and undergo hot conditioning which is

intended to remove much of the bound and chemisorbed water. Last, the MCO will be backfilled with an inert gas, sealed, and inserted into a storage tube at the CSB ("Management of Spent Nuclear Fuel from the K Basins at the Hanford Site, Richland, Washington," 1996).

The purpose of this thesis is to model the treatment process including various phenomena that contribute to pressure buildup, and hydrogen and oxygen generation. The final uncertainties of these values are also calculated. A Monte Carlo simulation was run to verify the results of the model. As many physical parameters have large uncertainties associated with them, it is intended that the model be modified as better data is acquired.

The model runs on Powerstation FORTRAN 4.0 for Windows NT.

2. Background

The MCOs will be loaded underwater at the K Basins and transported full of water to the Cold Vacuum Drying facility (CVD). Figure 1 shows a schematic of the treatment process. After draining and under vacuum, the inside temperature of the MCO will be raised to 100°C for no more than 10 hours. The MCO then will be transported to the CSB where hot conditioning will take place. The temperature inside the MCO will be raised to 300°C to remove chemisorbed water and waters of hydration from the fuel and sludge. This step is expected to take about 25 hours. At this point, the temperature may be lowered to 150°C and an argon and oxygen mixture admitted to form an oxide film on the fuel surface ("Dry Storage of N Reactor Fuel," 1994).

Figure 2 shows the treatment steps and phenomena that occur as the fuel-filled MCO moves from underwater storage in the K Basin to final dry storage at the CSB. Corrosion, radiolysis, and heat transfer are phenomena that must be examined at each step. Dehydration is of importance during CVD and HCS and must be considered in the staging step in between. Hydriding and dehydriding of uranium must be considered during all steps proceeding the initial staging step.

2.1 Previous Work

The initial impetus to develop a process similar to one of French design was a report prepared for the DOE by the Independent Technical Assessment team (ITA) entitled "Dry Storage of N Reactor Fuel Independent Technical Assessment." The ITA team examined related literature and the French treatment process and performed preliminary calculations for the feasibility study. The conclusion therein is that dry storage of N Reactor fuel using a treatment process as described above would be "environmentally benign, desirable, and feasible" ("Dry Storage of N Reactor Fuel," 1994).

In developing the treatment system, the DOE operating contractor, first Westinghouse Hanford Company, now Fluor-Daniel, Inc, has generated reports addressing fuel characterization and choice of models for various phenomena. Several of the most recent reports are listed in Table 1. A more complete listing of background literature is contained in the bibliography.

Table 1: Reports of Previous Work

Source	Title	Comments
Pajunen, A.L.	Evaluation of Radiolytic Gas Generation from Water Disassociation in a Multicanister Overpack	Recommends correlations to determine G values for oxygen and hydrogen and compares with literature data
Muhlestein, L.D, and M.R. Allen	Cumulative Oxygen Production in a Multi-Canister Overpack as a Function of Time	Calculates the expected cumulative inventory of oxygen in an MCO during storage.
Abrefah, J., et.al.	Reaction Rate Constant for Dry Air Oxidation of N Reactor Fuel	Establishes reaction rate constants for dry air or oxygen oxidation of the fuel
Thurman, Cooper D., A. Burt Johnson	Spent Nuclear Fuel Project Surface Area Estimates for N-Reactor Fuel in the K East Basin	Estimates the exposed uranium surface area of individual fuel assemblies

2.2 Corrosion

Generally, Zircaloy is an excellent cladding material and protects the uranium metal from aqueous corrosion. However, previous handling during reactor discharge and subsequent movement of the fuel produced mechanical defects in some of the cladding leaving uranium metal exposed to the environment. Uranium metal reacts with water or oxygen to produce oxides or hydrides ("Dry Storage of N Reactor Fuel," 1994). Oxidation of uranium is an exothermic process. The most general reactions are

(1) $U + 2H_2O \rightarrow UO_2 + 2H_2 \qquad \Delta G = -133.3 \frac{kcal}{mole}$ at 25°C

(2) $U + O_2 \rightarrow UO_2 \qquad \Delta G = -246.6 \frac{kcal}{mole}$ at 25°C

The hydrogen generated by equation one may then be available for hydride formation. Volumetric expansion results from the above reaction due to the low density of the oxide

as compared to that of metallic uranium. Other species stable at 25°C and neutral pH include U_4O_9, U_3O_8, and UO_3*H_2O. In addition, sub- and super- stoichiometric oxide forms are known to exist in the sludge and the oxide layer on the fuel surface. These forms are transient and difficult to test for material properties (Abrefah, 1995).

Experiments confirm that in dry air a fine, black, hyperstoichiometric (UO_{2+x}) oxide is produced which has an average oxygen/uranium ratio of 2.09 (Colmenares, 1984). In a dry oxygen atmosphere, oxygen is absorbed to the metal surface and forms an oxide film. As the film grows, the oxidation rate is controlled by the diffusion of oxygen interstitial ions through the film. Large internal stresses are generated as the surface oxide thickens and cracks. Below about 75μm thickness the oxide is protective. Thicker films begin to fracture due to compressive stresses. When the compressive yield stress, 1.93 GPa, is exceeded, the oxide spalls off (Robinson and Thomas, 1995).

When water is present, a similar film initially forms on the surface of the uranium metal. Then, water that is adsorbed to the oxide surface dissociates into hydroxyl and hydrogen ions. Hydroxyl ions may then diffuse through the oxide layer and react with uranium metal to form oxide and produce hydrogen. The hydrogen atoms can then diffuse through the oxide film or remain in the uranium. Hydrogen atoms at the surface may recombine to form hydrogen gas. When sufficient hydride is accumulated under the oxide film, hydrides may form (Colmenares, 1984). Hydrogen accumulation under the film is more likely than diffusion through the oxide film to the surface (Robinson and Thomas, 1995). Colmenares' research indicates that up to 100°C, uranium metal in the presence of water corrodes to become a fine black oxide, and at about 150°C exfoliation begins in sheets approximately 1μm thick. This process proceeds with laminae fracturing

first laterally then normal to the surface. Exfoliation can also be initiated by hydride formation and expansion (Colmenares, 1984).

The effect of oxygen concentration in water is evident in the stoichiometry of the oxides formed (Robinson and Thomas, 1995). Corrosion of uranium at ambient temperature in aerated water generally produces UO_2 and hyperstoichiometric oxides of the form UO_{2+x}. Corrosion in deaerated water produces UO_3*nH_2O and pitting attack of the uranium metal. In addition, the current density between aerated and deaerated cells has been observed to be near 50 $\mu amps/cm^2$ (Robinson and Thomas, 1995).

Corrosion of uranium will occur throughout the treatment process and in storage if oxygen is present. The reaction rates of uranium are usually calculated as weight gain per unit area per unit time. Therefore, to calculate the amount of uranium corroded, oxygen consumed, or hydrogen produced, the amount surface area participating in the reaction must be known. Currently, the amount of exposed uranium in each MCO that will participate in corrosion is highly uncertain. From visual inspections, estimated values range from 3000 cm^2 per assembly to 60000 cm^2 per assembly (Pajunen, 1997). Furthermore, the distribution of surface area values is not Gaussian. To calculate standard deviations of results and run the Monte Carlo simulation, a distribution representing the area values is required.

2.3 Radiolysis

Radiolytic disassociation of water produces over ninety different chemical species for which g-values give the number of atoms or molecules produced per 100 eV of energy deposition (Spinks and Woods, 1964). Most of the chemical species are short-lived chemical radicals with lifetimes of about 10^{-9} seconds. The long-lived products of

concern to this model are H_2 and O_2. G-values differ with type of radiation and with absorbing media. This characteristic of radiolysis makes calculation of overall g-values for hydrogen and oxygen production complicated (Spinks and Woods, 1964).

In addition to g-values, the amount of water available for radiolysis must be known. The sources of water in the MCO include free water, bound water, and chemisorbed water. Bound and chemisorbed water contained in the sludge are significant sources of water but are difficult to quantify. The amount of sludge in each MCO will range from nearly zero to over one hundred kilograms with an average around twelve kilograms ("Particulate Inventory in a MCO," 1997). Like fuel surface area, this distribution is not Gaussian. Furthermore, the composition of the sludge is needed to determine water inventory. Preliminary tests show that the fraction of water in the sludge before drying and conditioning is about 10% (Pajunen, 1997).

2.4 Dehydration

At stages of the process where the MCO is no longer filled with water, dehydration can occur at specific temperatures and pressures. Dehydration releases bound and water and makes it available for other reactions. Dehydration was not modeled for this thesis but can be added in later revisions. For this model, bound and chemisorbed water is released by radiolysis only. Since all of the bound water is expected to be removed during the HCS stage, this will not significantly affect the results.

2.5 Hydrides

Uranium hydride may be formed at ambient temperatures by the reaction of metal and hydrogen,

(3) $2U + 3H_2 \rightarrow 2UH_3 \qquad \Delta G = -17.9 \frac{kcal}{mole}$ at 25°C

The hydride may be decomposed in oxygen and water by the reactions

(4) $2UH_3 + 2O_2 \rightarrow 2UO_2 + 3H_2$ $\Delta G = -229.3 \frac{kcal}{mole}$ at 25°C

(5) $2UH_3 + 4H_2O \rightarrow 2UO_2 + 7H_2$ $\Delta G = -115.9 \frac{kcal}{mole}$ at 25°C

(Robinson and Thomas, 1995). The relationship between hydrogen overpressure in equilibrium with the hydride is such that uranium hydride is expected to be stable in air or mild vacuum. Though the hydride is not as stable as the oxide, small amounts can remain occluded in metal or oxide film ("Dry Storage of N Reactor Fuel," 1994).

The study of the kinetics and mechanisms of hydride formation is limited. Powell, Bloch, and Mintz conducted hydriding experiments in hydrogen gas (Robinson and Thomas, 1995). Their experiments, and others, have shown that uranium hydride follows a characteristic growth curve, which begins with a significant induction period, followed by a linear weight gain with time and ending with saturation and completion of the reaction. Nucleation then, appears to occur slowly (Bloch and Mintz, 1980). This behavior was explained by assuming that nucleation of hydride occurs beneath discontinuities and growth faults in the oxide layer and produces pitting attack. The growth of the hydride causes disruption and spalling of the oxide layer producing a larger reaction area and corresponding to the transition to linear weight gain with time. Additional experiments by Bloch and Mintz show that oxygen decreases the frequency of initiation events (Bloch and Mintz, 1980).

It has been well documented that uranium hydride can ignite spontaneously when suddenly exposed to air by the reaction

(6) $4UH_3 + 7O_2 \rightarrow 4UO_2 + 6H_2O$ $\Delta G = -508 \frac{kcal}{mole}$ at 25°C

(Robinson and Thomas, 1995). An ignition event of particular interest occurred at the Lawrence Livermore National Laboratory and is described in the article, "Regarding the the Chemistry of Metallic Uranium Stored in Steel," (Wood, et. al., 1994). An unexpected release of energy occurred when a sealed steel drum containing co-extruded, iron-clad, depleted-uranium was opened. The lid was tightly sealed, and while it was being loosened, the lid was propelled into the air and flames escaped the container. The analysts believe that while the lid was being loosened, air entered the drum and supported ignition of uranium hydride that in turn ignited hydrogen gas. The resulting analysis of drum content revealed that the gas inside the drum was 75% nitrogen and 25% hydrogen. Any oxygen that was present when the drum was originally sealed was consumed in the corrosion process. After oxygen was depleted, corrosion of uranium occurred which produced the hydrogen gas and caused hydriding of uranium metal. The water available in the drum was contained in the plastic liner, wood cross brace, plywood spacers, cardboard tubes, and silica gel.

3. Theoretical Modeling

3.1 Corrosion

Investigations of the corrosion of uranium in oxidizing atmospheres have been conducted since separation of the metal was achieved (Pearce, 1989). These atmospheres include dry air or oxygen, moist air, and water immersion. A significant body of data has been compiled for oxidation at temperatures below 300°C. Much of this data was collected by Pearce in "A Review of Reaction of Unirradiated Uranium in Gaseous Atmospheres" (1989).

3.1.1 Dry Air or Oxygen

In a dry air or oxygen atmosphere, oxygen is consumed and uranium oxide is produced. The most general reaction is

$$U + O_2 \rightarrow UO_2$$

Numerous other sub- and super- stoichiometric compounds have been identified, but the dioxide form is the only one that is considered for this thesis (Robinson and Thomas, 1995). Generally, oxidation rates are reported as weight gain per unit area per unit time. Near room temperature, oxidation rates appear to fit a logarithmic form such as

(7) $$R = k_1 \cdot \log(at + t_o)$$

or

(8) $$\frac{1}{R} = A - k_2 \cdot \log t$$

for which R is the weight gain per unit area, t is time, k is a reaction rate constant, and A, a, and t_o are other environment specific constants (Pearce, 1989). In experiments with sufficiently sensitive measuring techniques, parabolic rates have also been observed as shown by

(9) $$R = k_p \cdot t^{\frac{1}{2}}$$

In addition, linear oxidation rates described by

(10) $$R = k_l \cdot t + A$$

may be observed proceeding deposition of a protective oxide layer on the surface of the metal. Deposition generally takes place at temperatures greater than 100°C. The growth of an oxide layer slows the diffusion of atoms to the reaction surface (Pearce, 1989).

At very high temperatures, a continuously increasing reaction rate is observed. It is in this region that uranium can ignite (Pearce, 1989). For the atmospheres considered in this thesis, temperatures will not exceed 300°C.

Figure 3 shows data points extracted from plots provided by Pearce's report for uranium corrosion in dry air or oxygen at temperatures less than 300°C. This data was compiled from various authors and raw data was not available. Pearce reports a rate equation of

$$(11) \qquad \log k_l = 8.9464 - \frac{4638.2}{T}$$

where k_l is the reaction rate in $\frac{mg}{cm^2 \cdot hr}$ and T is the temperature in degrees Kelvin. However, unweighted linear extrapolation of the data yields the equation,

$$(12) \qquad \log k_l = 6.957 - \frac{3609}{T}$$

Similarly, data from French experiments reported in "Reaction Rate Constant for Dry Air Oxidation of N Reactor Fuel" (Abrefah, et. al., 1997) yields the expression,

$$(13) \qquad \log k_l = 7.223 - \frac{3745}{T}$$

Pearce does not report a weighting scheme with the data so one must assume that some of the data points were rejected from his extrapolation, or other points were included that did not appear on the plot.

3.1.2 Water Vapor

The reaction that takes place in water vapor depends on the partial pressure of the oxygen. Below about 10 Pa uranium reacts with water by equation one

$$U + 2H_2O \rightarrow UO_2 + 2H_2$$

("Dry Storage of N Reactor Fuel," 1994). Figure 4 shows the compiled data given by Pearce from experiments oxidizing uranium in water vapor with pressures between 0.006 and 101 kPa. Pearce gives a reaction rate equation,

$$(14) \quad \log\left(\frac{R}{P^{\frac{1}{2}}}\right) = 4.33 - \frac{2144}{T}$$

where R is the reaction rate in $\frac{mg}{cm^2 \cdot hr}$ and P is the water vapor pressure in kPa.

Extracting the data and performing a linear regression yields the equation

$$(15) \quad \log\left(\frac{R}{P^{\frac{1}{2}}}\right) = 4.59 - \frac{2274}{T}$$

When the oxygen partial pressure exceeds 10 Pa then uranium reacts directly with oxygen by equation two

$$U + O_2 \rightarrow UO_2$$

With 100% relative humidity and temperatures less than 100°C, Pearce reports a reaction rate equation,

$$(16) \quad \log k = 8.333 - \frac{3730}{T}$$

The data from Pearce was extracted and is shown in Figure 5. Linear regression on this data yielded the equation,

$$(17) \quad \log k = 8.541 - \frac{3797}{T}$$

For regimes in which the relative humidity is less than 100% and the temperature is less than 192°C, Pearce reports a different reaction rate,

(18) $$\log k = 10.566 - \frac{4990}{T} + 0.3 \log P$$

where P is the pressure in kPa. The data corresponding to this equation is shown in Figure 6. Linear regression of the data gives the equation

(19) $$\log k = 7.499 - \frac{3720}{T} + 0.3 \log P$$

When the temperature exceeds 192°C, Pearce reports a reaction rate equation,

(20) $$\log k = 6.1931 - \frac{2963}{T} + 0.3 \log P$$

Data for this equation could not be extracted.

3.1.3 Water Immersion

Like the vapor reaction, the reaction that takes place when uranium is immersed in water depends on the amount of dissolved oxygen in the water. However, there is no pressure dependence in the equations which are

(21) $$\log k = 4.33 - \frac{2144}{T}$$

for reactions without oxygen and

(22) $$\log k = 8.33 - \frac{3730}{T}$$

in oxygen-rich atmospheres.

3.2 Radiolysis

Radiolysis of free water in the MCO is by gamma radiation only due to the short range of alpha and beta particles. The g-value of hydrogen used from free water is 1.0 $\frac{H_2 \text{molecule}}{100 \text{ev}}$ which converts to 1.04e-7 $\frac{\text{moles } H_2}{\text{Watt} \cdot \text{s}}$. To determine the hydrogen

produced, the amount of energy deposited into the water must be determined. For this model, it was assumed that 1% of the decay power is deposited outside the fuel. In the immersed case, 100% of the power deposited outside the fuel is absorbed by the water. For the vapor case, the percentage should be adjusted according to the density of the vapor.

The g-value of hydrogen for radiolysis of water contained in sludge was set to 0.001 $\frac{H_2 molecule}{100\,ev}$ which converts to 1.04e-10 $\frac{Moles\,H_2}{Watt \cdot s}$. However the energy absorbed by the sludge must be calculated differently. The density of sludge is 4 $\frac{g}{cm^3}$ and contains mostly UO_2. The density of U is 19 $\frac{g}{cm^3}$ so that in the sludge there are 0.881 kgs of uranium per kg of sludge. The amount of uranium per fuel element is 23.43 kgs and there are 270 elements per MCO so that the total amount of uranium in each in MCO is 6326 kgs. Therefore, the fraction of uranium that has been removed from the fuel element is 0.000139 and is the fraction of decay heat-generating fission products that are contained in the sludge. In addition, this number is reduced by 30% due to the dissolution of strontium and cesium in water (Olander, 1976). Finally, the fraction of decay heat that is deposited to the sludge is 0.0000975. To determine the amount of hydrogen generated,

(23) $$H_2\ produced\ (moles) = 0.0000975 * g\ (\frac{Moles\ H_2}{Watt \cdot s}) * Decay\ power\ (W) * time(s)$$

3.3 Hydrides

Limited studies of the formation of uranium hydride from uranium metal pieces and powders have been conducted. For hydriding to occur, the partial pressure of hydrogen must exceed the equilibrium partial pressure that is calculated to be

$$(24) \quad \log P = \frac{-4500}{T} + 9.28$$

where P is the pressure in torr and T is the temperature in °K (Spedding, et. al., 1949). To predict the reaction rate of uranium hydride, Kirkpatrick and Condon (1991) generated a linear solution to fit data gathered from experiments with solid pieces of uranium metal. Their final solution is

$$(25) \quad \dot{wg} = s \cdot U_o \cdot V_s \cdot (1 - U_{stop}) + C_o \cdot V_s$$

where $\dot{wg}$ is the weight gain in $\frac{\text{mole fraction} \cdot m}{s}$ which can be multiplied by the molar density of uranium to get units of $\frac{\text{moles}}{m^2 \cdot s}$, s is a stoichiometric constant (s=3 for UH_3), U_o is the uranium molar fraction for unreacted material (U_o is unity when expressing terms in mole fractions), V_s is the spall front velocity in $\frac{m}{s}$, U_{stop} is the uranium molar fraction where hydriding and dehydriding rates are equal and the reaction rate is effectively zero, and C_o is the solubility molar fraction for free hydrogen in uranium. V_s is calculated using the equation,

$$(26) \quad V_s = \frac{1}{t_1} \cdot \left(\frac{D_E}{s \cdot k_1 \cdot U_E} \right)^{\frac{1}{2}}$$

for which t_1 is the time to the first spall in seconds, D_E is the effective diffusivity in $\frac{m^2}{s}$, k_1 is a hydriding rate constant in s^{-1} and U_E is a generalized effective uranium molar

fraction for spall velocity. For the temperatures of interest in this thesis, t_l can be calculated by the equation,

$$(27)\ t_l = \frac{-\ln(U_c)}{\left[k_l \cdot (C_o - C_{eq})\right]}$$

for which U_c is the uranium molar fraction for spall, and C_{eq} is the equilibrium hydrogen molar fraction. U_c is a function of temperature and for temperatures below 835.5°K is estimated to be,

$$(28)\ U_c = 1 - (1 - 0.989)^{\left[0.5 - 0.5 \cdot \tanh\left(-2950 \cdot \left(\frac{1}{T} - 0.00146\right)\right)\right]}$$

k_l is also a function of temperature and is expressed by,

$$(29)\ k_l = 10.4 \cdot e^{\left(\frac{1592}{T}\right)}$$

C_o is a function of temperature and hydrogen partial pressure (P) in Pa and is given by,

$$(29)\quad C_o = \frac{N \cdot 4.183E-6 \cdot e^{\left(\frac{-894}{T}\right)} \cdot P^{\frac{1}{2}}}{\left(N + 4.183E-6 \cdot e^{\left(\frac{-894}{T}\right)} \cdot P^{\frac{1}{2}}\right)}$$

N is the maximum concentration of free hydrogen atoms (in mole fraction) in the metal lattice. Kirkpatrick's and Condon's proposed fit for N is,

$$(30)\quad N = e^{\left(-2.362 - \frac{2305}{T}\right)}$$

For C_{eq} in equation 26, Kirkpatrick and Condon use a zero value and account for a plateau or equilibrium pressure of hydrogen over hydride powder by including a dehydriding reaction in the model.

D_E in equation 25 is the effective diffusivity in $\frac{m^2}{s}$, and is given by,

$$(31) \quad D_E = D \cdot \left[\frac{N}{(N - C_o)} \right]^{\frac{1}{2}}$$

for which N and C_o are given above and D is the diffusivity of free hydrogen in uranium in $\frac{m^2}{s}$ and is calculated as a function of temperature by the equation

$$(32) \quad D = 1.9E-6 \cdot e^{\frac{-5820}{T}}$$

U_E in equation 25, the effective uranium molar fraction for spall velocity, is calculated from U_o and U_c,

$$(33) \quad U_E = (U_o \cdot U_c)^{\frac{1}{2}}$$

U_{stop} in equation 24 is the uranium molar fraction where hydriding and dehydriding rates are equal and the reaction rate is effectively zero and must be solved for iteratively using the equation,

$$(34) \quad k_1 \cdot (C_o - C_{eq}) \cdot U_{stop} = k_2 \cdot (1 - U_{stop})^{\frac{1}{3}}$$

The results from this analysis at room temperature are shown in figure 7 and fit well with laboratory experimental data.

Hydriding data is also available from industrial experience. The plot of data given in Figure 2 of "Calculated vs. Experimental Hydrogen Reaction Rates with Uranium" (Condon, 1975) has the same shape as Figure 7 but hydriding rates are a factor of ten lower.

4. Computer Modeling

The primary objectives of the computer simulation are to predict the pressure and composition of gases inside the MCO as a function of time for a given temperature and initial conditions. A model was created for each of the three corrosion regimes. Table 2 gives a summary of the evolution of gases and water in the three regimes.

Table 2: Mechanisms of Gas Production and Consumption

	Water	Hydrogen	Oxygen
Water immersion • O_2< ppm • O_2> ppm	• ↓ by R • ↓ by C	• ↑ by R • ↑ by C	• ↑ by R • ↓ by C
Water vapor • P_{o2}< 10 Pa • P_{o2}> 10 Pa • P_{H2}>P_{eq}	• ↓ by R • ↓ by C	• ↑ by R • ↑ by C • ↓ by H	• ↑ by R • ↓ by C
Dry Oxygen • With bound or hydrated water • P_{H2}>P_{eq}	• • ↓ by R	• • ↑ by R • ↓ by H	• ↓ by C • ↑ by R

R-radiolysis, C-corrosion, H-hydriding, ↑-produced, ↓-consumed

4.1 Water Immersion

The model requires the input of several physical constants and initial values with associated standard deviations :

- Physical parameters

1. Temperature
2. Amount of sludge
3. Free volume (volume inside MCO not occupied by fuel or baskets)
4. Gas space (free volume not occupied by water)
5. Exposed fuel surface area
6. Fraction of sludge that is bound water
7. Fraction of sludge that is chemisorbed water
8. G values for oxygen and hydrogen
9. Decay power
10. Fraction of decay energy that is deposited outside the fuel
11. Fraction of decay power that is deposited in water
12. Reaction rate constants for corrosion equations

- Initial values

1. Partial pressure of hydrogen
2. Amount of oxygen in water
3. Amount of free water

In addition, the oxygen limit below which corrosion occurs by water must be known, and the timestep for each calculation loop must be input.

The first step of the model is to convert the amounts of water, hydrogen, and oxygen to moles using molecular weights or the ideal gas law. Also, the oxygen limit and amount of oxygen are converted to ppm and compared to determine which corrosion mechanism will take place. The reaction rate is given by

$$k = 10^{\left(b+\frac{m}{T}\right)} \tag{35}$$

where k is in units of $\frac{mg\ O}{cm^2 \cdot s}$, b and m are rate constants, and T is temperature in °K

4.1.1 Oxygen-depleted water

When the amount of oxygen is less than the limit, corrosion occurs by equation one. Regression of the available data was performed and yielded the following reaction rate constants and uncertainties,

$m = -2274 \quad \sigma_m = 42.7$

$b = 4.59 \quad \sigma_b = 0.12$

The reaction rate is then multiplied by the exposed surface and the length of the timestep to determine the amount of weight gain (oxygen) per step. For equation one, 7.437 mg of uranium react for every mg of oxygen. Also, for every mole of uranium reacted, two moles of water are consumed, and two moles of hydrogen gas are released.

At the same time, oxygen and hydrogen are being produced and water is being consumed by radiolysis. For the water-immersed atmosphere the only type of radiation considered was gamma radiation. For hydrogen, the g-value was determined to be 1.04e-7 $\frac{\text{moles } H_2}{\text{Watt} \cdot \text{s}}$. Since the short-lived products are not considered in this analysis one can assume that the effective reaction is

(36) $$H_2O + \gamma \rightarrow H_2 + \frac{1}{2}O_2$$

and the oxygen produced is half the value of the hydrogen produced.

At this point, accounting begins of the number of moles of water, hydrogen, and oxygen remaining at the end of the timestep. The amount of water consumed during the timestep is the sum of water consumed in radiolysis and corrosion,

(37) *Total H_2O consumed = H_2O consumed in corrosion + H_2O consumed in radiolysis*

As long as the total water consumed is less than the amount of water available at the beginning of the time step, the new amount of water is,

(38) *New moles of H_2O = Initial moles of H_2O - Total H_2O moles consumed*

However, if the amount consumed exceeds the initial amount, the new amount of water is zero and the amounts of hydrogen and oxygen produced in radiolysis and the amount of hydrogen produced in corrosion are altered. This analysis assumed that the water available participated in corrosion processes preferentially to radiolytic processes so that the adjusted production terms are,

(39) *H_2 produced from corrosion = Initial moles of H_2O*

(40) *H_2 produced from radiolysis = 0*

(41) *O_2 produced from radiolysis = 0*

The amounts of hydrogen and oxygen present at the end of the timestep are the sums of the production terms and the initial amounts of hydrogen and oxygen. These final values become the initial values for the next calculation loop.

4.1.2 Oxygen-rich water

When the amount of oxygen is greater than the limit, corrosion occurs by equation two. Regression of the available data was performed and yielded the following reaction rate constants,

$m = -3797 \quad \sigma_m = 345$

$b = 8.54 \quad \sigma_b = 1.00$

As for the oxygen-depleted case, the reaction rate is then multiplied by the exposed surface and the length of the timestep to determine the amount of weight gain (oxygen) per step. 7.437 mg of uranium react for every mg of oxygen.

Radiolysis is unaffected by the corrosion mechanism and the parameters used are the same as for the oxygen-depleted case. Accounting for the number of moles of water, hydrogen, and oxygen is different.

In this regime, water is consumed by radiolysis only. As long as the total water consumed is less than the amount of water available at the beginning of the time step, the new amount of water is,

(42) $New\ moles\ of\ H_2O = Initial\ moles\ of\ H_2O - H_2O\ consumed\ in\ radiolysis$

However, if the amount consumed exceeds the initial amount, the new amount of water is zero and the amounts of hydrogen and oxygen produced by radiolysis become,

(43) $H_2\ produced\ from\ radiolysis = \frac{2}{3} \cdot Initial\ moles\ of\ H_2O.$

(44) $O_2\ produced\ from\ radiolysis = \frac{1}{3} \cdot Initial\ moles\ of\ H_2O$

The total change in moles of oxygen in the timestep is,

(45) $\Delta O_2 = O_2\ produced\ from\ radiolysis - O_2\ consumed\ in\ corrosion$

If ΔO_2 is positive or if ΔO_2 is negative and the absolute value is less than the initial amount of oxygen, then the new amount of oxygen is,

(46) $New\ moles\ of\ O_2 = Initial\ moles\ of\ O_2 + \Delta O_2$

If ΔO_2 is negative and the absolute value is greater than the initial amount of oxygen, then all of the oxygen is consumed in this timestep by corrosion and the new amount of oxygen is zero.

The new amount of hydrogen is the sum of the initial moles of hydrogen and that produced by radiolysis.

4.2 Water Vapor

The physical parameters and initial values with standard deviations that are required for the water vapor regime are:

- Physical parameters

    1. Temperature
    2. Amount of sludge
    3. Free volume (volume inside MCO not occupied by fuel or baskets)
    4. Exposed fuel surface area
    5. Fraction of sludge that is bound water
    6. Fraction of sludge that is chemisorbed water
    7. G values for oxygen and hydrogen
    8. Decay power
    9. Fraction of decay energy that is deposited outside the fuel
    10. Fraction of decay power that is deposited in water
    11. Reaction rate constants for corrosion equations

- Initial values

    1. Partial pressure of hydrogen
    2. Partial pressure of oxygen
    3. Partial pressure of water vapor

In addition, the oxygen limit, in Pa, below which uranium reacts with water must be known, and the timestep for each calculation loop must be input. The amount of water that is available for corrosion is different from that available for radiolysis. Only water

vapor is available for corrosion while vapor, bound, and chemisorbed water is available for radiolysis. Thus, the model must account for both terms.

The first step is to calculate the total pressure by summing the partial pressures of hydrogen, oxygen, water vapor. Then the partial pressures of water, hydrogen, and oxygen are converted to moles using the ideal gas law. The reaction rate is given by

(47) $$k = 10^{\left(b + \frac{m}{T} + pterm\right)}$$

where k is in units of $\frac{mg\ O}{cm^2 \cdot s}$, b and m are rate constants, T is temperature in °K, and pterm is a term that incorporates the total pressure. At the end of the calculation loop, the new amount of water available for radiolysis is determined. Water can be consumed by corrosion and by radiolysis. Therefore the total consumption term is

(48) *H_2O consumed= H_2O consumed by corrosion + H_2O consumed by radiolysis*

and the new total amount of water available for radiolysis is

(49) *New H_2O for radiolysis = Initial amount - H_2O consumed*

The total pressure in the MCO is also calculated at the end of the loop by summing the partial pressures of water vapor, hydrogen, and oxygen.

4.2.1 Oxygen-depleted vapor

When the amount of oxygen is less than the limit, corrosion occurs by equation one. Regression of the available data was performed and yielded the following reaction rate constants,

$$m = -2274 \quad \sigma_m = 42.7$$
$$b = 4.59 \quad \sigma_b = 0.12$$
$$pterm = 0.3 \log(P)$$

for which P is the total pressure in kPa. The reaction rate is then multiplied by the exposed surface and the length of the timestep to determine the amount of weight gain (oxygen) per step. For equation one, 7.437 mg of uranium react for every mg of oxygen. Also, for every mole of uranium reacted, two moles of water are consumed, and two moles of hydrogen gas are released.

Radiolysis produces hydrogen and oxygen as in the water immersion case. In addition to corrosion- and radiolytic- induced changes in the atmosphere, uranium hydriding can occur in the water vapor atmosphere if the partial pressure of hydrogen exceeds the equilibrium pressure that is calculated to be,

$$(50) \quad P_{eq} = 133.29 \cdot 10^{\left(\frac{-4590}{T} + 9.39\right)} \qquad \text{(Spedding, et. al., 1949)}$$

Hydriding consumes hydrogen gas.

If the amount of water consumed in corrosion is less than the initial moles of water vapor then the new amount of water vapor is simply the difference between the two. Otherwise, the new amount of water vapor is zero, the amount of water consumed

in the timestep by corrosion equals the initial amount of water vapor, and the moles of hydrogen produced by corrosion equals the moles of water vapor consumed.

Hydrogen may be produced by radiolysis and corrosion so that

(51) *Total H_2 produced = H_2 produced by corrosion + H_2 produced by radiolysis*

and may be consumed by hydriding. The hydrogen partial pressure cannot be less than the equilibrium pressure given in equation 49. If the amount of hydrogen consumed is less than the initial amount available then the new amount of hydrogen is

(52) *New H_2 = Initial H_2 + Total H_2 produced – H_2 consumed by hydriding*

Otherwise the new amount of hydrogen gas is the total hydrogen produced in the timestep plus the minimum hydrogen partial pressure.

The new amount of oxygen is simply the initial amount plus the amount produced by radiolysis.

4.2.2 Oxygen-rich water vapor

When the amount of oxygen is greater than the limit, corrosion occurs by equation two. In this regime, the reaction rate constants depend upon temperature and relative humidity. When the relative humidity is 100% the reaction rate constants are,

$m = -3797$ $\sigma_m = 345$
$b = 8.54$ $\sigma_b = 1.00$
$pterm = 0$

If the humidity is less than 100% and the temperature is less than 192°C, the reaction rate constants are,

$$m = -3720 \quad \sigma_m = 93.6$$
$$b = 7.50 \quad \sigma_b = 0.24$$
$$pterm = 0.3 \cdot \log(P)$$

where P is the total pressure in kPa. Finally, if the temperature exceeds 192°C, the reaction rate constants are

$$m = -2963 \quad \sigma_m = 100$$
$$b = 6.1931 \quad \sigma_b = 0.02$$
$$pterm = 0.3 \cdot \log(P)$$

The reaction rate is then multiplied by the exposed surface and the length of the timestep to determine the amount of weight gain (oxygen) per step. 7.437 mg of uranium react for every mg of oxygen.

Radiolysis produces hydrogen and oxygen and consumes water, and hydriding may consume hydrogen as in the preceding case.

Hydrogen may be produced by radiolysis and corrosion so that

(53) *Total H_2 produced = H_2 produced by corrosion + H_2 produced by radiolysis*

and may be consumed by hydriding. If the amount of hydrogen consumed is less than the initial amount available then the new amount of hydrogen is

(54) *New H_2 = Initial H_2 + Total H_2 produced – H_2 consumed by hydriding*

Otherwise the new amount of hydrogen gas is the total hydrogen produced in the timestep.

4.3 Dry Oxygen

The physical parameters and initial values with standard deviations that are required for the dry oxygen regime are:

- Physical parameters

  1. Temperature
  2. Amount of sludge
  3. Free volume (volume inside MCO not occupied by fuel or baskets)
  4. Exposed fuel surface area
  5. Fraction of sludge that is bound water
  6. Fraction of sludge that is chemisorbed water
  7. G values for oxygen and hydrogen
  8. Decay power
  9. Fraction of decay energy that is deposited outside the fuel
  10. Fraction of decay power that is deposited in water
  11. Reaction rate constants for corrosion equations

- Initial values

  1. Partial pressure of hydrogen
  2. Partial pressure of oxygen
  3. Total pressure

The first step is to convert the partial pressures of hydrogen and oxygen to moles using the ideal gas law. Then the moles of inert gas can be calculated. The reaction rate equation is of the form

$$k = 10^{\left(b + \frac{m}{T}\right)} \tag{55}$$

Regression of the available data was performed and yielded the following reaction rate constants,

$$m = -3520 \quad \sigma_m = 100$$
$$b = 6.71 \quad \sigma_b = 0.02$$

The reaction rate is then multiplied by the exposed surface and the length of the timestep to determine the amount of weight gain (oxygen) per step. 7.437 mg of uranium react for every mg of oxygen. One mole of oxygen gas is consumed for every mole of uranium consumed.

As in the previous case, hydrogen and oxygen may be produced and water consumed by radiolysis and hydrogen may be consumed by uranium hydriding.

If the amount of water consumed by radiolysis is less than the initial amount of water then the new amount is simply the difference between the terms. Otherwise, the new amount of water available for the timestep is zero and the production terms for hydrogen and oxygen radiolysis become,

$$(56) \quad H_2 \text{ produced from radiolysis} = \frac{2}{3} \cdot \text{Initial moles of } H_2O$$

$$(57) \quad O_2 \text{ produced from radiolysis} = \frac{1}{3} \cdot \text{Initial moles of } H_2O$$

For hydrogen, while the amount of hydrogen consumed by hydriding is less than the initial amount of hydrogen available, then the new amount of hydrogen is

$$(58) \quad \text{New } H_2 = \text{Initial } H_2 + H_2 \text{ produced from radiolysis} - H_2 \text{ consumed by hydriding}$$

However if the amount consumed is greater than the initial amount of hydrogen, then the new amount of hydrogen is the amount produced by radiolysis.

If the amount of oxygen consumed in corrosion is less than the initial amount available then the new amount of oxygen is

(59) *New O_2 = Initial O_2 + O_2 produced from radiolysis - O_2 consumed by corrosion*

The new total pressure is calculated by adding the new number of moles of hydrogen and oxygen to the number of moles of inert gas.

4.4 Error propagation

Standard deviations were assigned to physical parameters such as temperature and the amount of sludge and to calculational parameters such as reaction rate constants. In the case of reaction rate constants with log-linear behavior, standard deviations were calculated formally from the data. Most standard deviations of physical parameters are best guesses and can be modified as more data is accumulated.

The errors were propagated through the model using standard statistical techniques. The approximation of the standard deviation σ_x for x that is a function of u and v is

$$(60) \qquad \sigma_x^2 = \sigma_u^2 \cdot \left(\frac{dx}{du}\right)^2 + \sigma_v^2 \cdot \left(\frac{dx}{dv}\right)^2 + 2\sigma_{uv}^2 \cdot \left(\frac{dx}{du}\right) \cdot \left(\frac{dx}{dv}\right)$$

where σ_{uv}^2 is the covariance between the variables u and v (Bevington and Robinson, 1992). For this model, all of the covariances between parameters were assumed to be zero. Equation sixty can be simplified for common mathematical operations such as addition, subtraction, multiplication, division, performing logarithms, and raising powers.

5. Results and Discussion

5.1 Monte Carlo analysis

A Monte Carlo analysis was used to attempt to verify the internal consistency of the model. For variable parameters listed above a random Gaussian sample population was generated with the exception of the exposed surface area and amount of sludge for which Poisson distributed samples were generated. The model was run with these sample distributions and the results were compared. Table 3 lists typical results. The following values were used for each regime:

- Free volume = 0.6 m^3
- Sludge = 10 kg $\sigma = 3$
- Exposed surface area = 3000 cm^2 $\sigma = 500$
- Decay power = 550 W $\sigma = 100$

The mean values of the parameters agreed. However, several of the calculated standard deviations differed. The most notable differences occur in the standard deviations of oxygen partial pressure and total pressure for the dry oxygen case. Most likely the difference in the standard deviation of the total pressure is caused by the difference in standard deviation in the oxygen pressure. Possibilities for these differences are:

Table 3: Comparison of Model and Monte Carlo Simulation Results

	Model	Monte Carlo
Dry air Temperature=25°C σ = 1 P_{total}=1.0135e5 Pa σ = 100 Initial P_{o2} = 0 Initial P_{H2} =P_{eq} Time=50 years		
Hydrogen partial pressure (Pa)	370.4	378.18
σ_{H2}	129.9	129.9
Oxygen partial pressure (Pa)	7946	7694
σ_{O2}	483.9	3488.7
Total pressure (Pa)	3.23e5	3.17e5
σ_{tot}	284.4	94657
Water-immersed Temperature=50°C σ = 10 Free H_2O=550 kg Gas space=0.05 m^3 Time=100 hours		
Hydrogen partial pressure (Pa)	30570	30574
σ_{H2}	10.47	13.39
Water (moles)	1.11e7	1.11e7
σ_{water}	4.12e4	2.31e4
Water vapor Temperature=25°C σ =10 Free H_2O=0.01 kg Initial P_{H2O}=1.013e5 Pa σ = 10 Time=100 hours		
Water vapor pressure (Pa)	101300	101400
σ_{water}	4807	3304
Hydrogen partial pressure (Pa)	45.89	65.16
σ_{H2}	51.56	60.25
Oxygen partial pressure (Pa)	354.3	355.0
σ_{O2}	13.98	79.27
Total pressure (Pa)	101700	101900
σ_{total}	4807	3272

1. The distributions are not appropriate for the variable sampled. In particular, the exposed surface area and amount of sludge may require more complex distributions than a Poisson distribution to model the behavior correctly.

2. Errors are not propagated correctly through the model. For instance, if the amount of oxygen consumed in one timestep is greater than the original amount of oxygen, then the final amount of oxygen present in the timestep is set to the amount generated by radiolysis, and the standard deviation is set to the standard deviation of the oxygen generated by radiolysis. A more complex means of calculating the error for this type of occurrence may be needed.
3. All covariances are assumed to be zero. Investigation of the dependence variables have on one another may reveal covariances that should be incorporated into the model.

5.2 Water-immersed staging

5.2.1 Effect of Temperature

Before cold vacuum drying (CVD), the loaded MCOs will be sealed and full of water. The primary concern at this time is accumulation of hydrogen in the gas space that could pressurize the container. Figure 8 shows the linear increase of hydrogen as a function of time for an MCO at 25°C containing typical amounts of sludge and exposed surface area and a gas space of 0.05 m^3. $\frac{\Delta P}{\Delta t}$ for this atmosphere is about 100 Pa/hr. The design limit of the MCO is 150 psi, or 1.03 Mpa, so one can assume that the containers will not pressurize in the short times that they will be sealed and full of water. Figure 9 shows the effect of raising the temperature on the hydrogen increase. Between 25 and 100°C the curves are nearly indistinguishable but at 130°C, the rate of increase jumps markedly after about 90 hours. This behavior is due to the consumption of oxygen and

the beginning of hydrogen production by corrosion. With the increase, $\frac{\Delta P}{\Delta t}$ is about 1000 Pa/hr and the containers would reach the pressure design limit in about 1000 hours which is an unreasonably long staging time. Figure 10 shows the standard deviation associated with the pressure values and that the uncertainty of the final pressure is quite small.

5.2.2 Effect of decay power

In Figure 11, the effect that decay power has on hydrogen partial pressure at 50°C after 200 hours of staging is shown. As the decay power increases, the amount of hydrogen and oxygen produced increases with small uncertainties in the final values. Oxygen remains in the water but hydrogen accumulates in the gas space. However, the total effect is small with $\frac{\Delta P}{\Delta \text{decay power}}$ approximately 40 Pa/W. The decay power that would cause overpressurization in this regime is about 25 kW. The fuel in the MCO is of low burnup values and the maximum decay power per MCO is approximately 900 W.

5.2.3 Effect of exposed surface area

Figure 12 displays the effect the exposed uranium surface area has on the hydrogen partial pressure. At values less than 10000 cm^2, increasing the surface area has no significant effect on the final hydrogen partial pressure. However, above 10000 cm^2 $\frac{\Delta P}{\Delta \text{surface area}}$ is about 19.4 Pa/cm^2. At the typical values of decay power and temperature given, the partial pressure after 200 hours approaches the pressure design limit above 55000 cm^2 of exposed surface area.

These results were for staging times much longer than the anticipated time of about 24 hours. However, the effect surface area has on the pressure should not be

disregarded and care should be taken to ensure that the anticipated time the MCO spends in water-immersed staging is not exceeded. In addition, it is imperative that the gas space in the MCO is sufficient to accommodate the accumulation of the hydrogen.

5.3 Staging in Water Vapor

After cold vacuum drying, the MCO will contain very little, if any free water, but will contain water vapor and water bound in the sludge. The MCO will be staged before hot conditioning. Figure 13 shows the effect temperature has on the total pressure over a storage period of 200 hours. As the temperature rises, the pressure reduction rate is increased significantly. At 25 and 50°C, it appears that the pressure remains constant, but Figure 14 shows how the pressure decreases at 50°C. In Figure 13 it appears that the pressure begins to approach a constant value after 200 hours at 130°C. The change in pressure drop is due to the increasing consumption rate of water by corrosion and hydrogen by hydriding.

The behaviors of the different components of the total pressure are shown in Figures 15-19. In Figure 15, the oxygen partial pressure rises with time at 25°C. However, the rise is offset by the decrease in the water vapor pressure as shown in Figures 17 and 18. In this regime, oxygen is produced by radiolysis and may be consumed by corrosion. Corrosion and radiolysis consume water. Figure 18 shows the decrease in water vapor pressure at 50°C. Last, the hydrogen partial pressure versus time is shown in Figure 19. After 5 hours, the hydrogen pressure drops to an equilibrium value. This behavior is explained by assuming that as the oxygen partial pressure increases, the corrosion mechanism changes from water consumption to oxygen consumption, and thus, hydrogen is no longer produced by oxidation. At this point,

hydrogen is produced only by radiolysis and is consumed by hydriding. This analysis shows that the total pressure inside the MCO will decrease during storage.

5.3.1 Effect of decay power

As the decay power at 25°C increases, the partial pressures of hydrogen and oxygen after 200 hours rise accordingly as shown in Figures 20 and 21. However, the changes in total pressure and standard deviation are small as shown in Figure 22. As with the water-immersed case, the decay power would have to be unrealistically high to warrant concern that the container would reach the pressure design limit.

5.3.2 Effect of exposed surface area

An increase in the exposed surface area increases the rate at which water vapor or oxygen is consumed. Figure 23 shows the resulting pressure decrease and small associated standard deviation. For this regime, corrosion consumes water and the pressure decreases relatively rapidly. For an atmosphere in which oxygen is consumed, the pressure drops more slowly. Regardless, an increase in surface area during staging in water vapor reduces the MCO pressure.

5.3.3 Effect of sludge

The amount of sludge in the MCO filled with vapor did not affect the final pressure in the model. However, the model for the vapor regime did not include the separate radiolysis mechanism that occurs in the sludge. It is expected that modifying the code to include the radiolysis to water and the radiolysis to sludge will result in an increase in hydrogen and oxygen partial pressures with an increase in the amount of sludge in the MCO.

5.4 Dry Interim Storage

After hot conditioning, the MCO will be placed in the CSB and stored for up to 50 years. During this time, the mechanisms that may occur to change the pressure are corrosion, which consumes oxygen, radiolysis, which consumes water and produces oxygen and hydrogen, and uranium hydriding, which consumes hydrogen. In addition to hydrogen and oxygen, a backfill gas will contribute to the total pressure.

As expected, Figure 24 shows that the total pressure increases with increasing storage temperature and reaches a constant value. Standard deviations are shown for pressure at 70°C in figures 25, 26, and 27 and are typical of the other temperatures of interest. The standard deviations for hydrogen and oxygen partial pressures are large percentages of the partial pressures, but are insignificant when incorporated into the total pressure.

5.4.1 Effect of decay power

Figure 28 shows that increasing decay power has no effect on the final pressure at 25°C until it exceeds 820W. This behavior corresponds to a significant increase in the oxygen partial pressure as shown in Figure 29. This behavior can be explained by assuming that at 820 W, the production of oxygen by radiolysis has exceeded the amount that is consumed by corrosion. The rise in pressure as a function of decay power is small; $\frac{\Delta P_{total}}{\Delta \text{decay power}}$ is about 0.2 Pa/W. Again, the decay power would have to be unreasonably high to pressurize the container in this regime. The final hydrogen partial pressure increases steadily with decay power as shown in Figure 30, but does not contribute significantly to the total pressure.

5.4.2 Effect of exposed surface area

The only corrosion mechanism during interim storage is that which consumes oxygen. Therefore, an increase in the reacting area produces a decrease in the oxygen partial pressure. Figure 31 shows this effect.

5.4.3 Effect of sludge

In the dry atmosphere, radiolysis of water occurs only to that water that is chemisorbed in the sludge. Therefore, an increase in the amount of sludge leads to an increase in the production of hydrogen and oxygen. Figures 32 and 33 confirm this effect. However, the final oxygen and hydrogen partial pressures after 50 years, even with 200 kgs of sludge contained in the MCO, are very small with a high degree of certainty.

6. Conclusions

A working model to predict the pressure and composition of gases inside a MCO containing metallic uranium, that includes radiolysis, corrosion, and hydriding, has successfully been created. The model allows investigators to predict how changing parameters will effect the pressure inside the MCO and generates a standard deviation associated with each calculation.

6.1 Significance of Results

The most important result of the model is proving that under typical conditions, the MCO will not pressurize during its expected service life and, during interim storage, will reach an acceptable constant pressure. In fact, for interim storage lasting 50 years, the total pressure is expected to decrease with time regardless of the amount of sludge that is present or the value of decay power. The regime in which greater concern is warranted is water-immersed staging. If the amount of exposed surface area remains

largely uncertain, it becomes imperative that maximum staging times are determined and not exceeded. It is also important that sufficient gas space is available to accommodate hydrogen production. As better estimates of variables and associated uncertainties are developed, the model will be easily modified to more accurately predict the pressure inside the MCO.

6.2 Future Work

It is intended that the model be continuously modified as characterization data is accumulated and better physical models are developed. A model to predict the release of water by dehydration will be added. Also, a transient thermal analysis is a likely improvement. A thermal analysis would be required to incorporate decay heat, reaction rate heat from corrosion and hydriding, and heat removal through the MCO walls. This may be especially important for interim storage for which the MCO will be located in a storage tube with very little surrounding gas space.

In addition, the statistical analysis will be improved. While Gaussian distributions are appropriate for most of the variables, two key parameters, the exposed surface area and the amount of sludge, do not physically fit such a distribution. The Poisson distribution is a slight improvement, in that values below zero are not permitted, but other candidate distributions may yield more accurate results. These distributions include the Rayleigh distribution and the Subbotin distribution (Christensen, 1990). Also, the manner in which errors are propagated through the model may be improved by rigorous statistical analysis. Such improvements would yield an accurate picture of how the standard deviation of one variable affects the standard deviation of the final pressure.

The radiolysis routine for the vapor regime should also be modified. In the water vapor case, radiolysis of water vapor yields different amounts of hydrogen and oxygen than radiolysis of bound and chemisorbed water in the sludge. The current model lumps together the total amount of water in the MCO to calculate the amounts of oxygen and hydrogen generated. Improvement of this routine would yield more accurate estimates of gas generation during staging between cold vacuum drying and hot conditioning.

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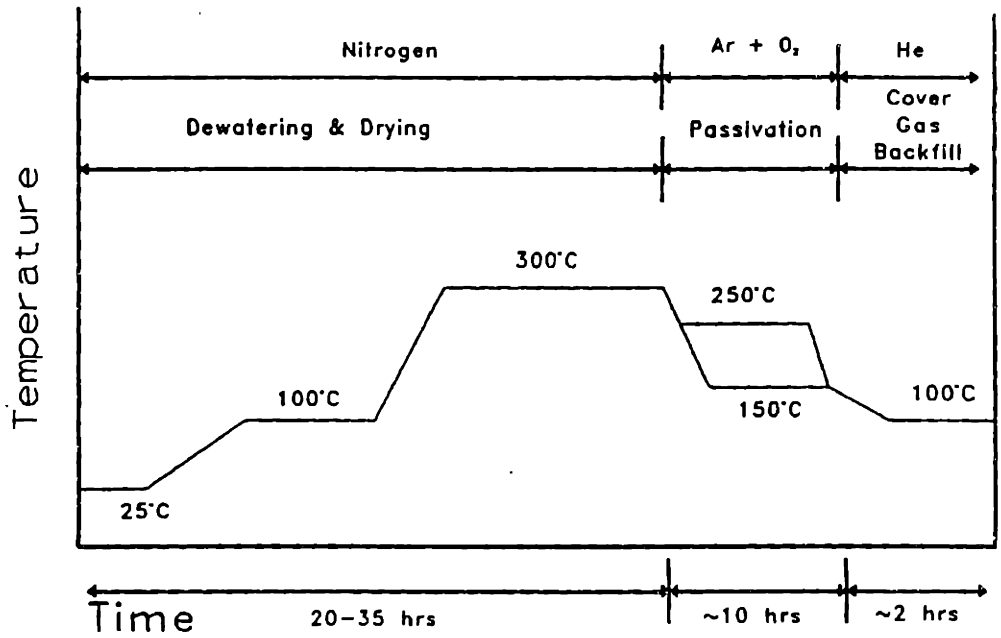
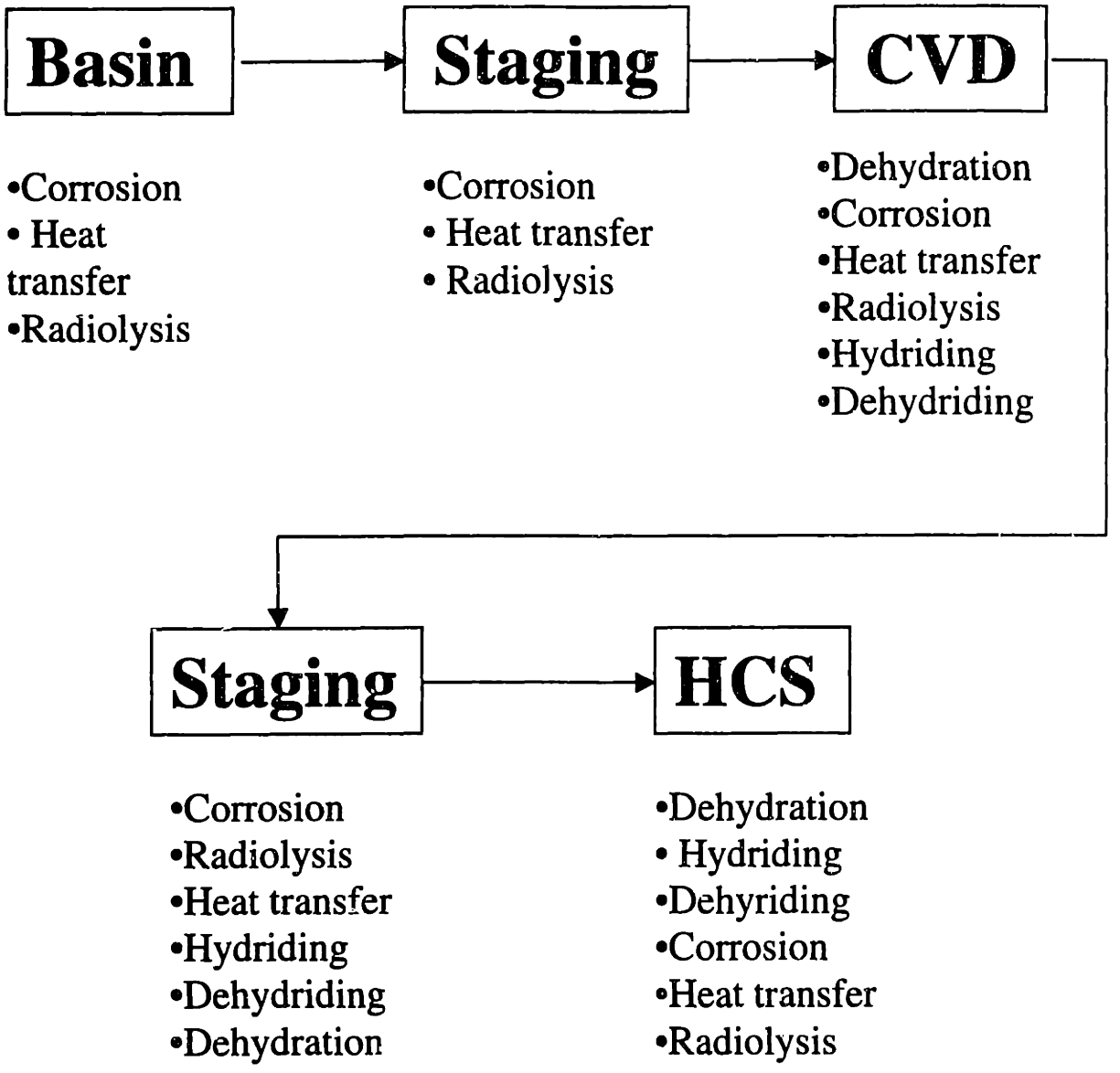


Figure 1: Conditioning Process ("Dry Storage of N Reactor Fuel, 1994)



2: Treatment Steps and Associated Phenomena

Figure 3: Comparison of Reaction Rates from French Data, Pearce Data Regression, and Pearce Equation

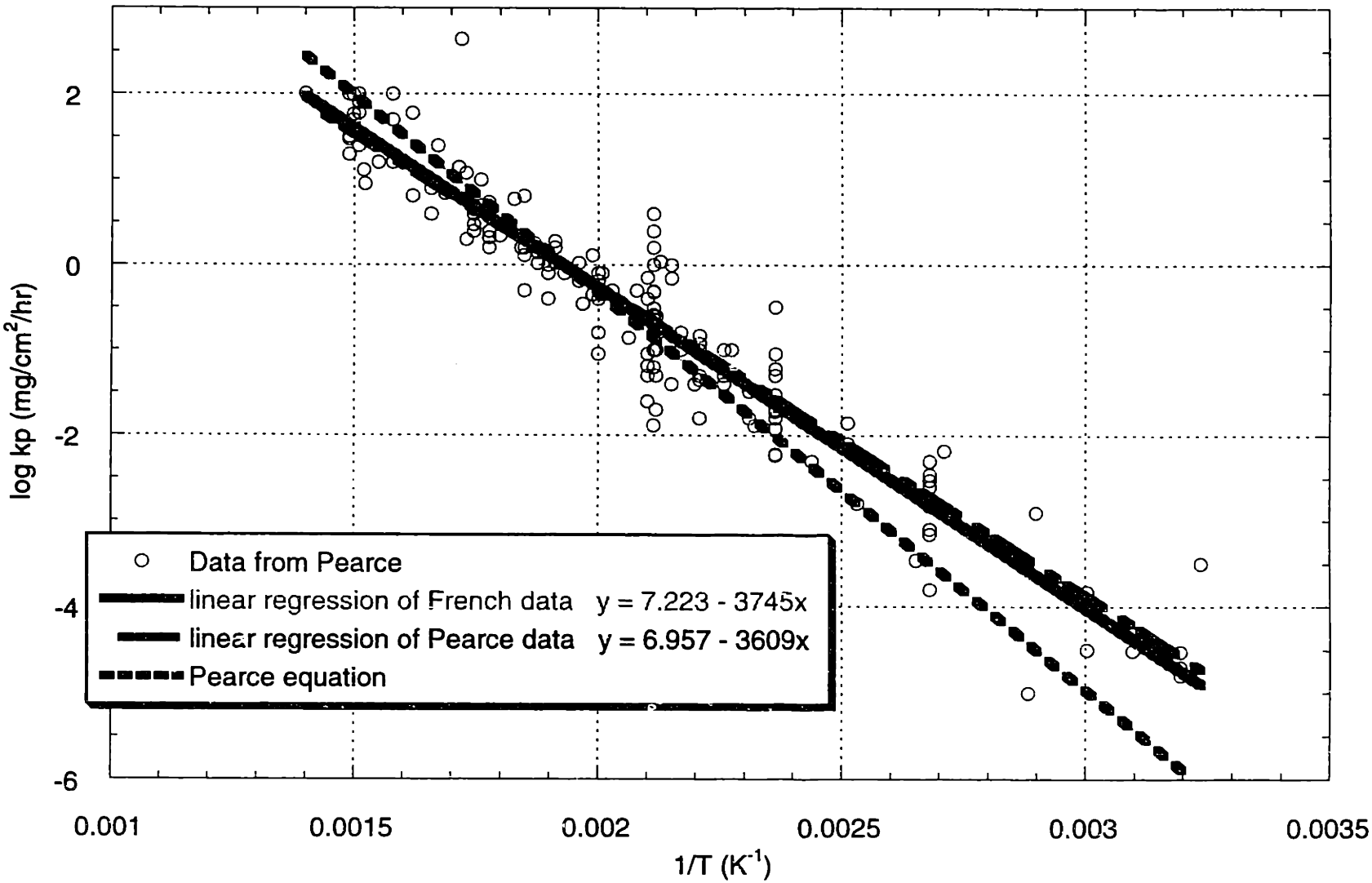


Figure 4: Summary of Reaction Rate Data, U-H_2O (Vapor) Reaction, Water Vapor Pressure Range 0.006-101kPa (Pearce, 1989)

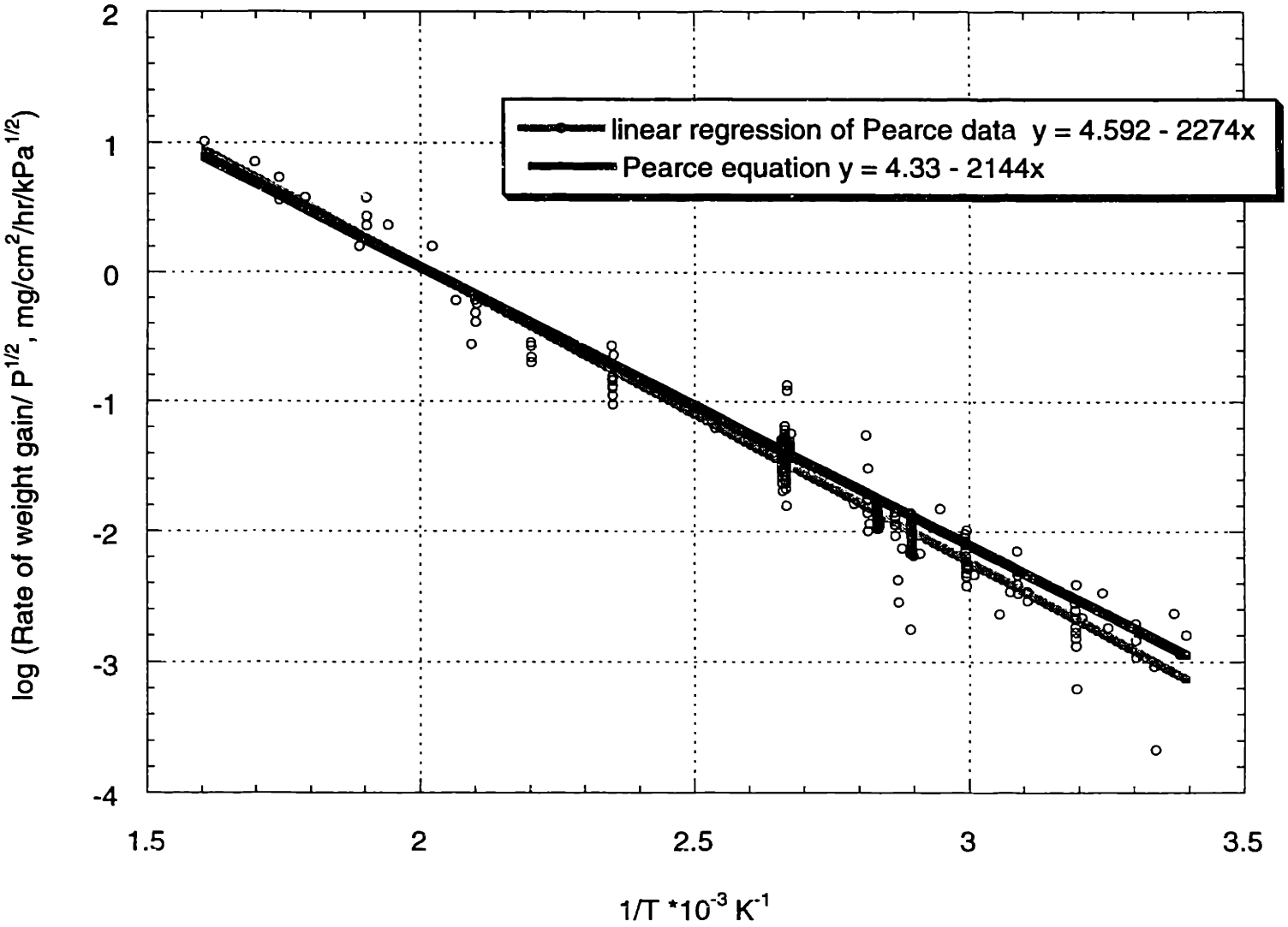


Figure 5: Rates of Reaction at 100% RH in the U-H_2O-O_2 System (Pearce 1989, Fig. 13)

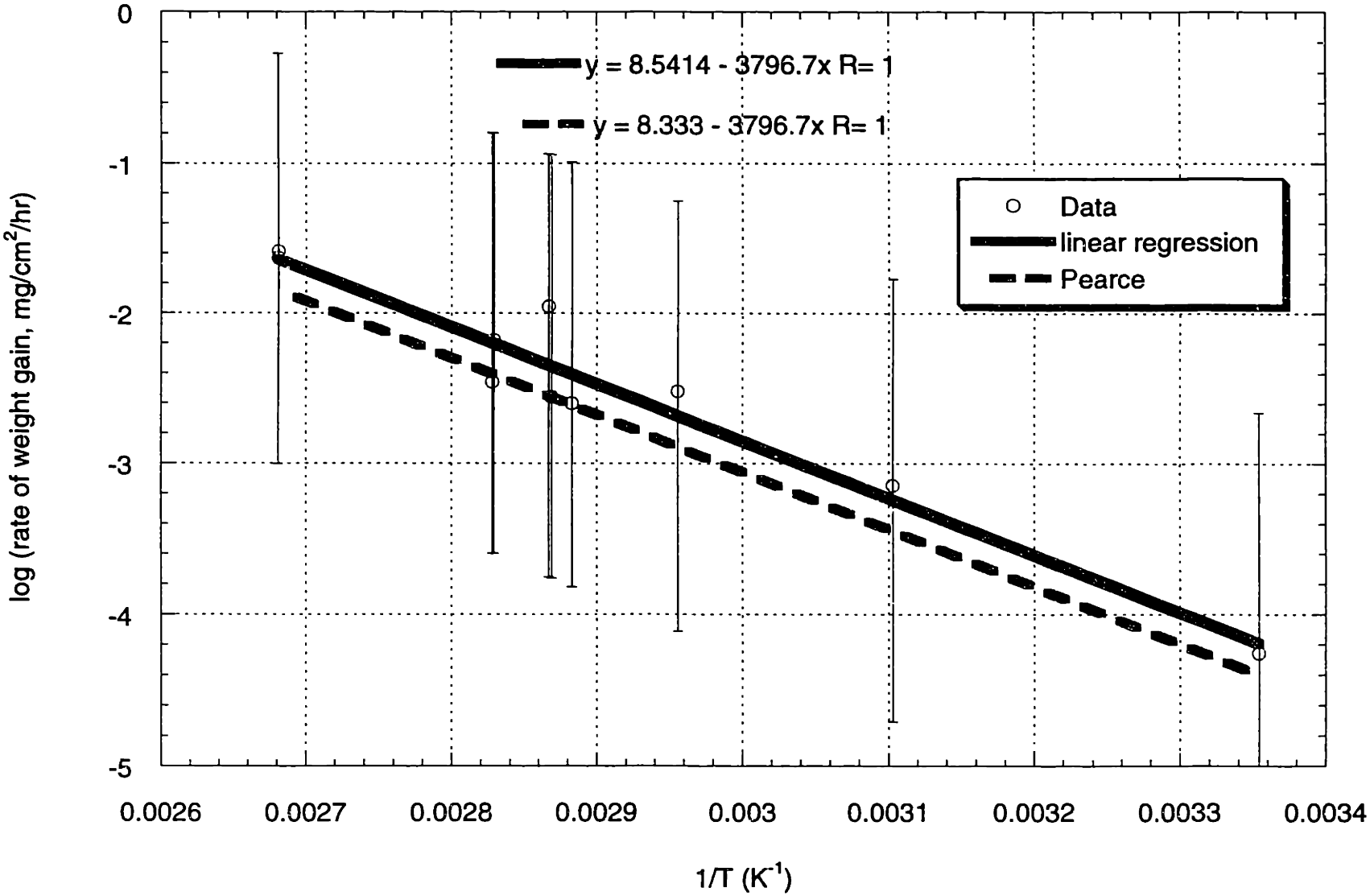


Figure 6: Reaction Rate Data, U-H_2O (Vapor) Reaction (Pearce, 1989)

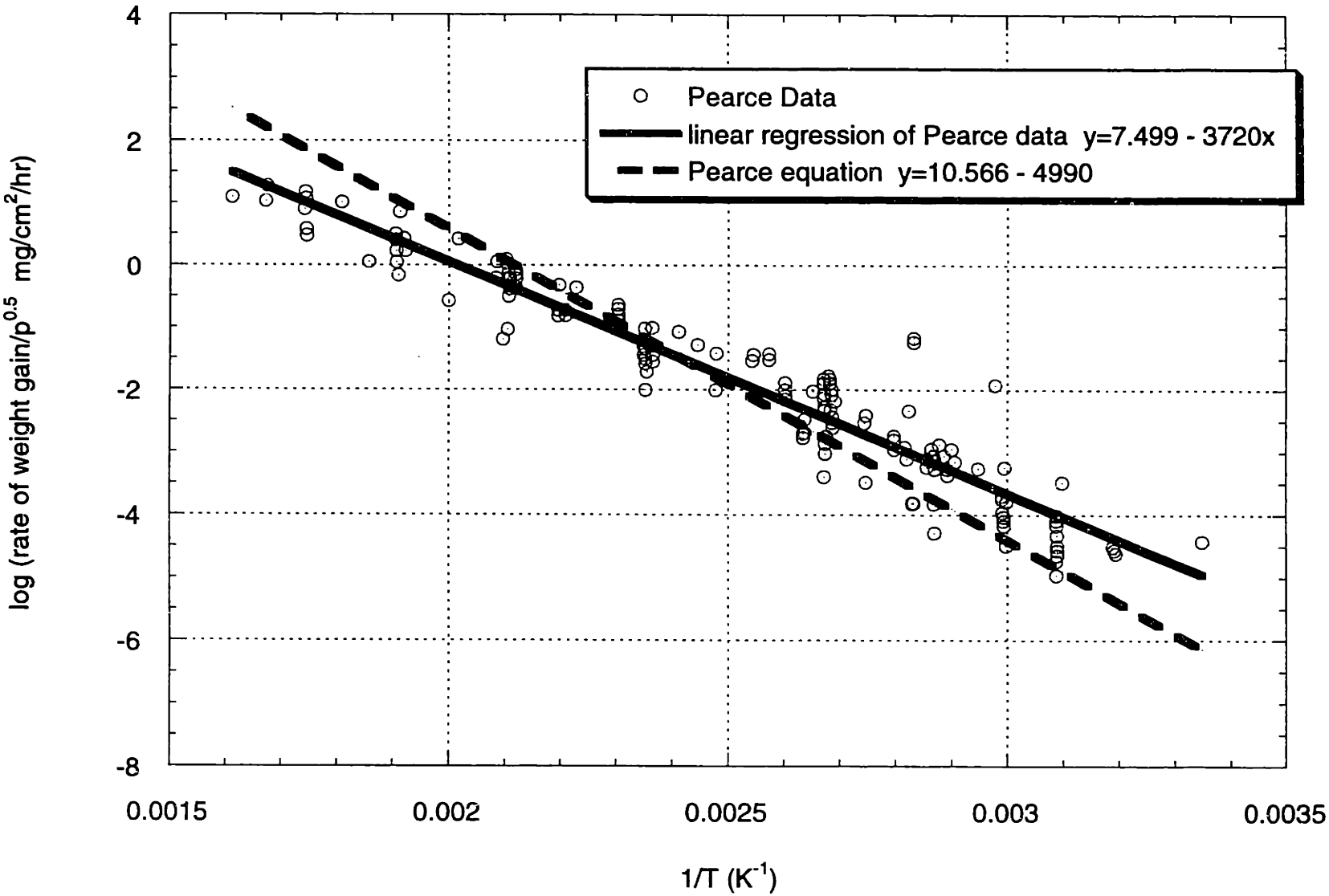


Figure 7: Results of Hydride Model at 25°C

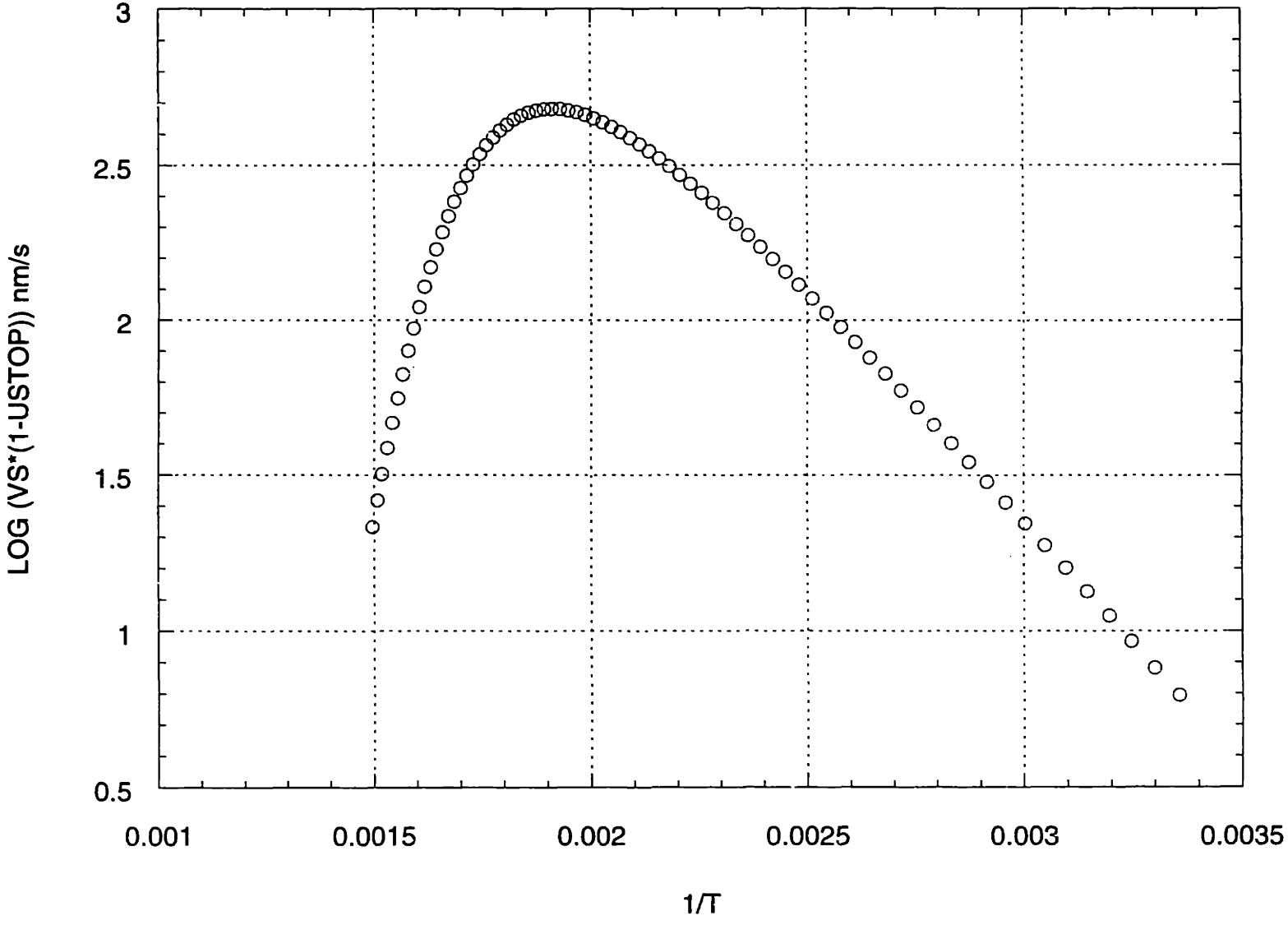


Figure 8: Hydrogen partial pressure vs. Time during water-immersed staging at 25°C

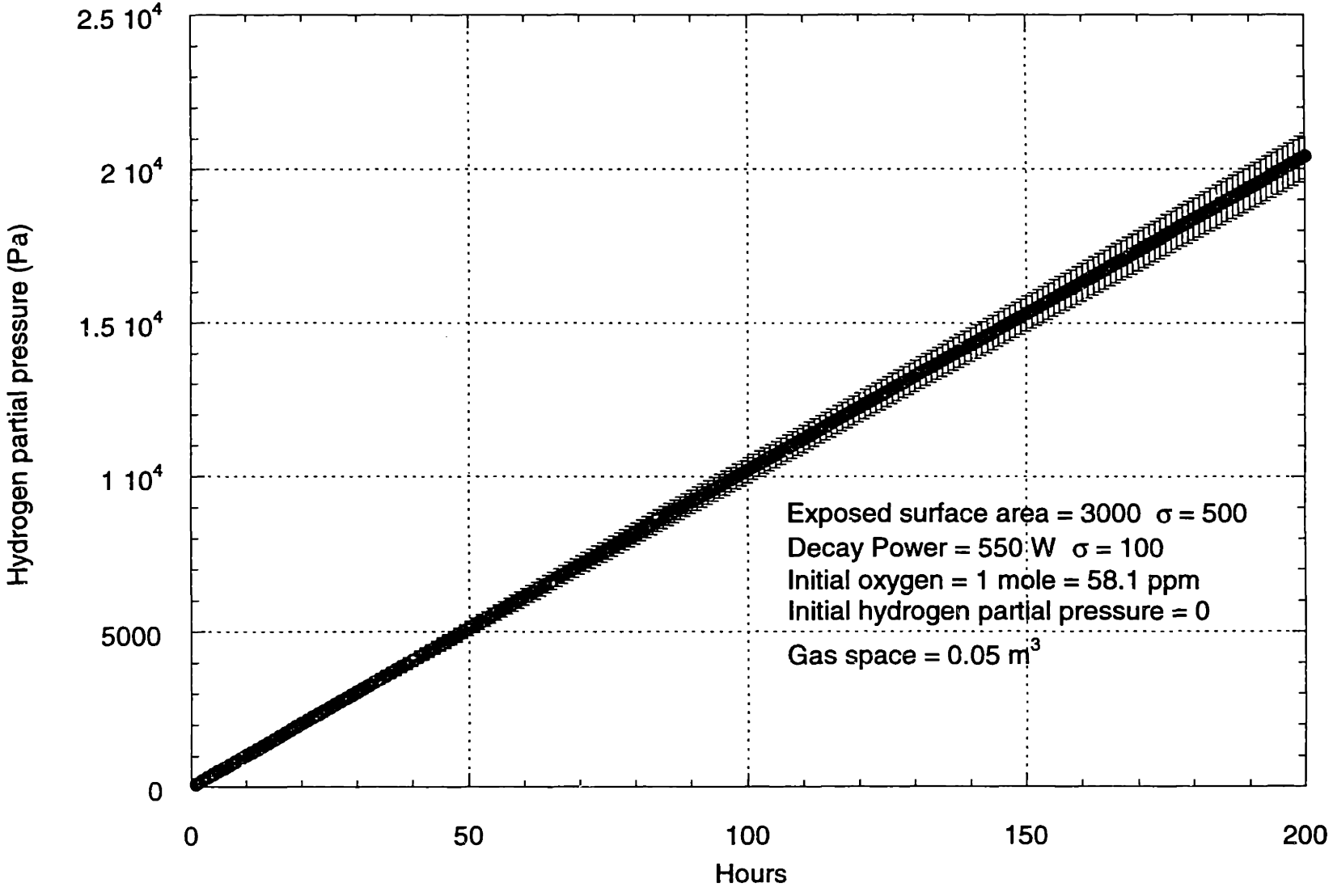


Figure 9: Hydrogen Partial Pressure vs. Time during water-immersed staging at several temperatures

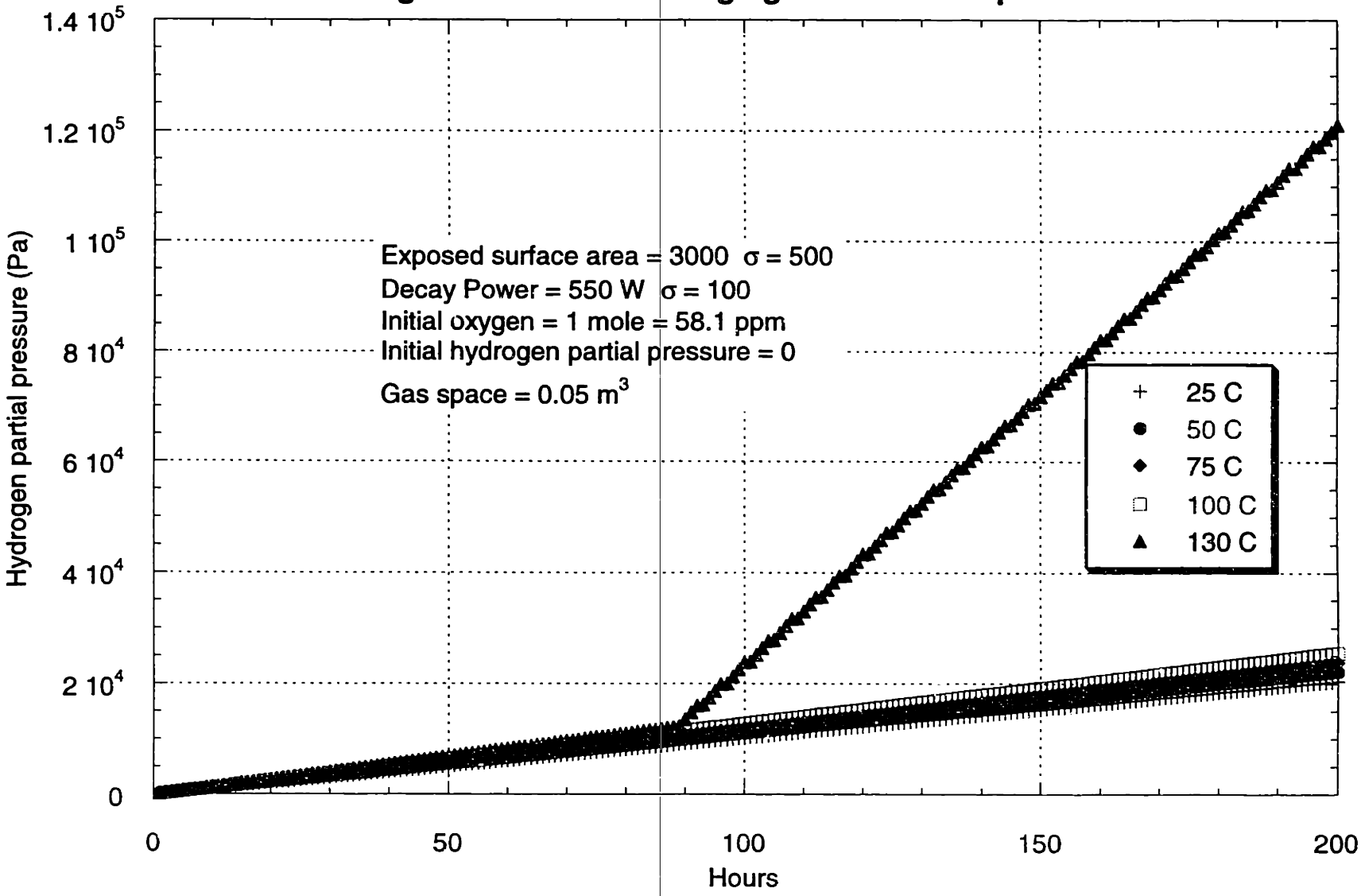


Figure 10: Hydrogen Partial Pressure vs. Time during water-immersed staging at 130°C

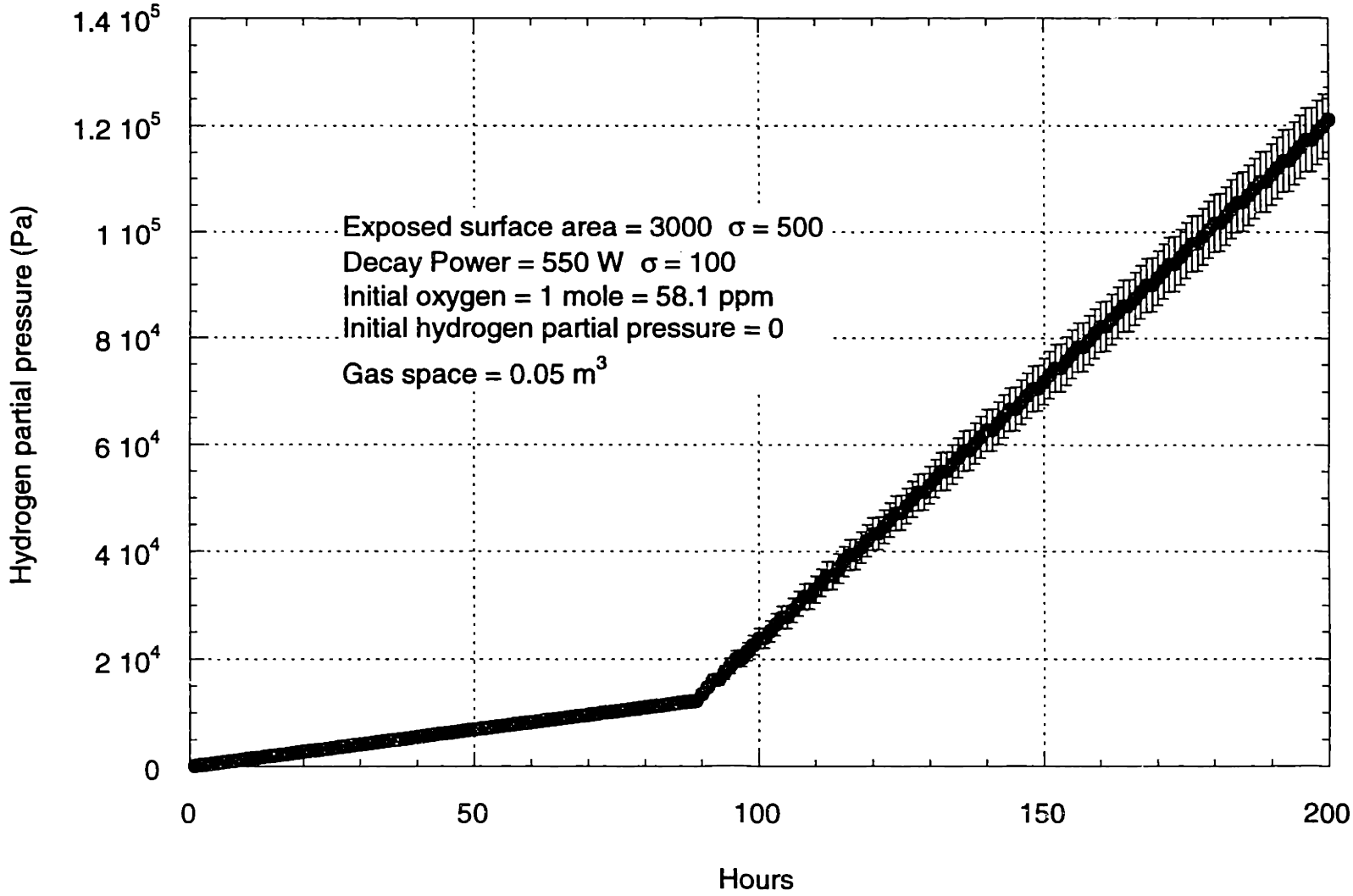


Figure 11: Hydrogen Partial Pressure vs. Decay Power after 200 hours of water-immersed staging at 25°C

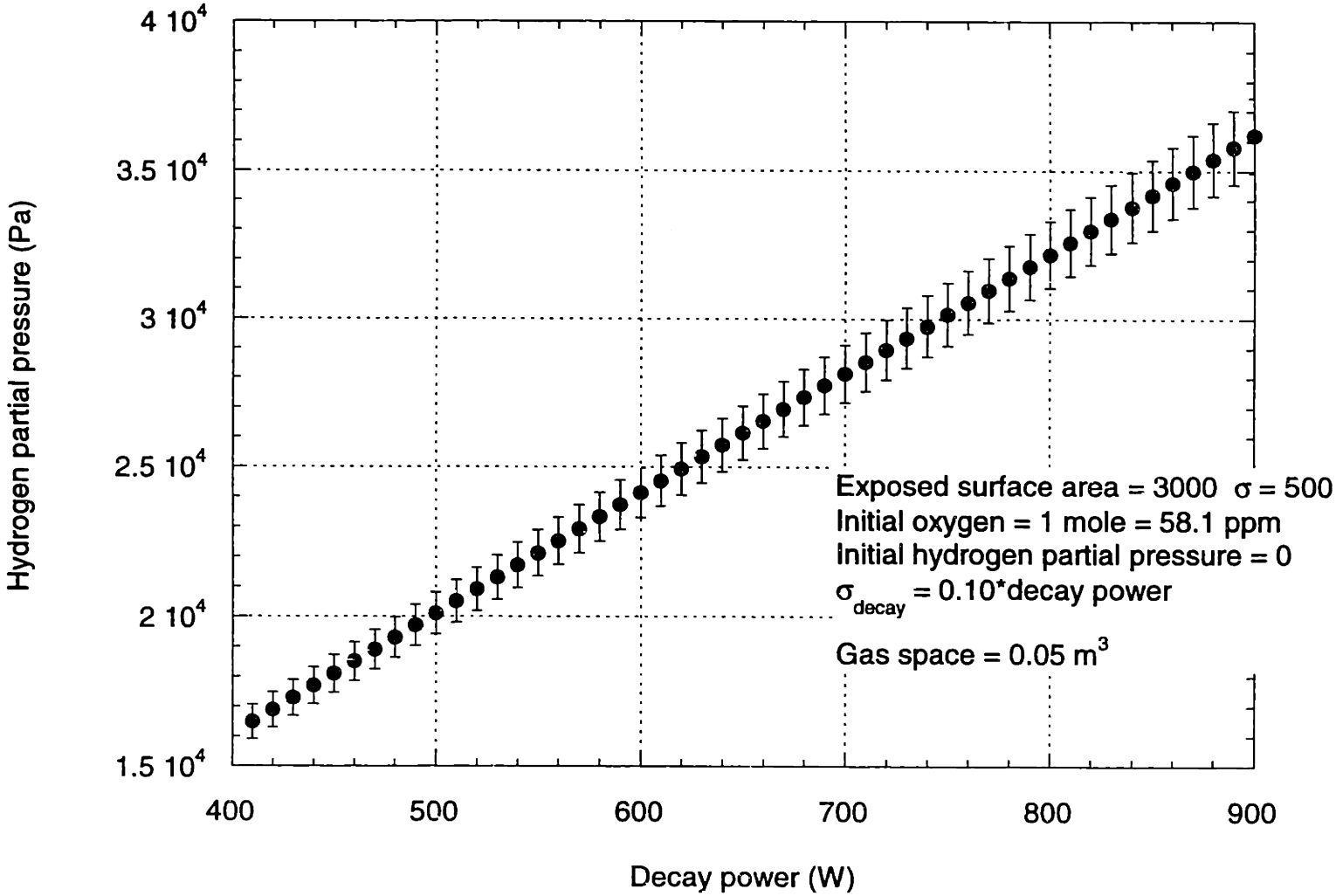


Figure 12: Hydrogen Partial Pressure vs. Exposed Surface Area after 200 hours of water-immersed staging at 25°C

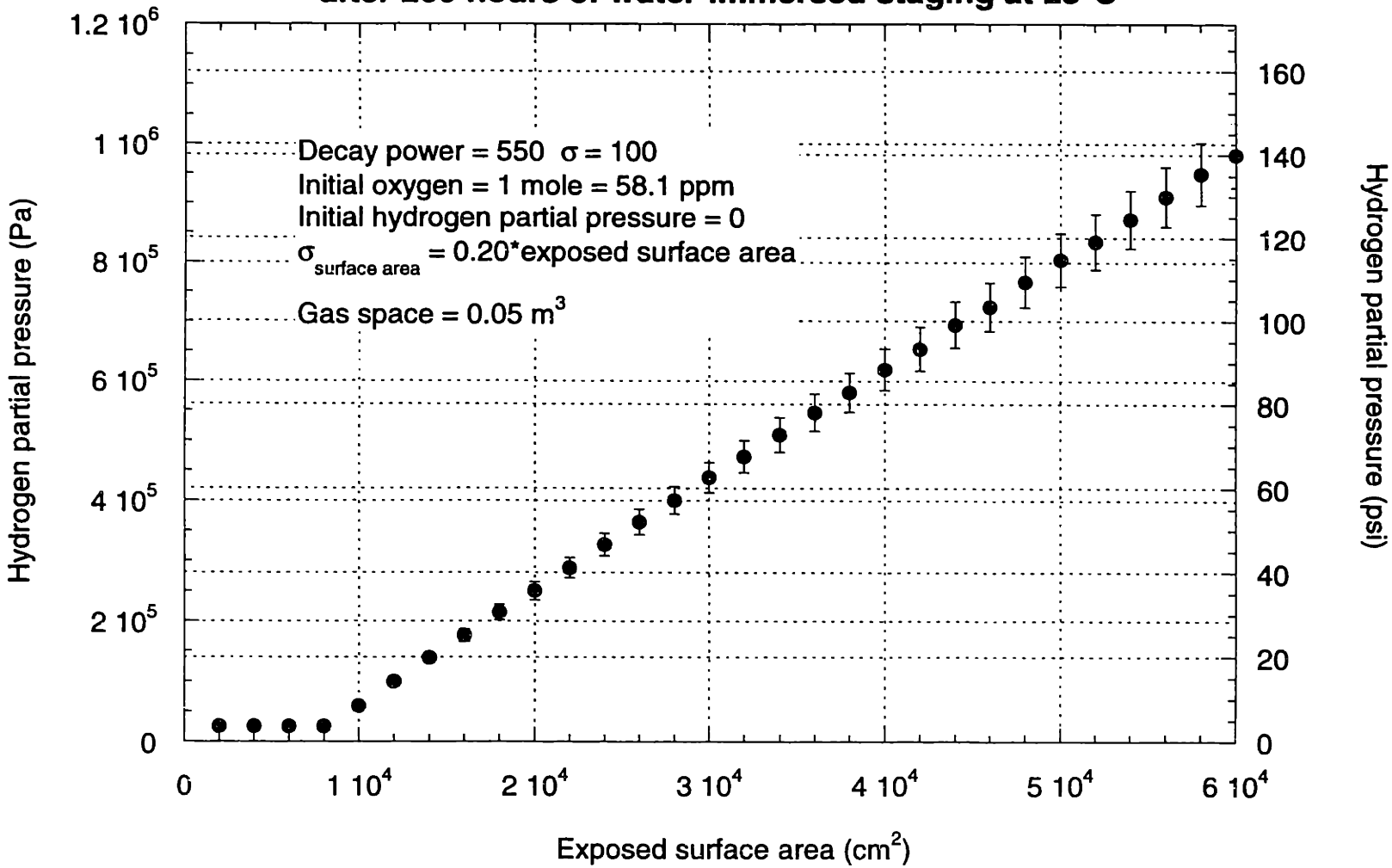


Figure 13: Total Pressure vs. Time during staging at different temperatures

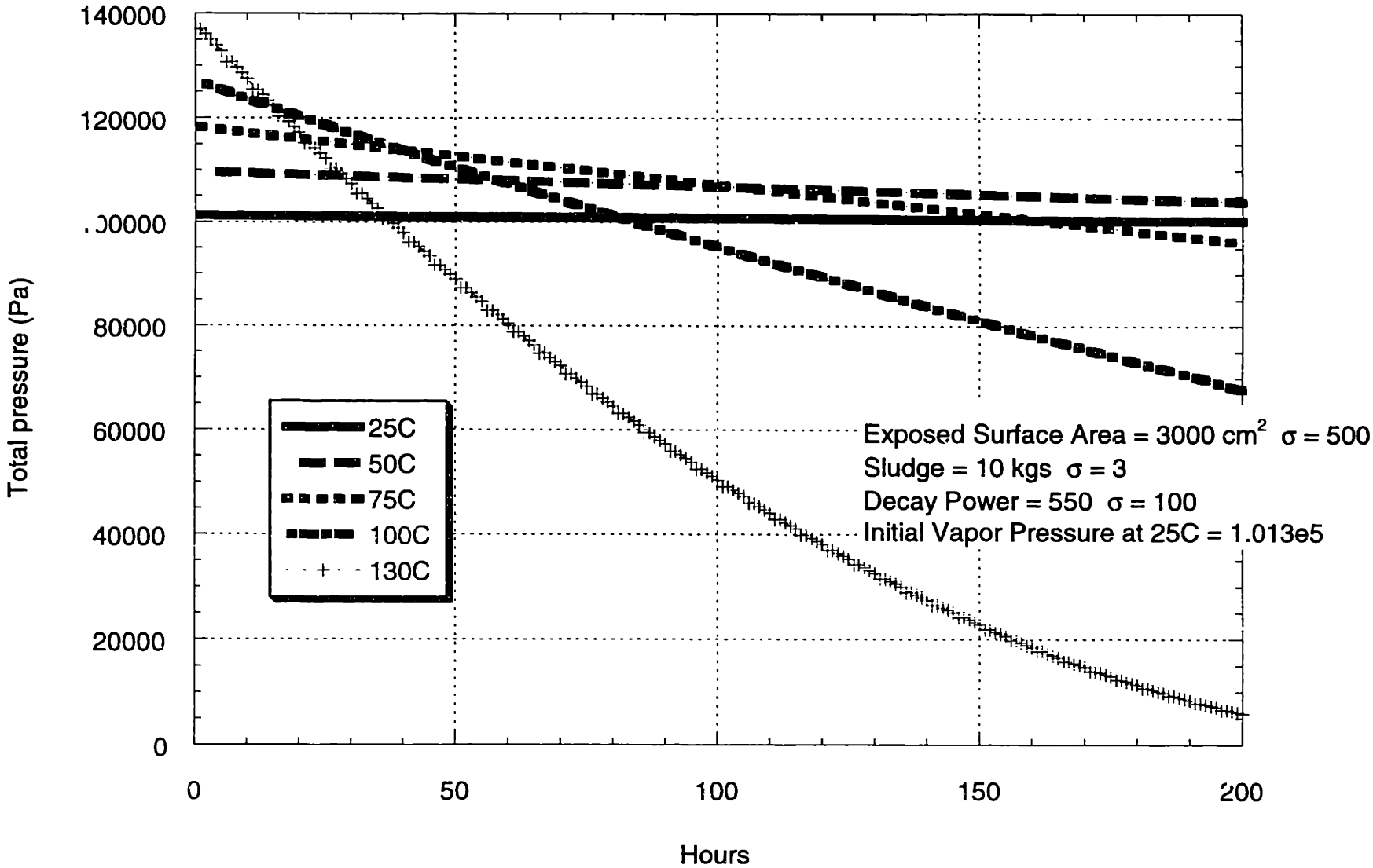


Figure 14: Total Pressure vs. Time during staging at 50°C

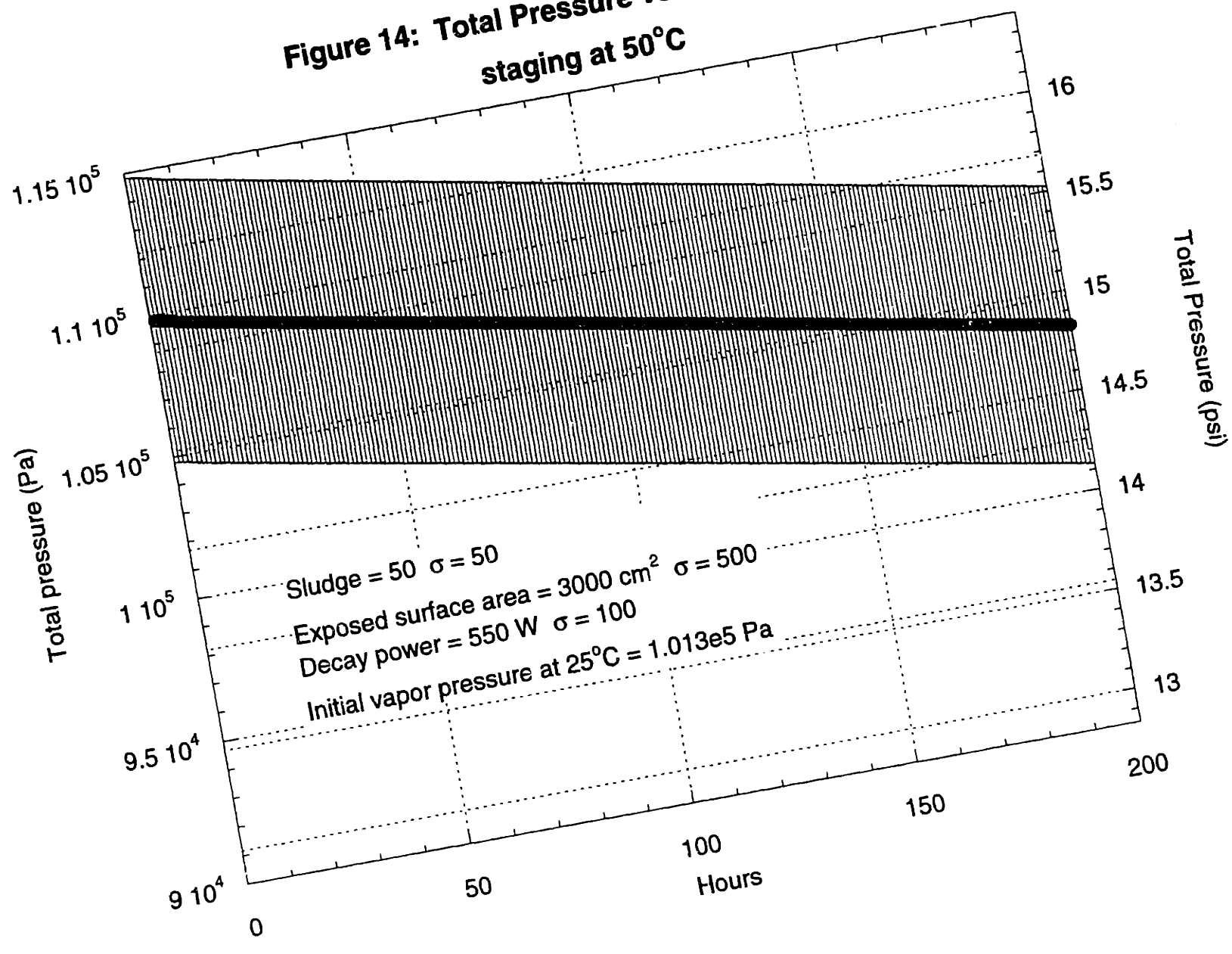


Figure 15: Oxygen Partial Pressure vs. Time during staging at 25°C

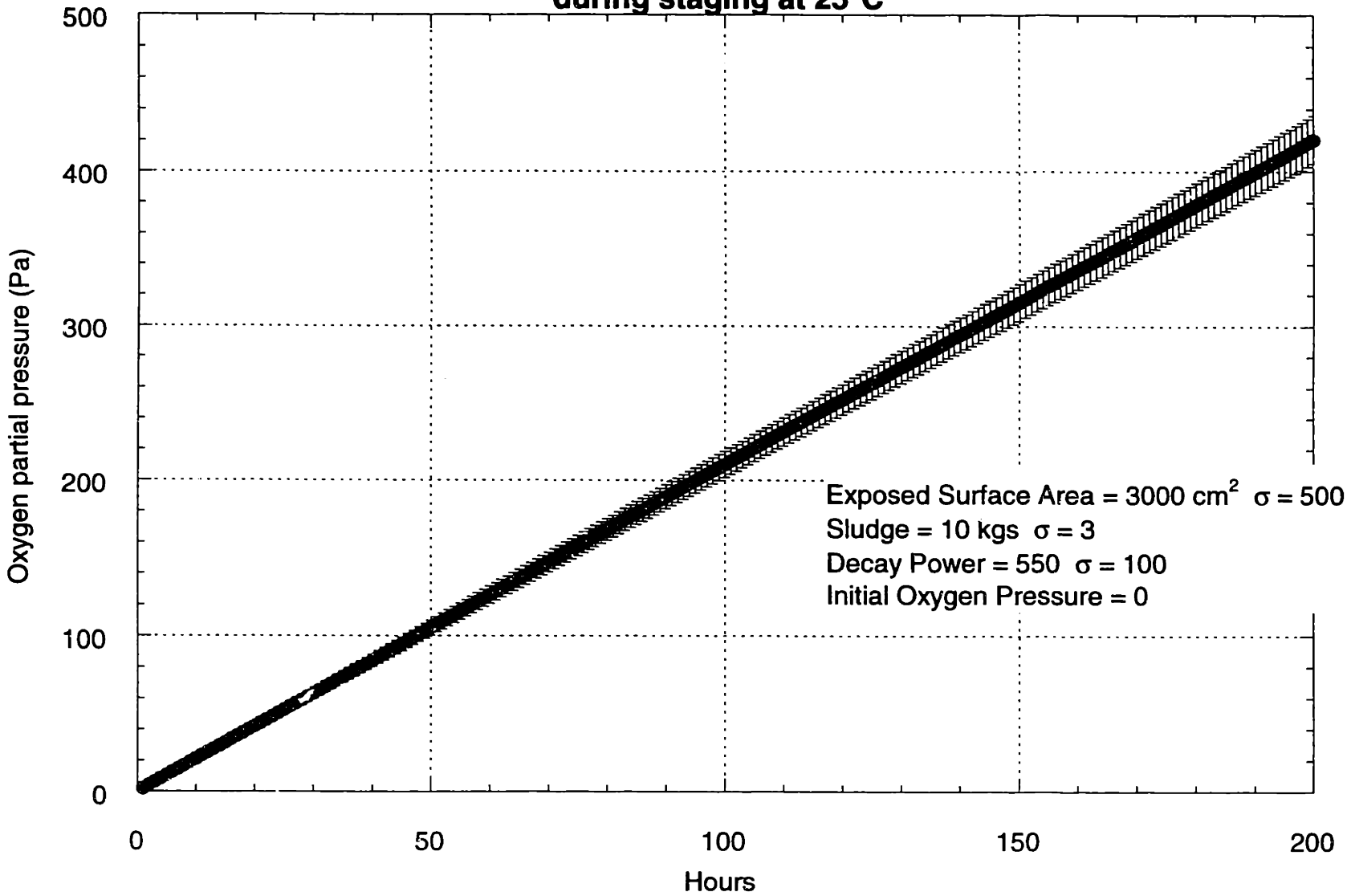


Figure 16: Water Vapor Pressure vs. Time during staging at 25°C

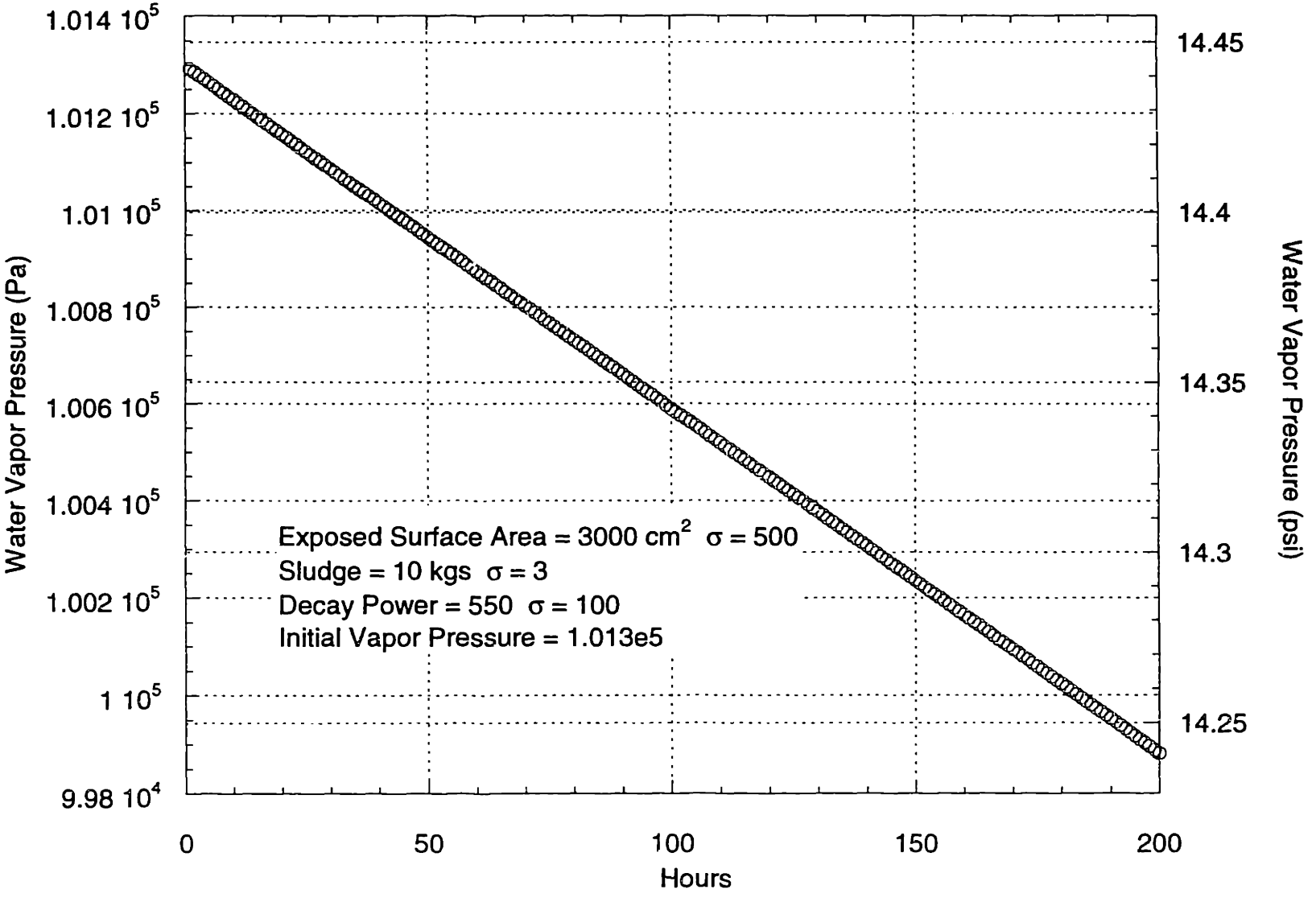


Figure 17: Water Vapor Pressure vs. Time during staging at 25°C

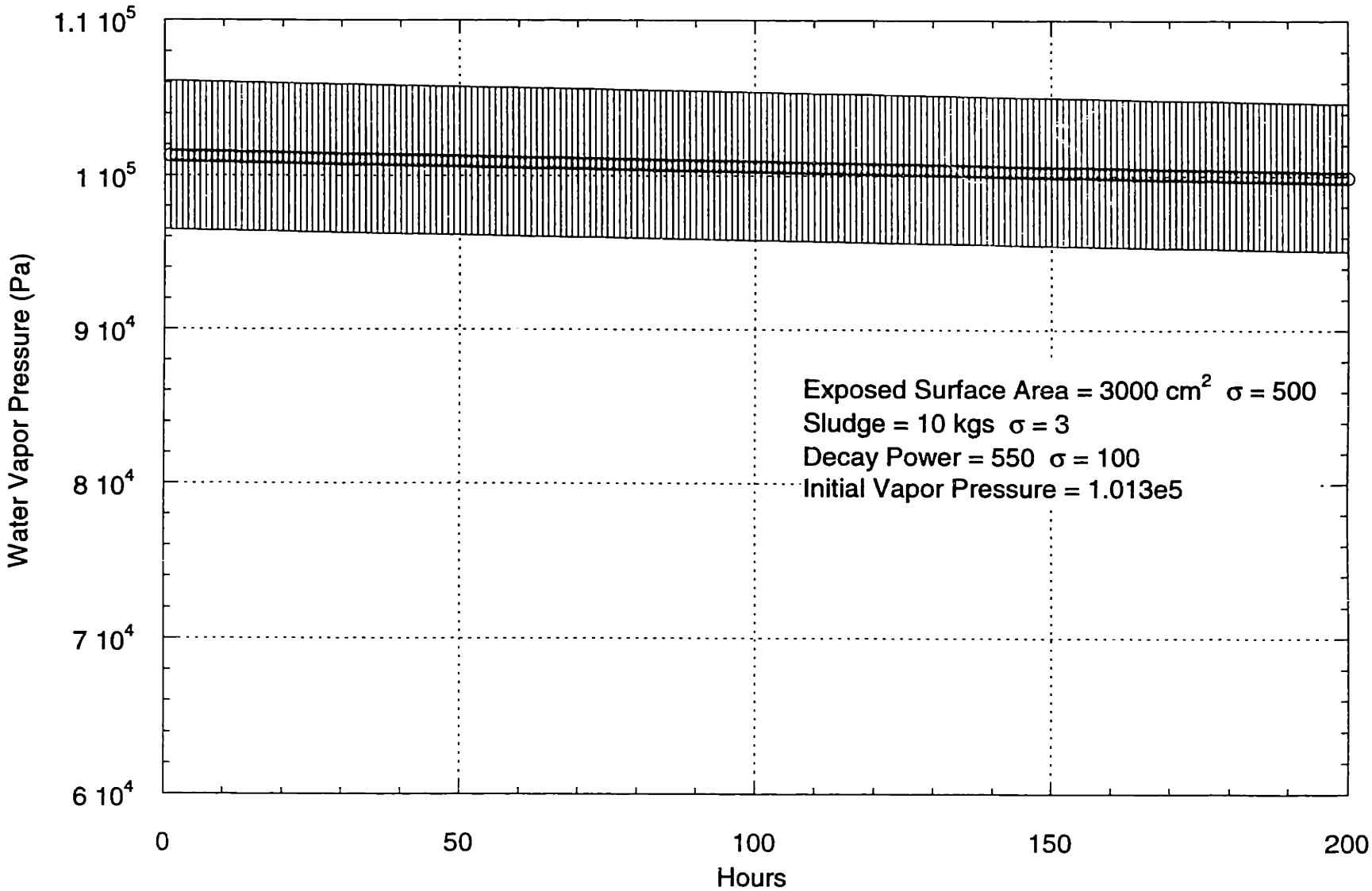


Figure 18: Water Vapor Pressure vs. Time during staging at 50°C

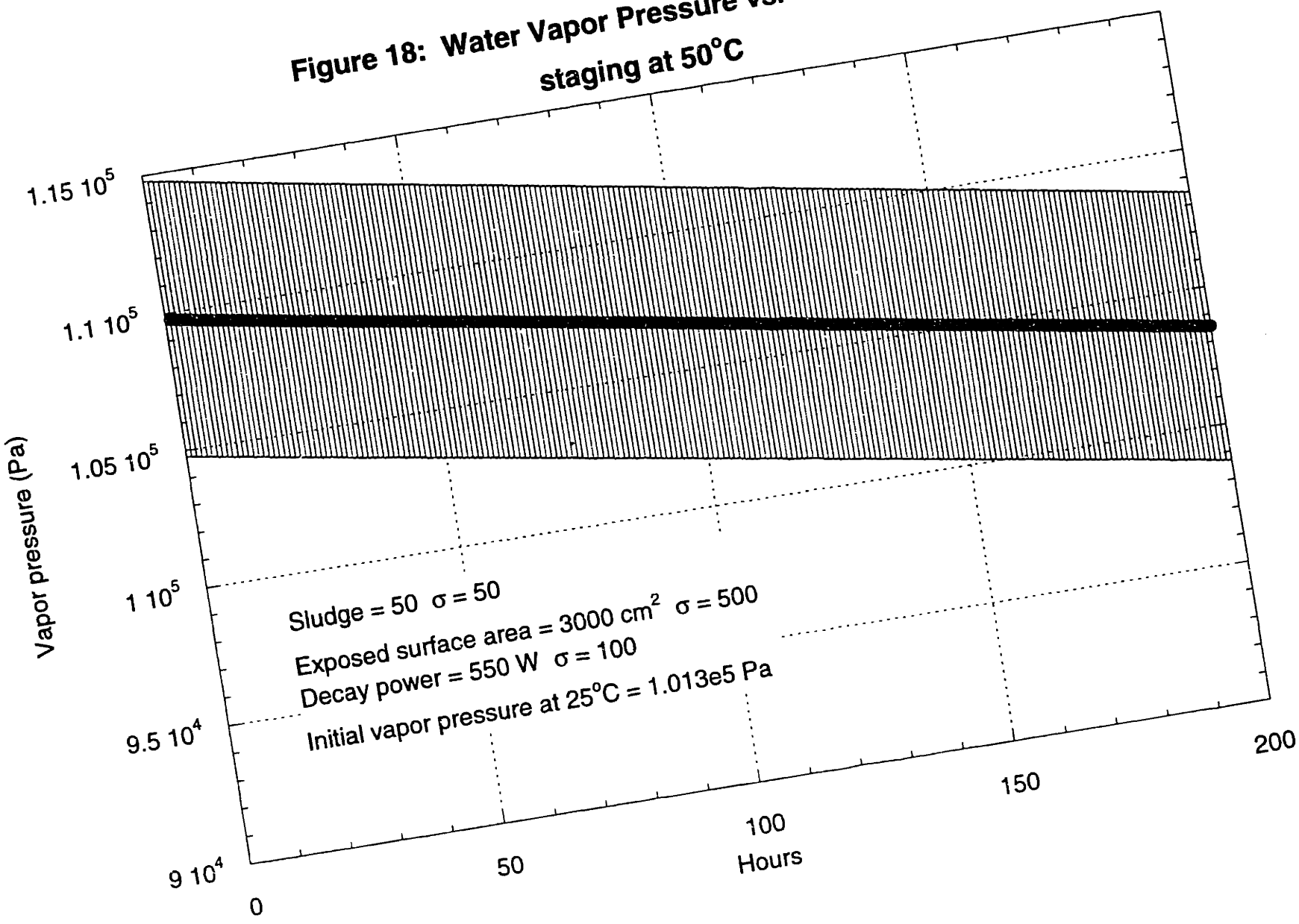


Figure 19: Hydrogen Partial Pressure vs. Time during staging at 25°C

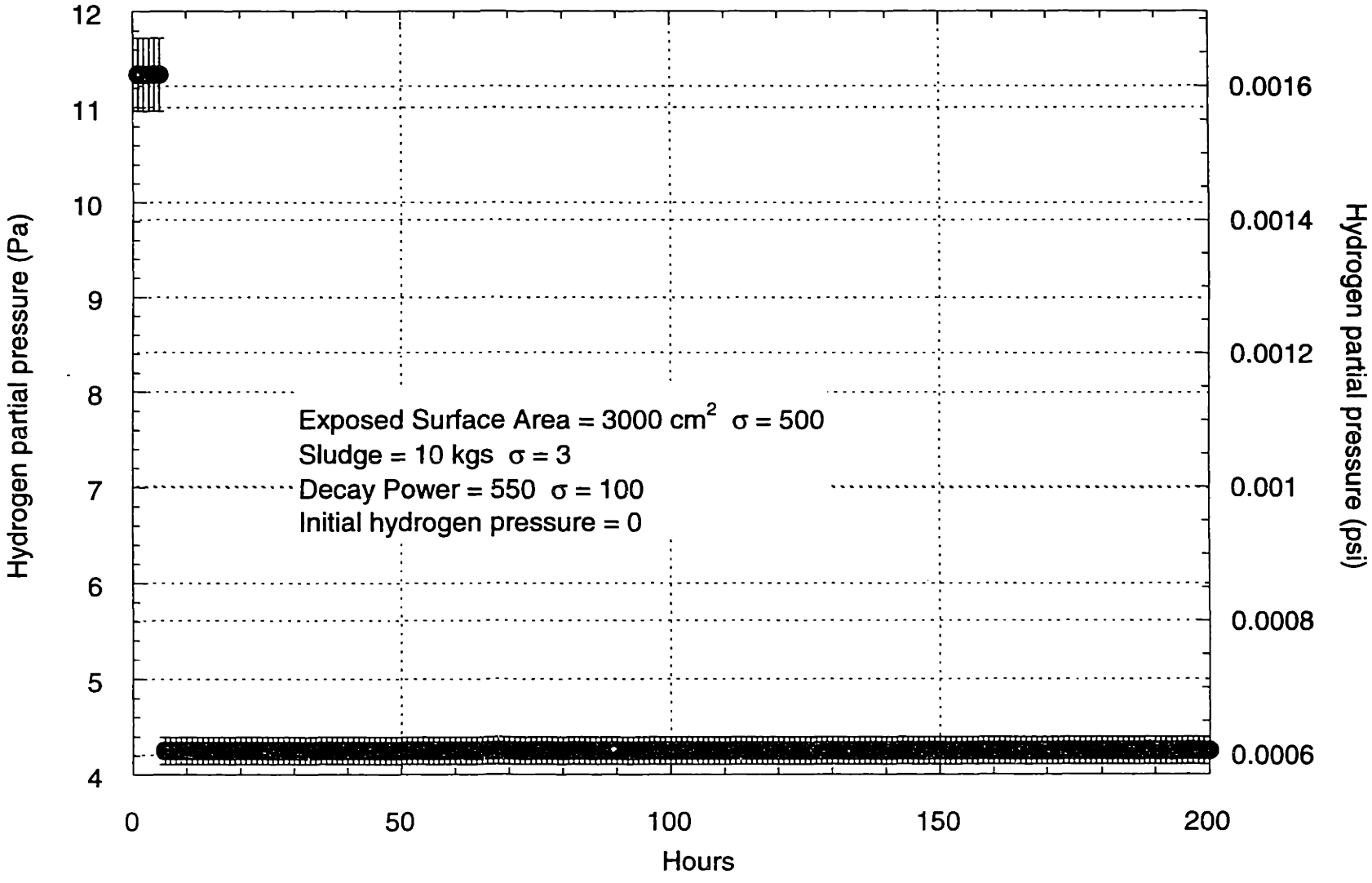


Figure 20: Oxygen Partial Pressure vs. Decay Power

Staging for 200 hours at 25°C

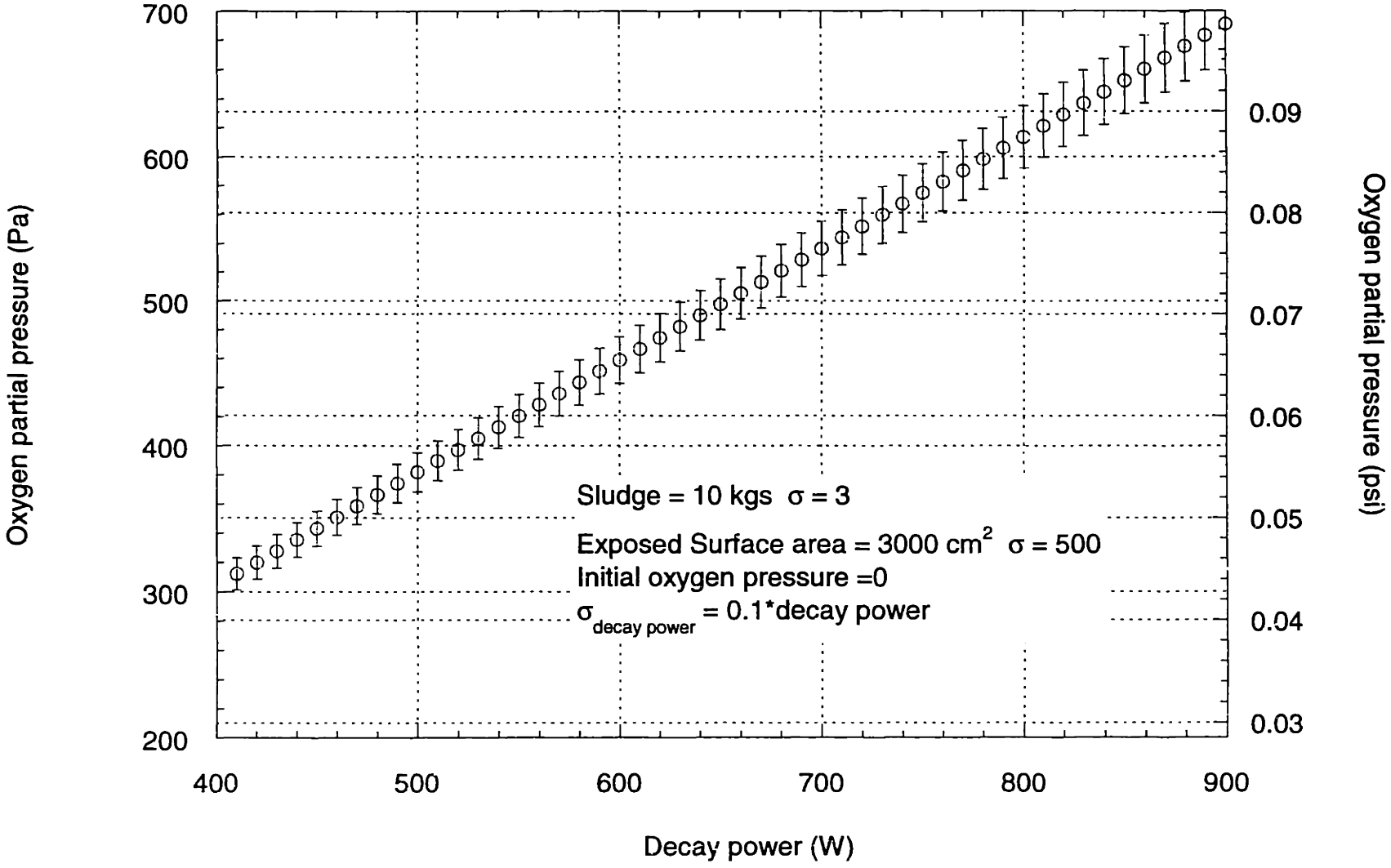


Figure 21: Hydrogen Partial Pressure vs. Decay Power

Staging for 200 hours at 25°C

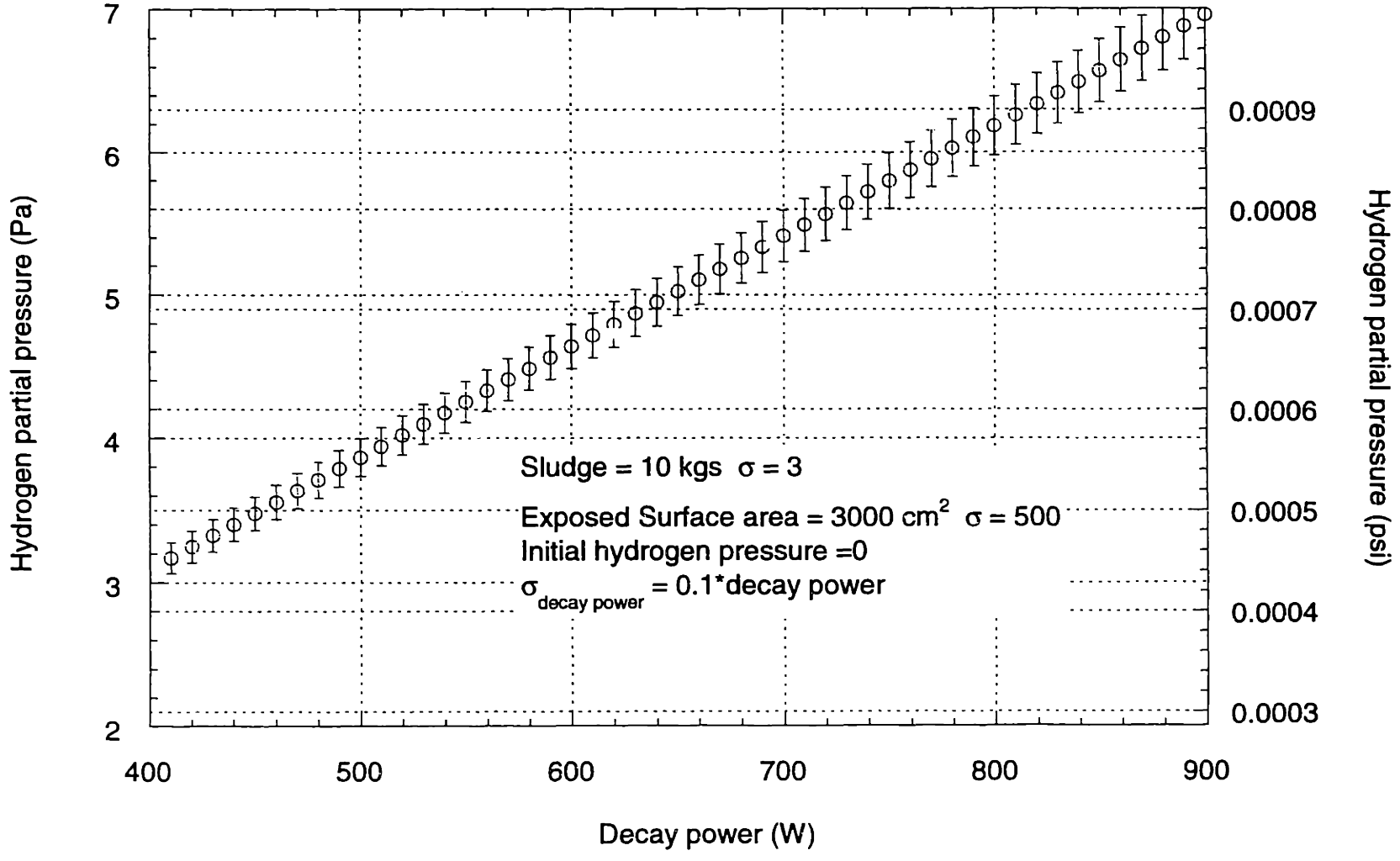


Figure 22: Total Pressure vs. Decay Power

Staging for 200 hours at 25°C

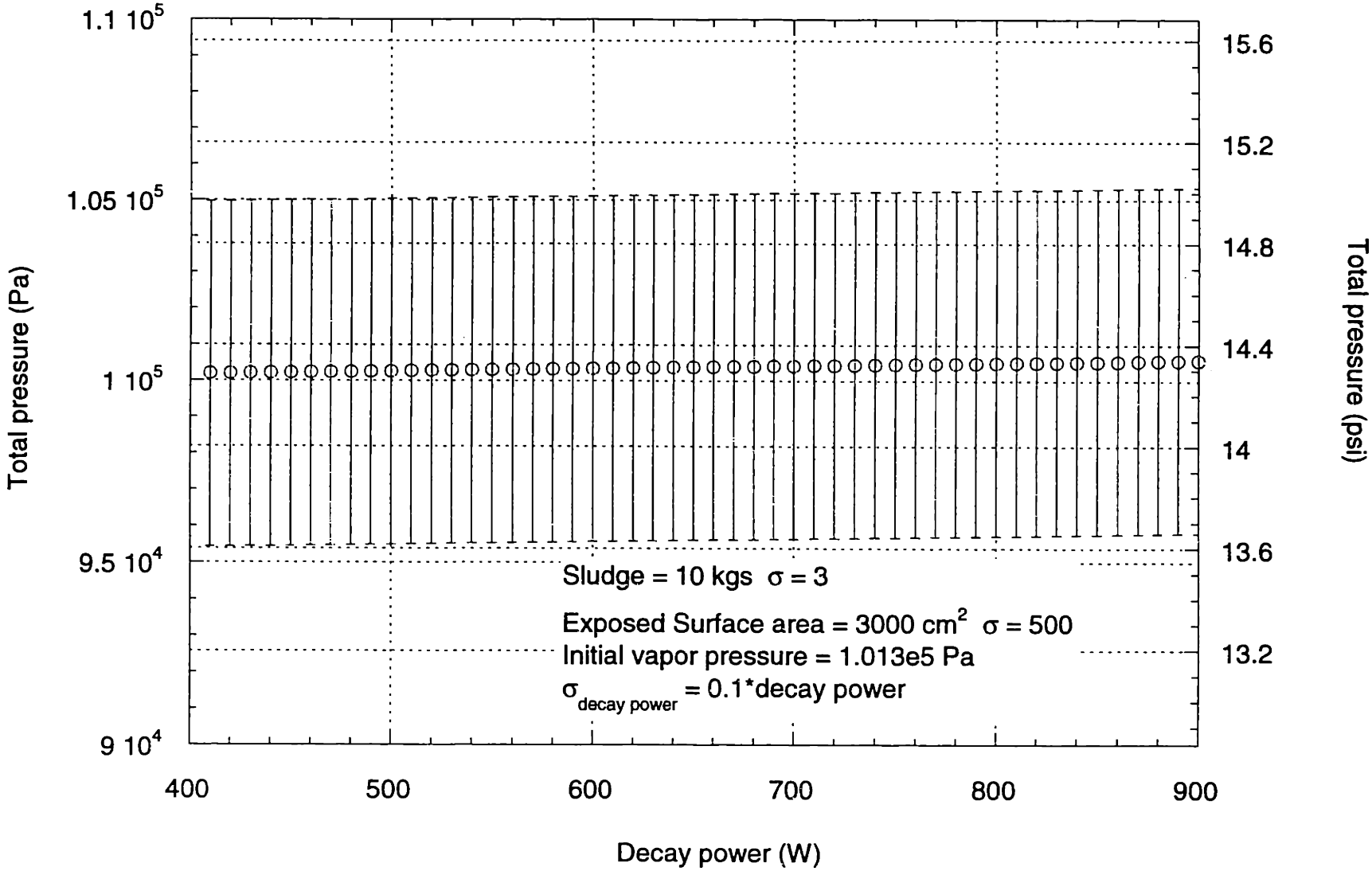


Figure 23: Total Pressure vs. Exposed Surface Area after Staging for 200 hours at 25°C

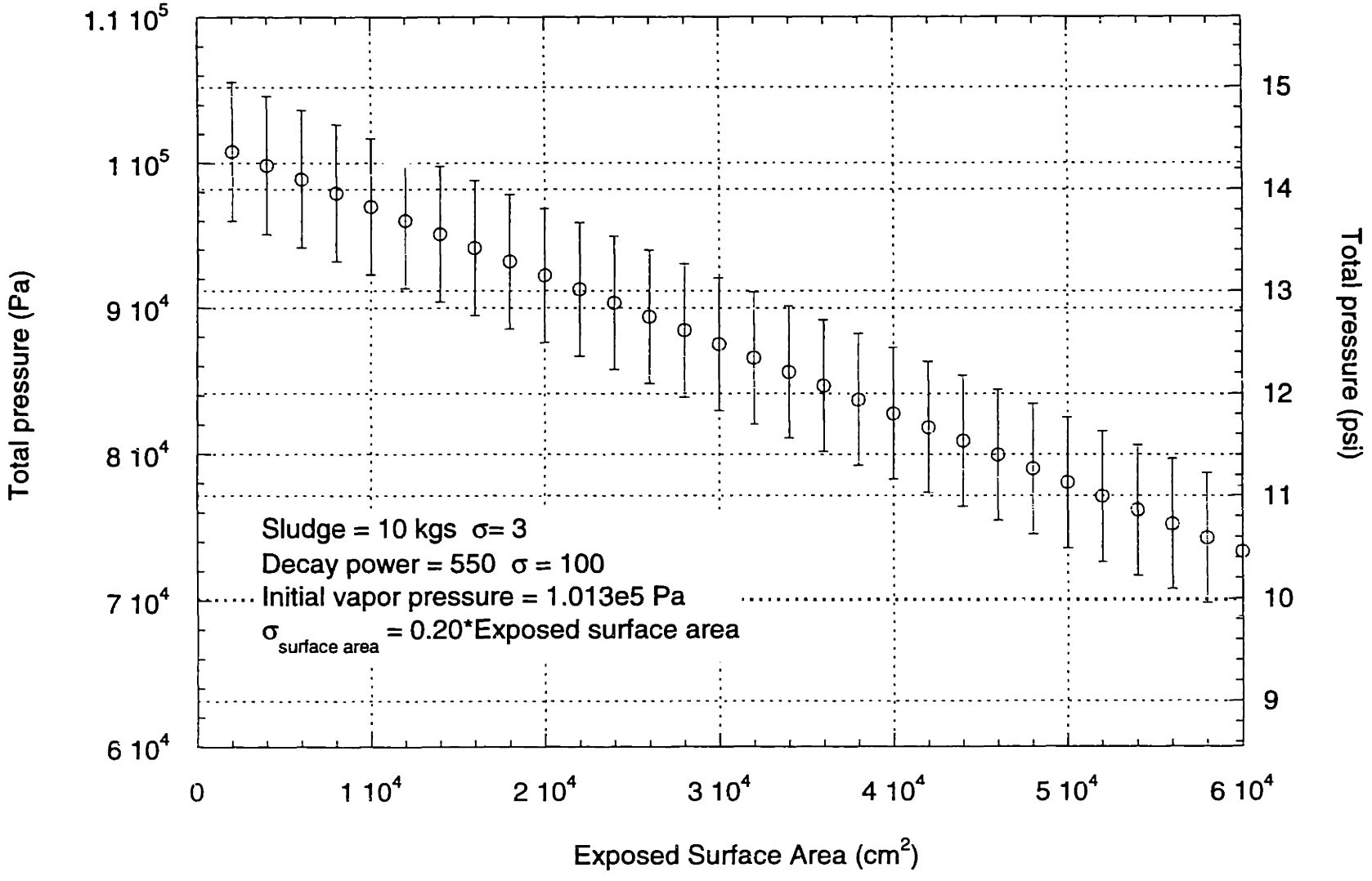


Figure 24: Total Pressure vs. Time during interim storage at several temperatures

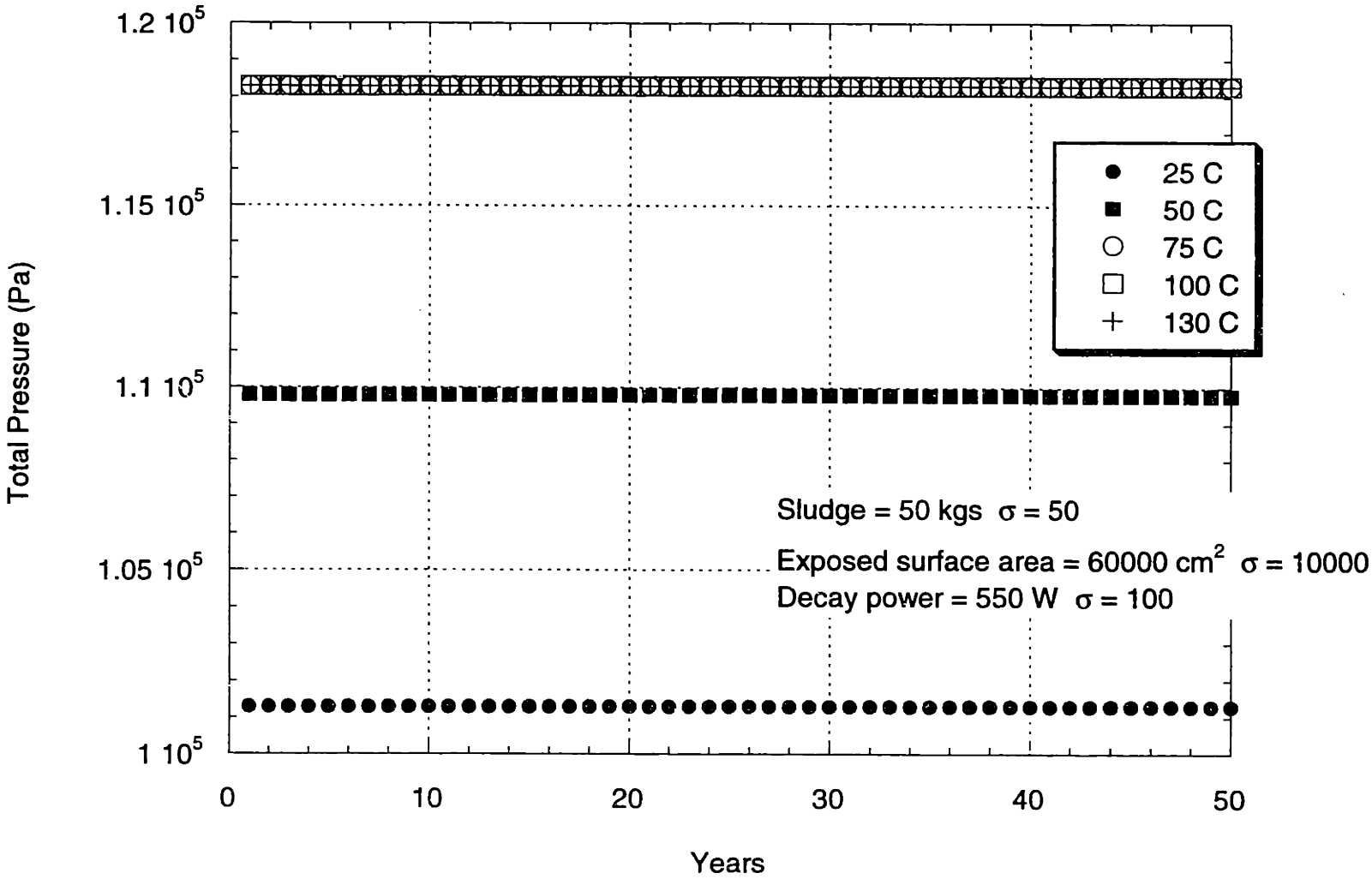


Figure 25: Total Pressure vs Time

During Interim Storage at 70°C

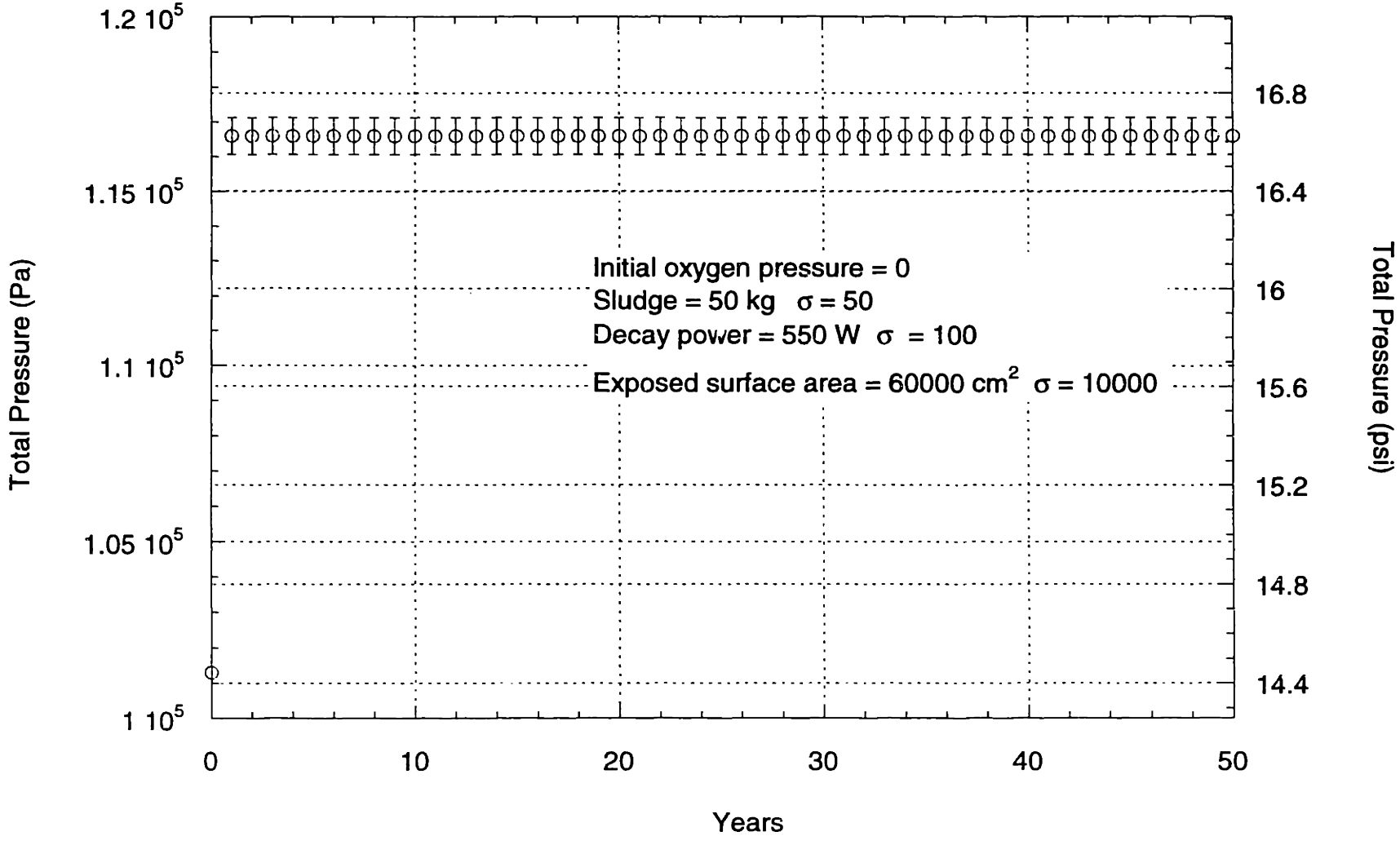


Figure 27: Oxygen Partial Pressure vs Time During Interim Storage at 70°C

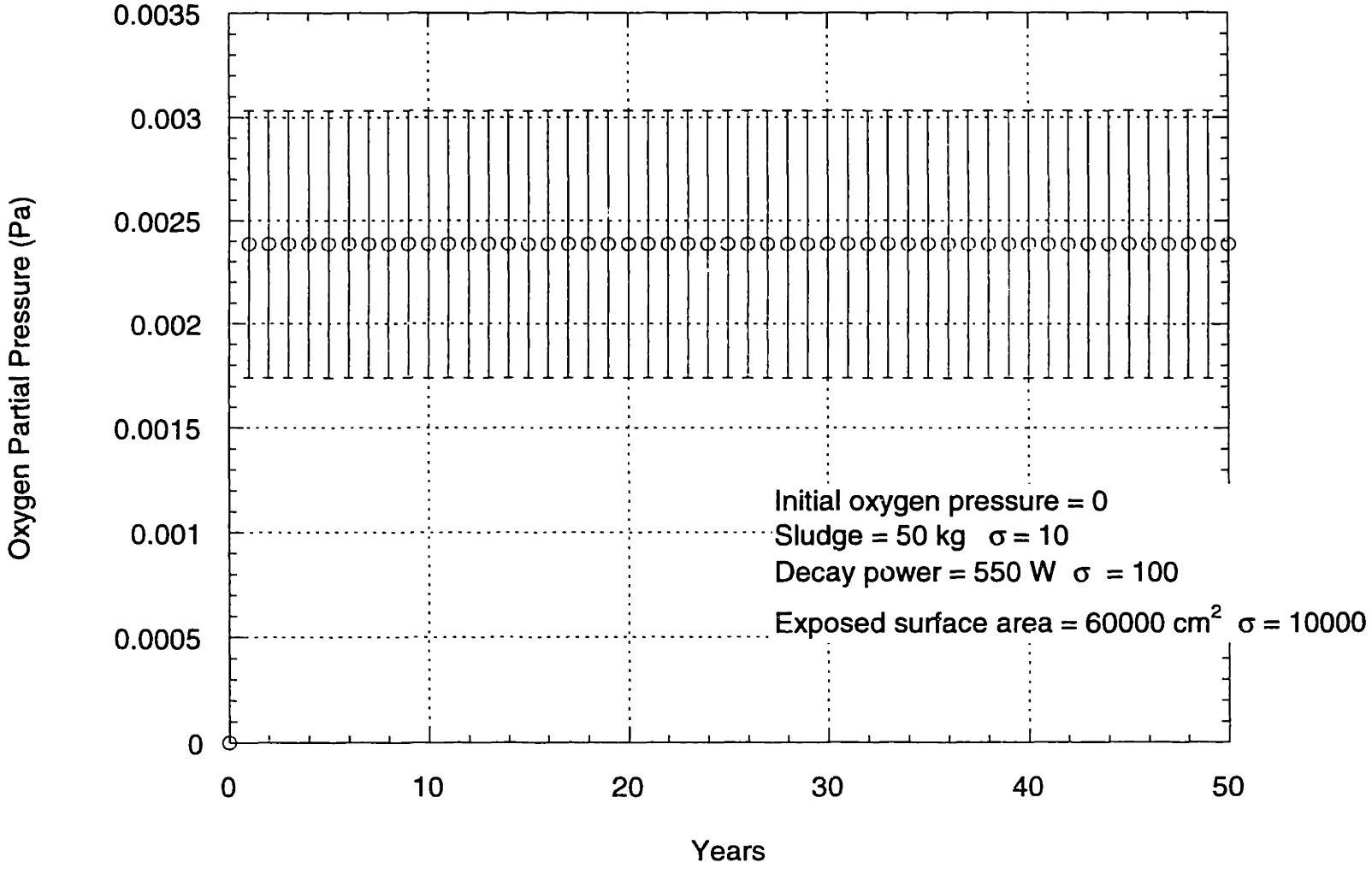


Figure 26: Hydrogen Partial Pressure vs. Time During Interim Storage at 70° C

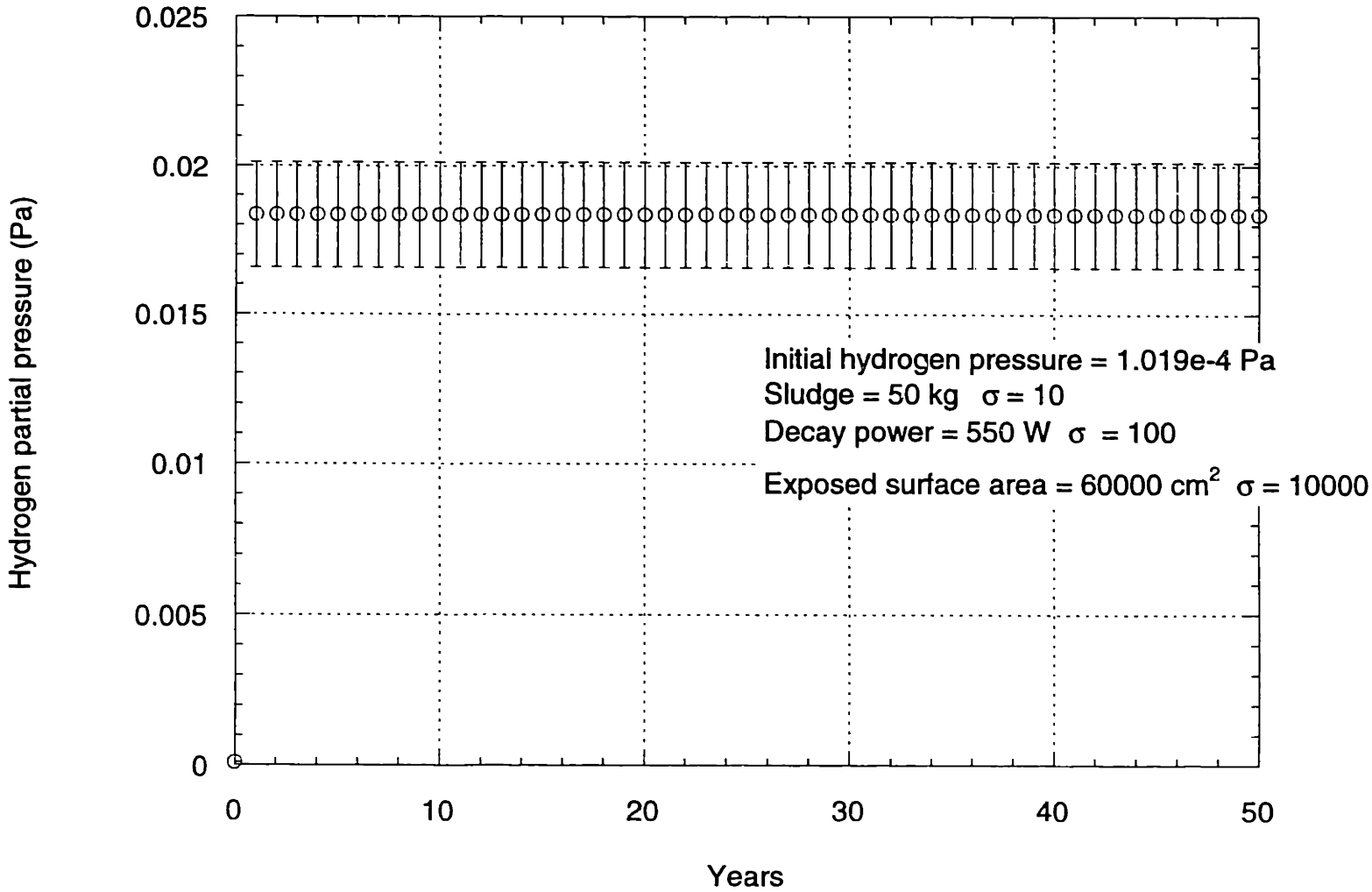


Figure 28: Total Pressure vs. Decay Power after 50 years of interim storage at 25°C

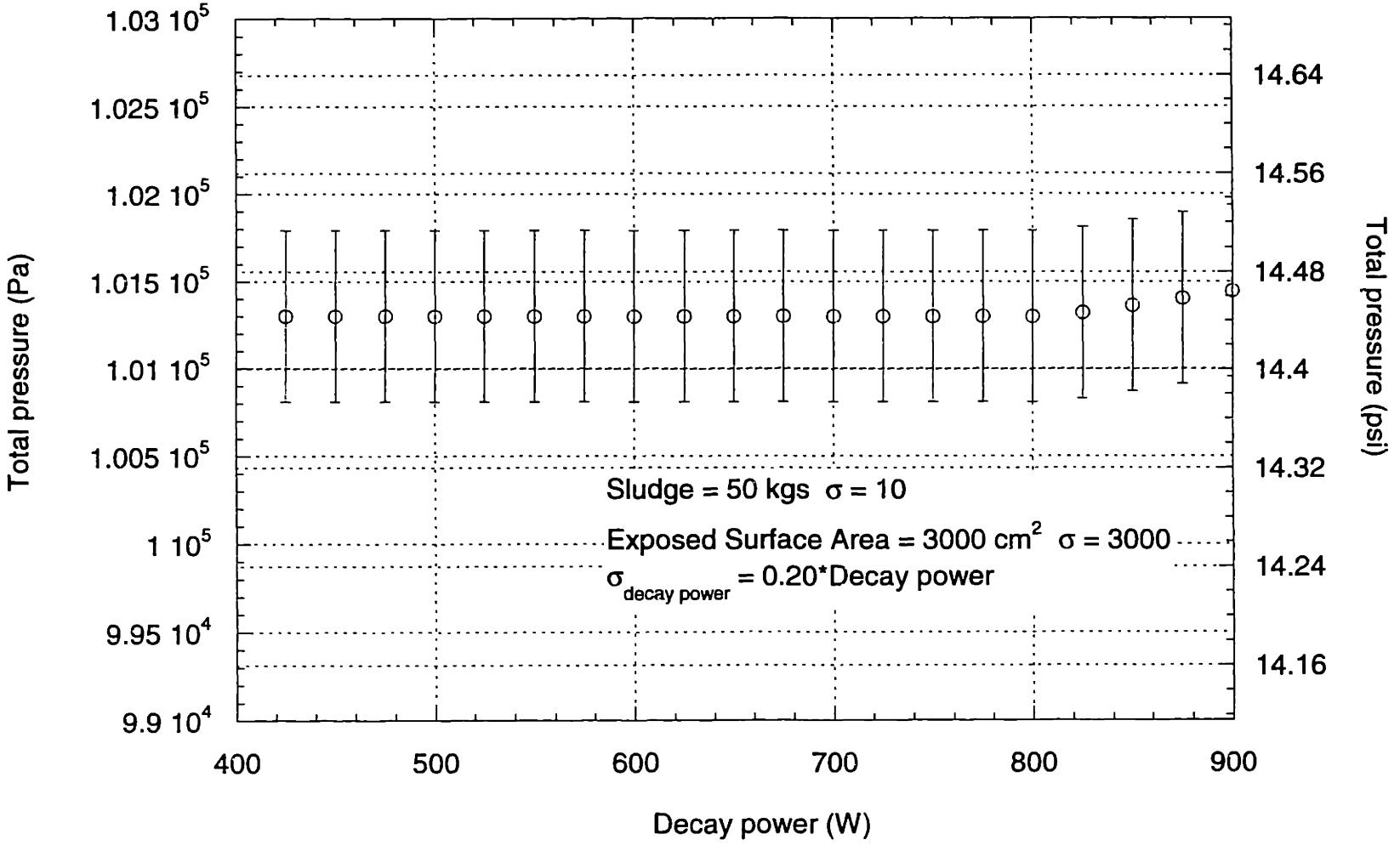


Figure 29: Oxygen Partial Pressure vs. Decay Power after 50 years of interim storage at 25°C

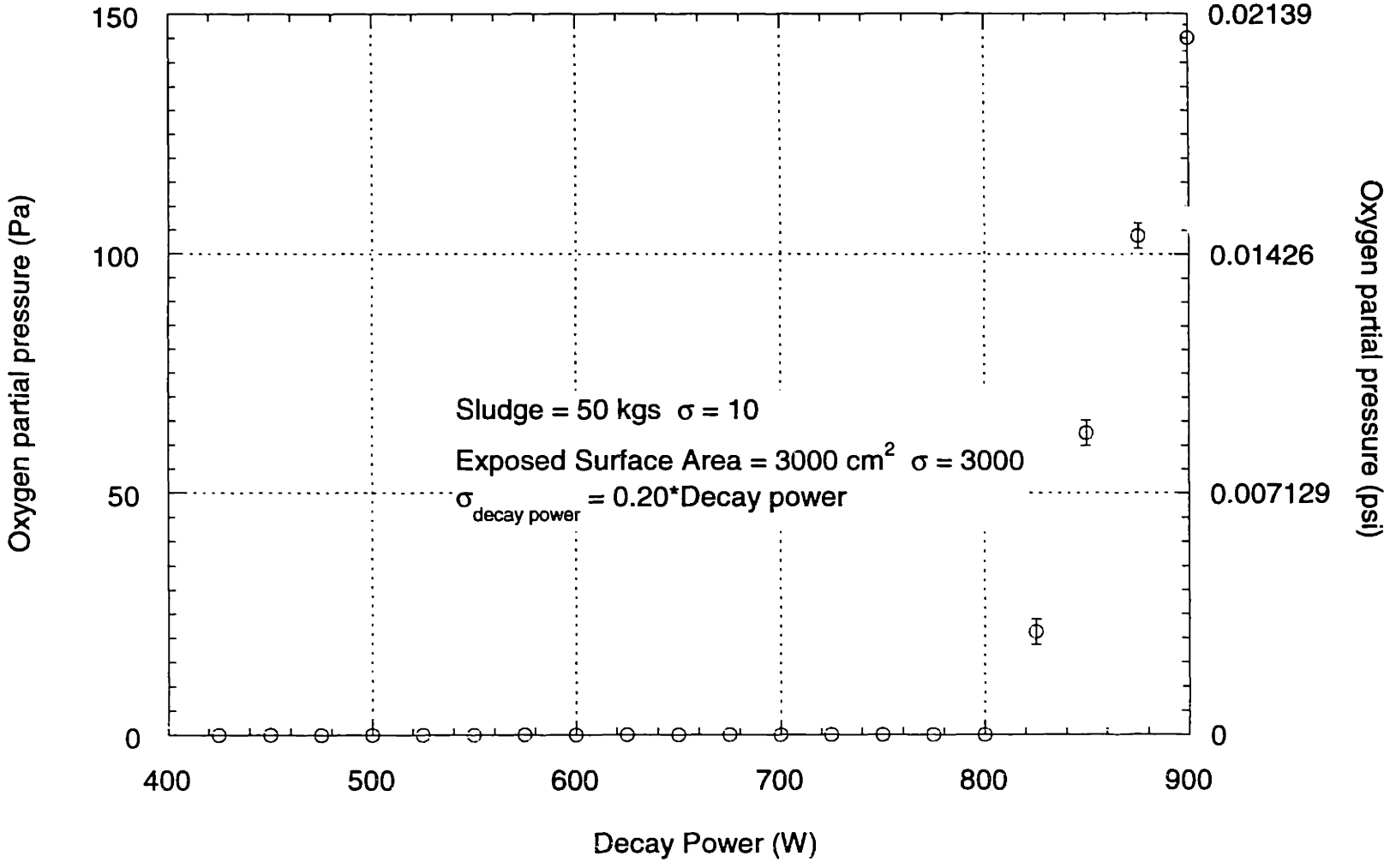


Figure 30: Hydrogen Partial Pressure vs. Decay Power after 50 years of interim storage at 25°C

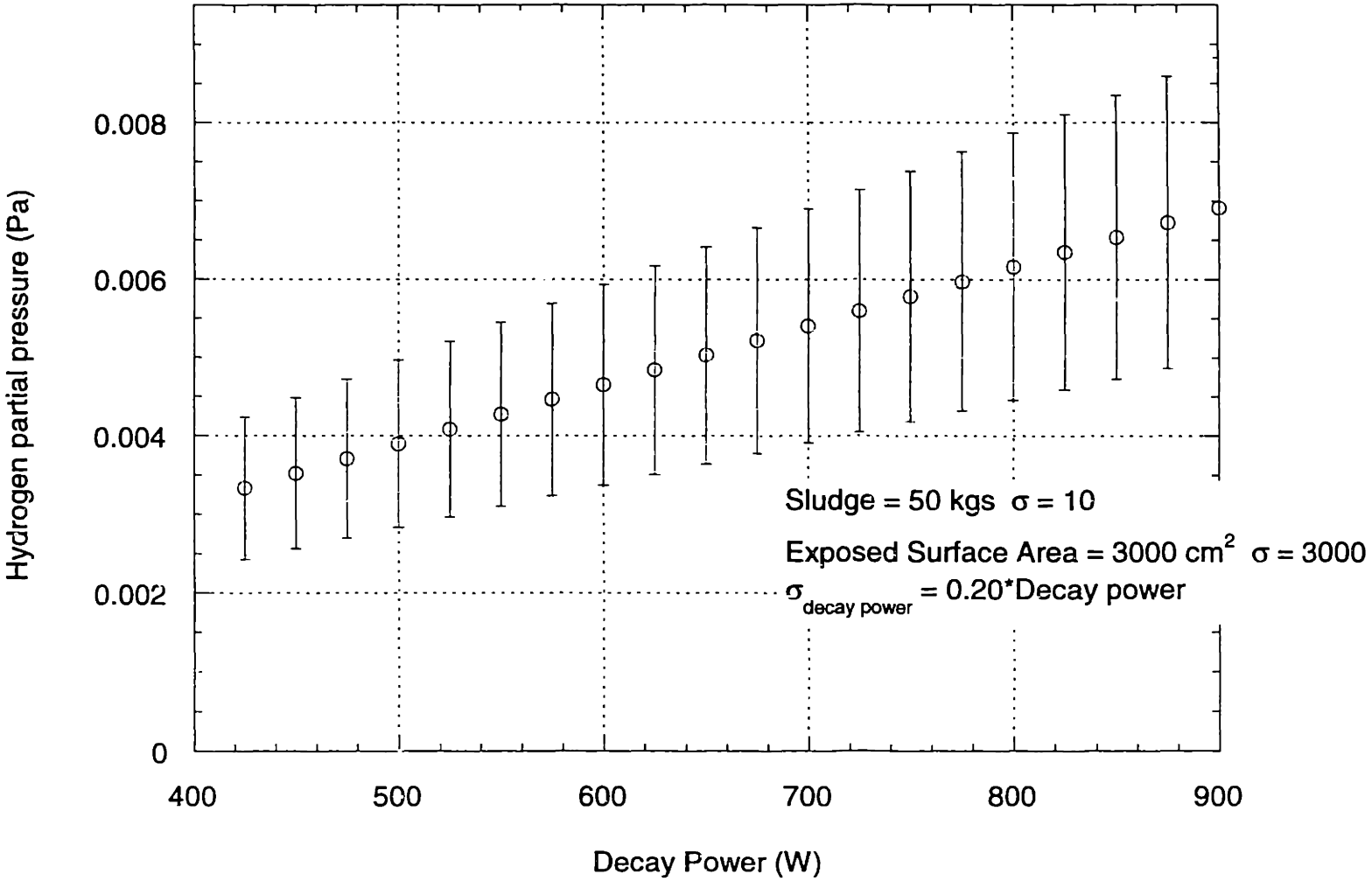


Figure 31: Oxygen Partial Pressure vs. Exposed Surface Area after 50 years of interim storage at 25°C

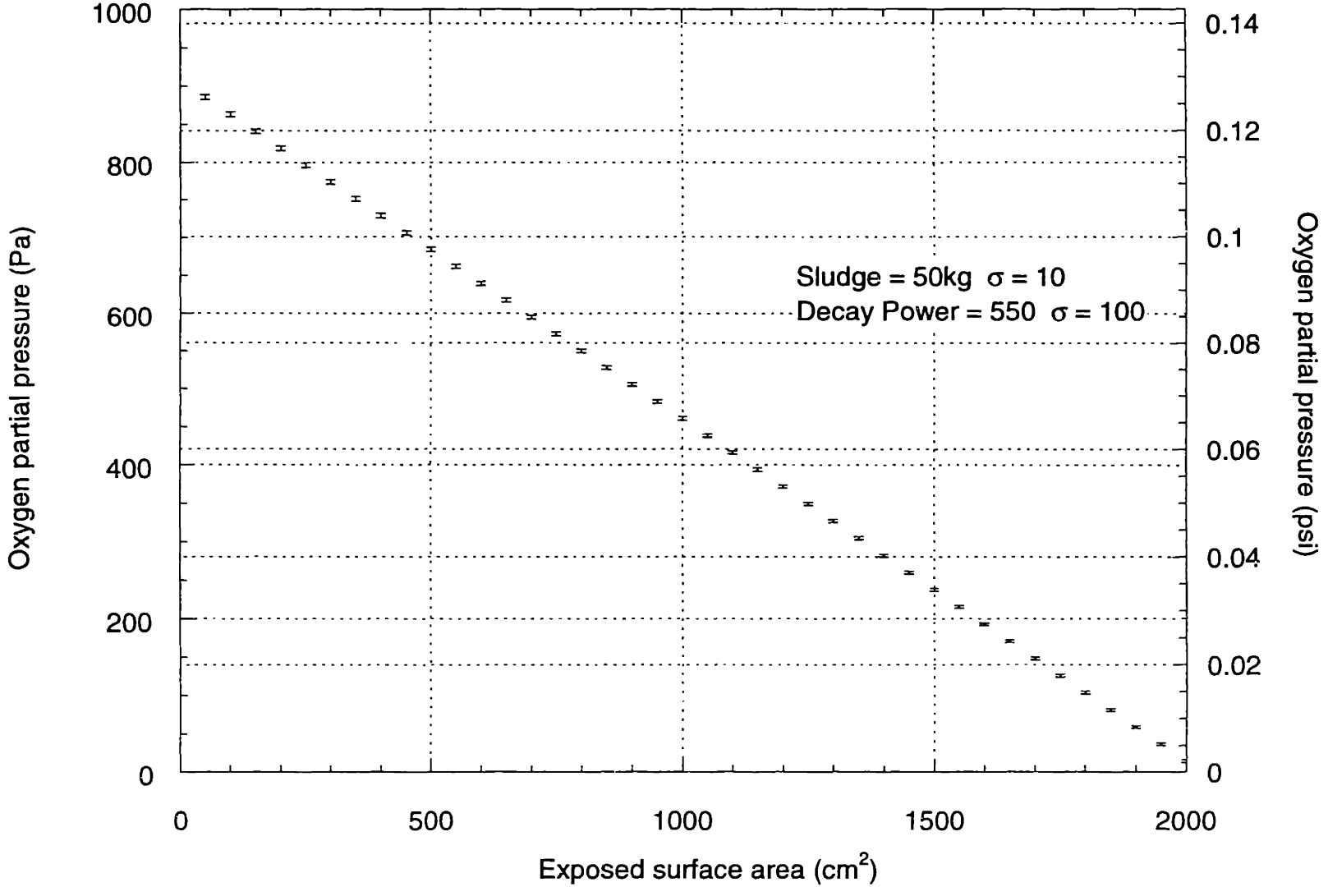


Figure 32: Hydrogen Partial Pressure vs Sludge

After 50 years of Interim Storage at 25°C

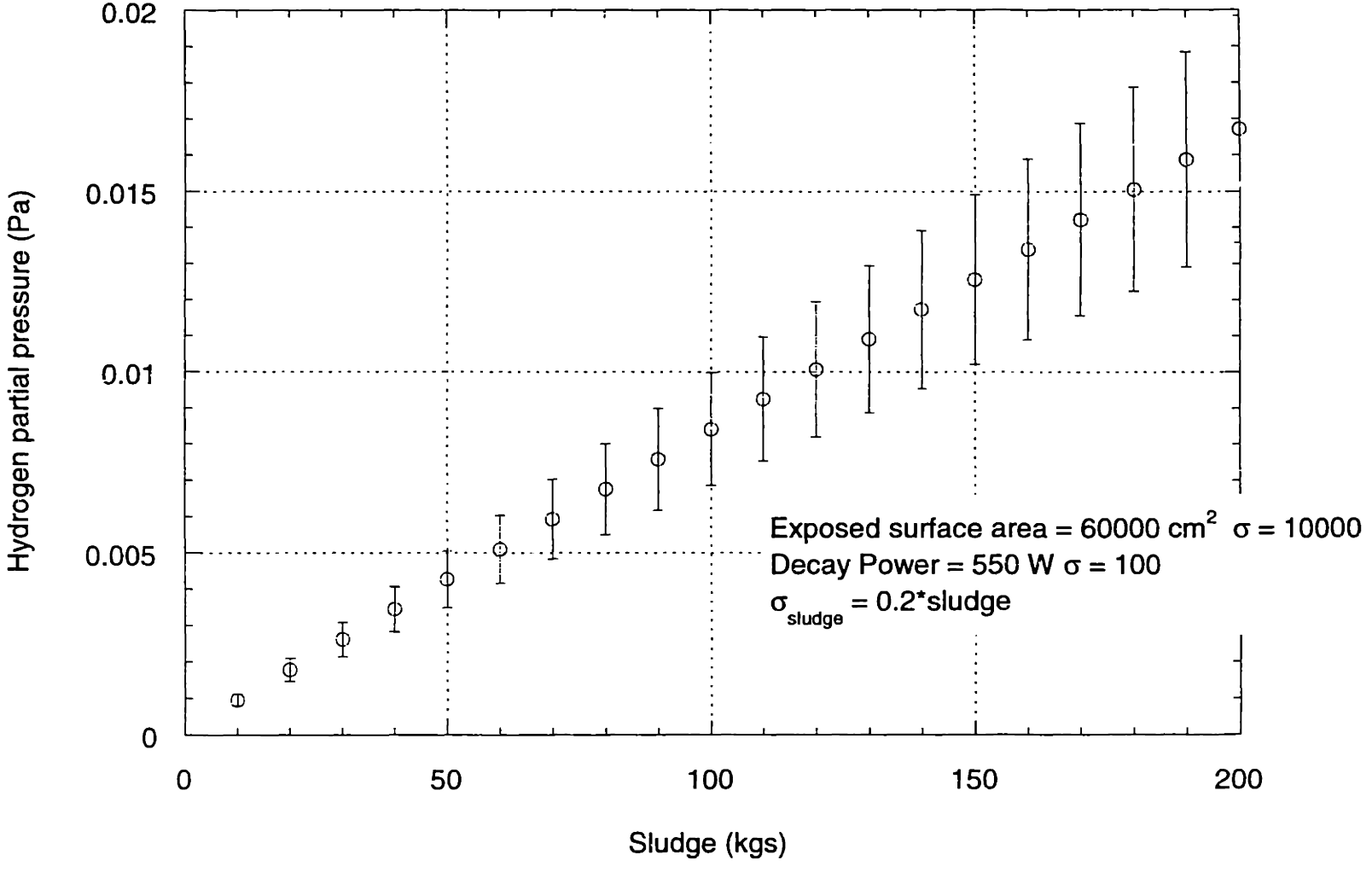
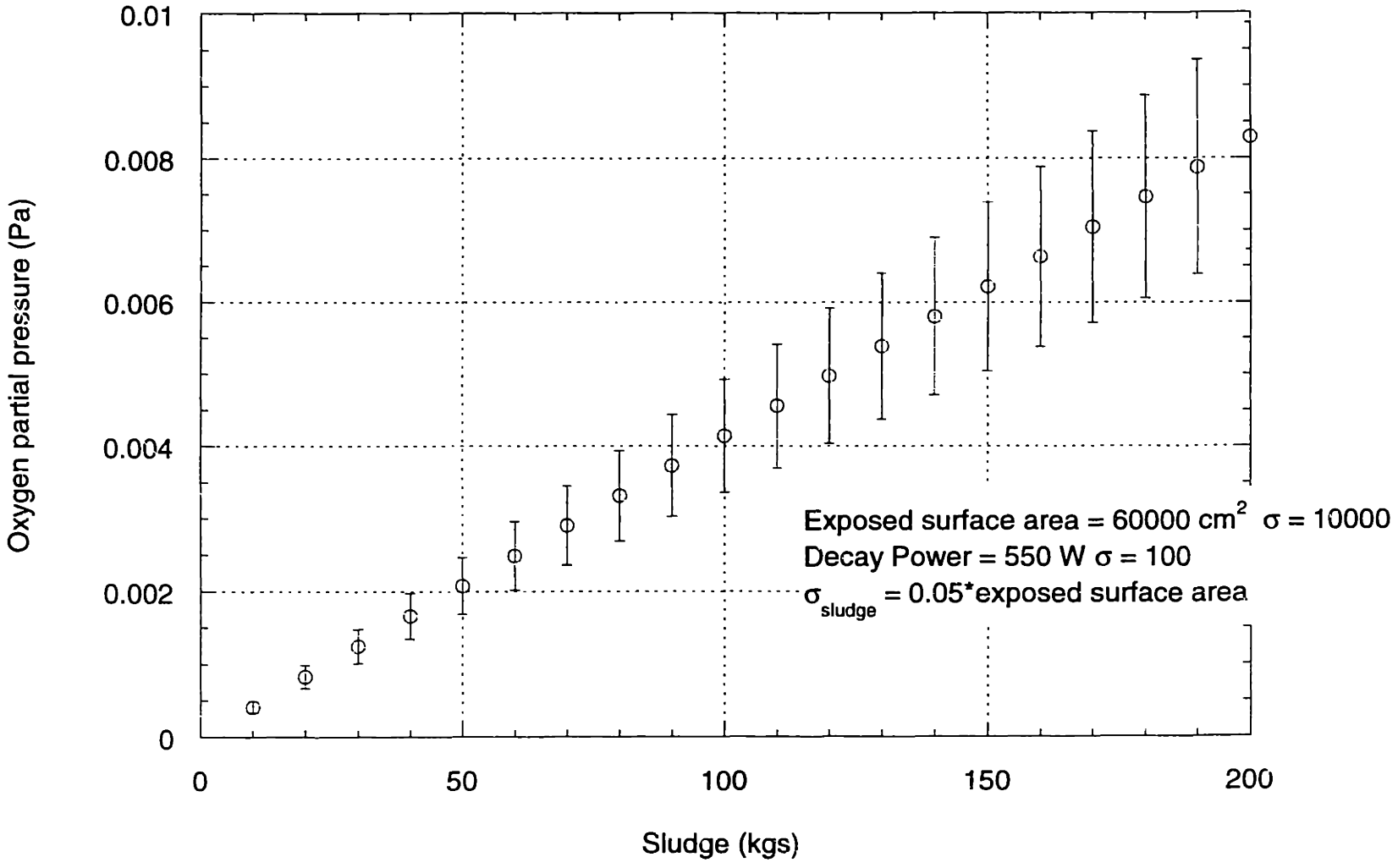


Figure 33: Oxygen Partial Pressure vs Sludge

After 50 years of Interim Storage at 25°C



Appendix A: User Instructions

For every model and subroutine, the first step is to assign each variable a data type and for user input or output values, a definition and appropriate units are given.

Water-immersed staging

At the beginning of the main program the user may input the following values;

- Temperature and standard deviation (K)
- Initial moles of oxygen in the water and standard deviation
- Initial hydrogen partial pressure and standard deviation (Pa)
- Gas volume (m^3)
- Amount of water and standard deviation (kgs)
- Sludge and standard deviation (kgs)
- Decay power and standard deviation (W)
- Exposed surface area (cm^2)
- Timestep for calculation loop (hours)
- Number of steps in loop

The user may also change the following parameters in other parts of the model:

- The reaction rate parameters and standard deviations may be changed in subroutine UIMMERSEDE for the deaerated case and UIMMERSEO2 for the oxygenated case
- The fraction of decay power that is deposited outside the fuel may be changed in subroutine RADIOL
- The fraction of decay power deposited outside the fuel which is absorbed in water may be changed in subroutine RADIOL
- The g-value for radiolysis may be changed in subroutine RADIOL

As currently written, the output is shown in columns in the following order: hours, moles of water, standard deviation of moles of water, hydrogen pressure, standard deviation of hydrogen pressure.

Water vapor-filled staging

At the beginning of the main program the user may input the following values;

- Temperature and standard deviation (K)
- Initial oxygen partial pressure and standard deviation (Pa)
- Initial water vapor pressure and standard deviation (Pa)
- Initial hydrogen partial pressure and standard deviation (Pa)

- Free volume (m^3)
- Free water and standard deviation (kgs)
- Sludge and standard deviation (kgs)
- Decay power and standard deviation (W)
- Exposed surface area (cm^2)
- Timestep for calculation loop (hours)
- Number of steps in loop

The user may also change the following parameters in other parts of the model:

- The reaction rate parameters and standard deviations may be changed in subroutine UVAPOR
- Fractions of bound and chemisorbed water in the sludge may be changed in subroutine WATER
- The fraction of decay power that is deposited outside the fuel may be changed in subroutine RADIOL
- The fraction of decay power deposited outside the fuel which is absorbed in water may be changed in subroutine RADIOL
- The g-value for radiolysis may be changed in subroutine RADIOL

As currently written, the output is shown in columns in the following order: hours, water vapor pressure, standard deviation of water vapor pressure, hydrogen partial pressure, standard deviation of hydrogen partial pressure, oxygen partial pressure, standard deviation of oxygen partial pressure, total pressure, standard deviation of total pressure.

Interim Storage in Dry Oxygen

At the beginning of the main program the user may input the following values:

- Temperature and standard deviation (K)
- Initial oxygen partial pressure and standard deviation (Pa)
- Initial total pressure and standard deviation (Pa)
- Free volume (m^3)
- Sludge and standard deviation (kgs)
- Free H_2O and standard deviation (kgs, should be zero for this case)
- Decay power and standard deviation (W)
- Exposed surface area (cm^2)
- Timestep for calculation loop (hours)
- Number of steps in loop

The user may also change the following parameters in other parts of the model:

- The reaction rate parameters and standard deviations may be changed in subroutine UDRYAIR
- The fraction of water that is chemisorbed in sludge and standard deviation may be changed in subroutine WATER
- The g-value for radiolysis may be changed in subroutine RADIOL

As currently written, the output is shown in columns in the following order: hours, hydrogen pressure, standard deviation of hydrogen pressure, oxygen pressure, standard deviation of oxygen pressure, total pressure, standard deviation of total pressure.

Appendix B: Executable file and output for water-immersed staging regime

```
$FREEFORM

!PROGRAM NAME imm+rad.F90

!MODIFIED 7/19/97
!MODIFIED 7/8/97  FOR IMMERSED CASE CALCULATES CONSUMPTION OF O2 FOR AERATED WATER
! AND CONSUMPTION OF WATER AND PRODUCTION OF H2 FOR DEAEPATED CASE
!MODIFIED 7/13/97 TO ADD RADIOLYSIS

  DOUBLE PRECISION TEMP,SIGTEMP      !TEMPERATURE IN KELVIN,STD DEV
  DOUBLE PRECISION SAEXP,SIGSAEXP    !TOTAL EXPOSED SURFACE AREA,CM^2
  DOUBLE PRECISION UCORR,SIGUCORR    !TOTAL CORROSION IN MG
  DOUBLE PRECISION MOLESH2,SIGMOLESH2    !MOLES OF H2 PRODUCED DURING CORROSION IN WATER, MOLES/HR
  DOUBLE PRECISION MOLESO2,SIGMOLESO2    !MOLES OF O2 IN IMMERSED CASE, STD DEV
  DOUBLE PRECISION O2LIM             !CUTOFF FOR DEAERATED REACTION, PPM,FROM ITA REPORT, APPENDIXG
  DOUBLE PRECISION FREEH2O,SIGFREEH2O    !KGS OF H2O IN CANISTER ASSUMES 23" DIAMETER, FUEL DIAM OF 1
4",
                    !HEIGHT OF 140", REST IS WATER,
  DOUBLE PRECISION FREEMOLES,SIGFREEMOLES    !MOLES OF FREE WATER
  DOUBLE PRECISION SLUDGE,SIGSLUDGE !AMOUNT OF SLUDGE IN MCO, KGS
  DOUBLE PRECISION MOLESH2O,SIGMOLESH2O !AMOUNT OF WATER AVAILABLE FOR RADIOLYSIS
  DOUBLE PRECISION DECAY,SIGDECAY    !DECAY HEAT IN WATTS
  DOUBLE PRECISION TIMESTEP          !CHOOSE TIMESTEP, INHOURS
  DOUBLE PRECISION NEWO2,SIGNEWO2,NEWH2O,SIGNEWH2O
  DOUBLE PRECISION NEWH2,SIGNEWH2
  DOUBLE PRECISION H2CORRPROD,SIGH2CORRPROD      !MOLES OF H2 PRODUCED DURING CORROSION
  DOUBLE PRECISION H2OCORRCONS,SIGH2OCORRCONS    !MOLES OF H2O CONSUMED DURING CORROSION
  DOUBLE PRECISION O2CORRCONS,SIGO2CORRCONS      !MOLES OF O2 CONSUMED DURING CORROSION
  DOUBLE PRECISION H2RADFROD,SIGH2RADPROD        !MOLES OF H2 PRODUCED DURING RADIOLYSIS
  DOUBLE PRECISION O2RADPROD,SIGO2RADPROD,H2ORADCONS,SIGH2ORADCONS
  DOUBLE PRECISION TEMP1,SIGTEMP1
  DOUBLE PRECISION FREEVOL,PH2,SIGPH2        !VOLUME IN MCO NOT TAKEN BY FUEL, PH2 IN Pa
  DOUBLE PRECISION GASVOL            !GAS VOLUME IN M^3
  INTEGER MO,YR,DAY,STEPS

OPEN(UNIT=2,FILE='C:\USERS\LISAS\OUTPUT\IMMERSE\IMM.TXT')
CALL GETDAT(YR,MO,DAY)

TEMP=323.D0
SIGTEMP=10.D0
O2LIM=0.129D0      !FROM ITA REPORT, APP.G
SLUDGE=10.D0
SIGSLUDGE=3.D0
DECAY=550.D0
SIGDECAY=100.D0
TIMESTEP=1.D0
STEPS=50
FREEVOL=0.6
GASVOL=0.05
PH2=0.D0
SIGPH2=0.D0

SAEXP=3000.D0
SIGSAEXP=500.D0
MOLESH2=0.0D0
SIGMOLESH2=0.0D0
MOLESO2=1.0D0
SIGMOLESO2=0.02D0
FREEH2O=550.D0
SIGFREEH2O=0.D0
FREEMOLES=FREEH2O*1.D3/18.D0
SIGFREEMOLES=SIGFREEH2O*1.D3/18.D0

!CALCULATE MOLES OF WATER AVAILABLE
CALL WATER(SLUDGE,SIGSLUDGE,FREEH2O,SIGFREEH2O,MOLESH2O,SIGMOLESH2O)

WRITE(2,*)'FILE GENERATED ON ',MO,'-',DAY,'-',YR

DO 10 I=1,STEPS

!CALCULATE O2 CONCENTRATION IN PPM
O2PPM=(MOLESO2*32.D3/(FREEMOLES*18.0D-3))

!CHECK TO SEE IF IN OXYGENATED OF DEAERATED REGIME
!O2-PRODUCED BY RADIOLYSIS
!H2-PRODUCED BY RADIOLYSIS AND CORROSION
!H2O-CONSUMED BY RADIOLYSIS AND CORROSION
IF (O2PPM .LT. O2LIM)THEN
    CALL UIMMERSDE(TEMP,SIGTEMP,TIMESTEP,SAEXP,SIGSAEXP,UCORR,SIGUCORR,H2CORRPROD,SIGH2CORRPROD,H2OC
ORRCONS,SIGH2OCORRCONS)
```

```
    CALL RADIOL(DECAY,SIGDECAY,TIMESTEP,H2RADPROD,SIGH2RADPROD,O2RADPROD,SIGO2RADPROD,H2ORADCONS,SIG
H2ORADCONS)

    !CALCULATE NEW MOLES OF H2O
    TEMP1=H2OCORRCONS+H2ORADCONS
    CALL STATSUM(H2OCORRCONS,1.D0,SIGH2OCORRCONS,H2ORADCONS,1.D0,SIGH2ORADCONS,0.D0,TEMP1,SIGTEMP1)
    IF(TEMP1 .LT. MOLESH2O)THEN
        NEWH2O=MOLESH2O-TEMP1
        CALL STATSUM(MOLESH2O,1.D0,SIGMOLESH2O,TEMP1,-1.D0,SIGTEMP1,0.D0,NEWH2O,SIGNEWH2O)
        MOLESH2O=NEWH2O
        SIGMOLESH2O=SIGNEWH2O

    !IF WATER CONSUMED IS MORE THAN THAT AVAILABLE, THEN MOLESH2PROD=MOLESH2O AT BEGINNING
    !OF TIMESTEP STD DEV FOR MOLESH2O IS SET ARBITRARILY
    ELSE
        H2CORRPROD=MOLESH2O
        SIGH2CORRPROD=SIGMOLESH2O
        H2RADPROD=0.D0
        SIGH2RADPROD=1.D-6
        O2RADPROD=0.D0
        SIGO2RADPROD=1.D-6
        MOLESH2O=0.D0
        SIGMOLESH2O=1.D-6
    ENDIF


    !CALC MOLES OF H2-FROM DEAERATED CORROSION AND RADIOLYSIS
    TEMP1=H2CORRPROD+H2RADPROD
    CALL STATSUM(H2CORRPROD,1.D0,SIGH2CORRPROD,H2RADPROD,1.D0,SIGH2RADPROD,0.D0,TEMP1,SIGTEMP1)
    NEWH2=MOLESH2+TEMP1
    CALL STATSUM(MOLESH2,1.D0,SIGMOLESH2,TEMP1,1.D0,SIGTEMP1,0.D0,NEWH2,SIGNEWH2)
    MOLESH2=NEWH2
    SIGMOLESH2=SIGNEWH2

    !CALCULATE MOLES OF O2-FROM LAST LOOP MOLES AND FROM RADIOLYSIS
    NEWO2=MOLESO2+O2RADPROD
    CALL STATSUM(MOLESO2,1.D0,SIGMOLESO2,O2RADPROD,1.D0,SIGO2RADPROD,0.D0,NEWO2,SIGNEWO2)
    MOLESO2=NEWO2
    SIGMOLESO2=SIGNEWO2


ELSE

    !O2-PRODUCED BY RADIOLYSIS, CONSUMED BY CORROSION
    !H2-PRODUCED BY RADIOLYSIS
    !H2O-CONSUMED BY RADIOLYSIS
    CALL UIMMERSO2(TEMP,SIGTEMP,TIMESTEP,SAEXP,SIGSAEXP,UCORR,SIGUCORR,O2CORRCONS,SIGO2CORRCONS)

    CALL RADIOL(DECAY,SIGDECAY,TIMESTEP,H2RADPROD,SIGH2RADPROD,O2RADPROD,SIGO2RADPROD,H2ORADCONS,SIG
H2ORADCONS)

    !CALCULATE MOLES OF H2O
    IF (H2ORADCONS.LT.MOLESH2O)THEN
        NEWH2O=MOLESH2O-H2ORADCONS
        CALL STATSUM(MOLESH2O,1.D0,SIGMOLESH2O,H2ORADCONS,-1.D0,SIGH2ORADCONS,0.D0,NEWH2O,SIGNEWH2O)
        MOLESH2O=NEWH2O
        SIGMOLESH2O=SIGNEWH2O
    ELSE
        H2RADPROD=2.D0/3.D0*MOLESH2O
        SIGH2RADPROD=2.D0/3.D0*SIGMOLESH2O
        O2RADPROD=0.5D0*H2RADPROD
        SIGO2RADPROD=0.5D0*H2RADPROD
        MOLESH2O=0.D0
        SIGMOLESH2O=1.0D-6  !ARBITRARY VALUE
    ENDIF

    !CALCULATE MOLES OF H2
    NEWH2=MOLESH2+H2RADPROD
    CALL STATSUM(MOLESH2,1.D0,SIGMOLESH2,H2RADPROD,1.D0,SIGH2RADPROD,0.D0,NEWH2,SIGNEWH2)
    MOLESH2=NEWH2
    SIGMOLESH2=SIGNEWH2

    !CALCULATE MOLE OF O2
    TEMP1=O2RADPROD-O2CORRCONS
    CALL STATSUM(O2RADPROD,1.D0,SIGO2RADPROD,O2CORRCONS,-1.D0,SIGO2CORRCONS,0.D0,TEMP1,SIGTEMP1)
        IF (TEMP1.GE. 0.D0)THEN
            NEWO2=MOLESO2+TEMP1
            CALL STATSUM(MOLESO2,1.D0,SIGMOLESO2,TEMP1,1.D0,SIGTEMP1,0.D0,NEWO2,SIGNEWO2)
            MOLESO2=NEWO2
```

```
            SIGMOLESO2=SIGNEWO2
        ELSEIF ((TEMP1 .LT.0.D0).AND.(ABS(TEMP1).LT.MOLESO2))THEN
           NEWO2=MOLESO2+TEMP1
            CALL STATSUM(MOLESO2,1.D0,SIGMOLESO2,TEMP1,1.D0,SIGTEMP1,0.D0,NEWO2,SIGNEWO2)
            MOLESO2=NEWO2
            SIGMOLESO2=SIGNEWO2
        ELSE
            MOLESO2=0.D0
            SIGMOLESO2=1.D-6     !ARBITRARY VALUE
        ENDIF

ENDIF

    CALL PRESSURE(MOLESH2,SIGMOLESH2,TEMP,SIGTEMP,GASVOL,PH2,SIGPH2)
    WRITE(2,60)I,MOLESH2O,SIGMOLESH2O,PH2,SIGPH2

    IF(MOLESH2O.EQ.0.D0)THEN
    WRITE(2,*)'ALL H2O CONSUMED IN ',I,'HOURS'
    PRINT*,'ALL H2O CONSUMED IN ',I,'HOURS'
    EXIT
    ENDIF

10  CONTINUE

50 FORMAT(I3,2X,F7.3,2X,2(E9.3,2X),F12.5,2X,E9.3,2X,E11.6,2X,E8.3)
60 FORMAT(I3,4(3X,E9.4))

END


SUBROUTINE UIMMERSDE(TEMP,SIGTEMP,TIME,SAEXP,SIGSAEXP,UCORR,SIGUCORR,H2CORRPROD,SIGH2CORRPROD,H2OCOR
RCONS,SIGH2OCORRCONS)

!THIS SUBROUTINE CALCULATES URANIUM CORROSION RATES FOR DEAERATED WATER,
!mg O/CM^2*HR AND mg/HR, GIVEN A TEMPERATURE
!USES RATE EQUATIONS EXTRACTED FROM DATA IN PEARCE. 10/89

!INTEGER O2      !IF O2=0, USE OXYGEN DEPLETED
!DOUBLE PRECISION O2LIM      !CUTOFF FOR DEAERATED WATER
!DOUBLE PRECISION PTOT,PO2  !TOTAL PRESSURE AND PARTIAL PRESSURE OF OXYGEN
DOUBLE PRECISION M,SIGM
DOUBLE PRECISION B,SIGB
DOUBLE PRECISION ADD
DOUBLE PRECISION TEMP,SIGTEMP    ! TEMPERATURE IN KELVIN,CALL FROM MAIN
DOUBLE PRECISION TIME            !TIME STEP IN HOURS
DOUBLE PRECISION SAEXP,SIGSAEXP ! TOTAL EXPOSED SURFACE AREA IN BASINS, CM^2,CALL FROM MAIN
DOUBLE PRECISION UCORR,SIGUCORR !VALUE THAT IS SENT TO MAIN, MG O2
DOUBLE PRECISION H2CORRPROD,SIGH2CORRPROD    !MOLES/HR OF H2 PRODUCED
DOUBLE PRECISION H2OCORRCONS,SIGH2OCORRCONS      !MOLES OF H2O CONSUMED ASSUMES 23"
                        !DIAMETER, FUEL DIAM OF 14",HEIGHT OF 140", REST IS WATER

    M=-2274.D0
    SIGM=42.7D0
    B=4.59D0
    SIGB=0.12D0
    ADD=0.D0
    CALL CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,UCORR,SIGUCORR)
    CALL H2PRES(UCORR,SIGUCORR,H2CORRPROD,SIGH2CORRPROD,H2OCORRCONS,SIGH2OCORRCONS)


END SUBROUTINE


SUBROUTINE UIMMERSO2(TEMP,SIGTEMP,TIME,SAEXP,SIGSAEXP,UCORR,SIGUCORR,O2CORRCONS,SIGO2CORRCONS)

!THIS SUBROUTINE CALCULATES URANIUM CORROSION RATES, mg O/CM^2*HR AND mg/HR,
!FOR AERATED CONDITIONS GIVEN A TEMPERATURE
!USES RATE EQUATIONS FROM PEARCE. 10/89

!INTEGER O2      !IF O2=0, USE OXYGEN DEPLETED
!DOUBLE PRECISION O2LIM      !CUTOFF FOR DEAERATED WATER
!DOUBLE PRECISION PTOT,PO2  !TOTAL PRESSURE AND PARTIAL PRESSURE OF OXYGEN
DOUBLE PRECISION M,SIGM
DOUBLE PRECISION B,SIGB
DOUBLE PRECISION ADD
DOUBLE PRECISION TEMP,SIGTEMP    ! TEMPERATURE IN KELVIN,CALL FROM MAIN
DOUBLE PRECISION TIME,O2CORRCONS,SIGO2CORRCONS
DOUBLE PRECISION SAEXP,SIGSAEXP ! TOTAL EXPOSED SURFACE AREA IN BASINS, CM^2,CALL FROM MAIN
```

```
DOUBLE PRECISION UCORR,SIGUCORR !VALUE THAT IS SENT TO MAIN, MG O2/HR
!DOUBLE PRECISION MOLESH2,SIGMOLESH2    !MOLES/HR OF H2 PRODUCED
!DOUBLE PRECISION WATER          !MOLES OF H2O IN CANISTER ASSUMES 23" DIAMETER, FUEL DIAM OF 14",
                    !HEIGHT OF 140", REST IS WATER

!WATER=15059
!O2PPM=(MOLESO2/WATER)*10000

M=-3797.D0
SIGM=345.D0
B=8.54D0
SIGB=1.0D0
ADD=0.D0
CALL CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,UCORR,SIGUCORR)
CALL O2PRES(UCORR,SIGUCORR,O2CORRCONS,SIGO2CORRCONS)

END SUBROUTINE



SUBROUTINE CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,UCORR,SIGUCORR)

DOUBLE PRECISION M,SIGM      !SLOPE AND STD DEV
DOUBLE PRECISION B,SIGB      !INTERCEPT AND STD DEV
DOUBLE PRECISION SAEXP,SIGSAEXP !EXPOSED FUEL AREA,CM^2
DOUBLE PRECISION TEMP,SIGTEMP   !TEMPERATURE IN KELVIN
DOUBLE PRECISION TIME
DOUBLE PRECISION ADD,TERM1,SIGTERM1,TERM2,SIGTERM2,EXPO,SIGEXPO
DOUBLE PRECISION KP,SIGKP       !REACTION RATE IN MG/HR/CM^2
DOUBLE PRECISION UCORR,SIGUCORR     !REACTION RATE IN MG/HR

!CALC REACTION RATE AND VARIANCE
TERM1=M/TEMP
CALL STATDIV(M,1.D0,SIGM,TEMP,SIGTEMP,0.D0,TERM1,SIGTERM1)
TERM2=TERM1+ADD
CALL STATSUM(TERM1,1.D0,SIGTERM1,ADD,1.D0,0.D0,0.D0,TERM2,SIGTERM2)
EXPO=B+TERM2
CALL STATSUM(B,1.D0,SIGB,TERM2,1.D0,SIGTERM2,0.D0,EXPO,SIGEXPO)
KP=10**EXPO
CALL STATPOW10(EXPO,SIGEXPO,1.D0,1.D0,KP,SIGKP)

!MULTIPLY BY SURFACE AREA, STD DEV ASSUMES NO CORRELATION
UCORR=TIME*KP*SAEXP
CALL STATPROD(KP,TIME,SIGKP,SAEXP,SIGSAEXP,0.D0,UCORR,SIGUCORR)

END SUBROUTINE CORRODE


SUBROUTINE H2PRES(UCORR,SIGUCORR,H2PROD,SIGH2PROD,H2OCONS,SIGH2OCONS)

DOUBLE PRECISION UCORR,SIGUCORR     !REACTION RATE IN MG O/HR
DOUBLE PRECISION H2PROD,SIGH2PROD
DOUBLE PRECISION H2OCONS,SIGH2OCONS

!CALC MOLES OF H2 PRODUCED, 7.347 CONVERTS FROM MG O TO MG U
H2PROD=UCORR*7.437D0*2.D0/2.38D5
SIGH2PROD=SIGUCORR*7.437D0*2.D0/2.38D5

!FOR EVERY MOLE OF HYDROGEN PRODUCED, 1 WATER CONSUMED
H2OCONS=H2PROD
SIGH2OCONS=SIGH2PROD

END SUBROUTINE H2PRES


SUBROUTINE O2PRES(UCORR,SIGUCORR,O2CONS,SIGO2CONS)

DOUBLE PRECISION UCORR,SIGUCORR     !REACTION RATE IN MG O
DOUBLE PRECISION O2CONS,SIGO2CONS

!CALC MOLES OF O2 CONSUMED
O2CONS=UCORR*1.D-3/32.D0
SIGO2CONS=SIGUCORR*1.D-3/32.D0

END SUBROUTINE O2PRES
```

```
SUBROUTINE STATSUM(VAR1,COEF1,SIG1,VAR2,COEF2,SIG2,SIG12,X,SIGX)

!USE TO CALCULATE STD DEV OF X IN EQUATION X=COEF1*VAR1+COEF2*VAR2 WITH
!STD DEVS OF VAR1 AND VAR2 EQUAL TO SIG1 AND SIG2, RESPECTIVELY AND
!COVARIANCE SIG12

DOUBLE PRECISION VAR1,VAR2
DOUBLE PRECISION COEF1,COEF2
DOUBLE PRECISION SIG1,SIG2,SIG12
DOUBLE PRECISION X,SIGX

SIGX=SQRT(COEF1**2*SIG1**2+COEF2**2*SIG2**2+2*COEF1*COEF2*SIG12)

END SUBROUTINE STATSUM


SUBROUTINE STATPROD(VAR1,COEF1,SIG1,VAR2,SIG2,SIG12,X,SIGX)

!USE TO CALCULATE THE STD DEV OF X IN EQUATION X=COEF1*VAR1*VAR2 WITH
!STD DEVS OF VAR1 AND VAR2 EQUAL TO SIG1 AND SIG2, RESPECTIVELY AND
!COVARIANCE SIG12

DOUBLE PRECISION VAR1,VAR2
DOUBLE PRECISION COEF1
DOUBLE PRECISION SIG1,SIG2,SIG12
DOUBLE PRECISION X,SIGX

SIGX=SQRT(X**2*((SIG1/VAR1)**2+(SIG2/VAR2)**2+2*SIG12**2/(VAR1*VAR2)))

END SUBROUTINE STATPROD


SUBROUTINE STATDIV(VAR1,COEF1,SIG1,VAR2,SIG2,SIG12,X,SIGX)

!USE TO CALCULATE THE STD DEV OF X IN EQUATION X=COEF1*(VAR1/VAR2) WITH
!STD DEVS OF VAR1 AND VAR2 EQUAL TO SIG1 AND SIG2, RESPECTIVELY, AND
!COVARIANCE SIG12

DOUBLE PRECISION VAR1,VAR2
DOUBLE PRECISION COEF1
DOUBLE PRECISION SIG1,SIG2,SIG12
DOUBLE PRECISION X,SIGX

SIGX=SQRT(X**2*((SIG1/VAR1)**2+(SIG2/VAR2)**2-2*SIG12**2/(VAR1*VAR2)))

END SUBROUTINE STATDIV


SUBROUTINE STATPOW10(VAR,SIG,COEF1,COEF2,X,SIGX)

!USED TO CALCULATE STD DEV OF X IN EQUATION X=COEF1*10**(COEF2*VAR)
!WITH STD DEV OF VAR EQUAL TO SIQ

DOUBLE PRECISION VAR,SIG,COEF1,COEF2,X,SIGX

SIGX=X*2.303D0*COEF2*SIG

END SUBROUTINE STATPOW10



SUBROUTINE WATER(SLUDGE,SIGSLUDGE,FREEH2O,SIGFREEH2O,MOLESH2O,SIGMOLESH2O)

!CALCULATES THE MOLES OF WATER (FREE,BOUND,CHEMISORBED) AVAILABLE FOR RADIOLYSIS

DOUBLE PRECISION SLUDGE,SIGSLUDGE   !KGS OF SLUDGE IN THE Mco
DOUBLE PRECISION FREEH2O,SIGFREEH2O !KGS OF FREE WATER
DOUBLE PRECISION BOUNDFRAC,SIGBOUNDFRAC !FRACTION OF SLUDGE THAT IS BOUND WATER
DOUBLE PRECISION CHEMFRAC,SIGCHEMFRAC   !FRACTION OF SLUDGE THAT IS CHEMISORBED WATER
DOUBLE PRECISION BOUNDKG,SIGBOUNDKG,CHEMKG,SIGCHEMKG,ABSKG,SIGABSKG
DOUBLE PRECISION MOLESH2O,SIGMOLESH2O !TOTAL INITAL MOLES OF WATER IN MCO

!PHYSICAL PARAMETERS
BOUNDFRAC=0.05D0
```

```
SIGBOUNDFRAC=0.01D0
CHEMFRAC=0.015D0
SIGCHEMFRAC=0.003D0

!CALCULATE KGS OF BOUND AND CHEMISORBED WATER
BOUNDKG=SLUDGE*BOUNDFRAC
CALL STATPROD(SLUDGE,1.0D0,SIGSLUDGE,BOUNDFRAC,SIGBOUNDFRAC,0.D0,BOUNDKG,SIGBOUNDKG)
CHEMKG=SLUDGE*CHEMFRAC
CALL STATPROD(SLUDGE,1.0D0,SIGSLUDGE,CHEMFRAC,SIGCHEMFRAC,0.D0,CHEMKG,SIGCHEMKG)
!ADD BOUND AND CHEMISORBED WATER
ABSKG=BOUNDKG+CHEMKG
CALL STATSUM(BOUNDKG,1.0D0,SIGBOUNDKG,CHEMKG,1.0D0,SIGCHEMKG,0.0D0,ABSKG,SIGABSKG)

!ADD TOTAL TO FREE WATER AND CONVERT TO MOLES WITH 1/0.018 MOLES/KG
MOLESH2O=55.56D0*(FREEH2O+ABSKG)
CALL STATSUM(FREEH2O,55.56D0,SIGFREEH2O,ABSKG,55.56D0,SIGABSKG,0.0D0,MOLESH2O,SIGMOLESH2O)


END SUBROUTINE WATER

SUBROUTINE RADIOL(DECAY,SIGDECAY,TIMESTEP,RADH2,SIGRADH2,RADO2,SIGRADO2,H2OCONS,SIGH2OCONS)

DOUBLE PRECISION GR !MOLES OF HYDROGEN PRODUCED PER SECOND PER WATT OF E ABSORBED, FROM RADIOL CODE
DOUBLE PRECISION RADFRAC,SIGRADFRAC !FRAC OF ENERGY DEPOSITED OUTSIDE FUEL
DOUBLE PRECISION ABSFRAC,SIGABSFRAC !FRAC OF RADFRAC DEPOSITED IN WATER
DOUBLE PRECISION DECAY,SIGDECAY     !DECAY POWER IN WATTS
DOUBLE PRECISION FRAC,SIGFRAC,MOLRATE,SIGMOLRATE
DOUBLE PRECISION RADH2,SIGRADH2,RADO2,SIGRADO2  !MOLES OF H2 AND O2 PRODUCED BY RADIOLYSIS
DOUBLE PRECISION H2OCONS,SIGH2OCONS
DOUBLE PRECISION TIME               !TIME IN SECONDS
DOUBLE PRECISION TIMESTEP

TIME=TIMESTEP*3600
!PHYSICAL PARAMETERS

GR=1.04E-5
RADFRAC=0.01D0
SIGRADFRAC=0.001D0
ABSFRAC=1.0D0
SIGABSFRAC=0.0D0

!CALCULATE FRACTION OF ENERGY DEPOSITED IN WATER
FRAC=ABSFRAC*RADFRAC
CALL STATPROD(ABSFRAC,1.0D0,SIGABSFRAC,RADFRAC,SIGRADFRAC,0.D0,FRAC,SIGFRAC)

!CALCULATE MOLES OF H2 PRODUCED
MOLRATE=GR*DECAY
CALL STATPROD(GR,1.0D0,0.D0,DECAY,SIGDECAY,0.D0,MOLRATE,SIGMOLRATE)

RADH2=MOLRATE*TIME*FRAC
CALL STATPROD(MOLRATE,TIME,SIGMOLRATE,FRAC,SIGFRAC,0.0D0,RADH2,SIGRADH2)

!0.5 MOLES OF O2 PRODUCED PER MOLE OF H2 PRODUCED
RADO2=0.5D0*RADH2
SIGRADO2=0.5D0*SIGRADH2

H2OCONS=RADH2
SIGH2OCONS=SIGRADH2

END SUBROUTINE RADIOL


 SUBROUTINE PRESSURE(MOLES,SIGMOLES,TEMP,SIGTEMP,FREEVOL,PRES,SIGPRES)

  DOUBLE PRECISION MOLES,SIGMOLES
  DOUBLE PRECISION TEMP,SIGTEMP
  DOUBLE PRECISION FREEVOL
  DOUBLE PRECISION PRES,SIGPRES
  DOUBLE PRECISION COEF
  COEF=8.314D0/FREEVOL
  PRES=COEF*MOLES*TEMP
  CALL STATPROD(MOLES,COEF,SIGMOLES,TEMP,SIGTEMP,0.D0,PRES,SIGPRES)

  END SUBROUTINE PRESSURE
```

1	3.06E+04	1.05E+01	1.11E+04	2.32E+03
2	3.06E+04	1.05E+01	2.21E+04	3.32E+03
3	3.06E+04	1.05E+01	3.32E+04	4.11E+03
4	3.06E+04	1.05E+01	4.42E+04	4.79E+03
5	3.06E+04	1.05E+01	5.53E+04	5.41E+03
6	3.06E+04	1.05E+01	6.64E+04	5.99E+03
7	3.06E+04	1.05E+01	7.74E+04	6.53E+03
8	3.06E+04	1.05E+01	8.85E+04	7.05E+03
9	3.06E+04	1.05E+01	9.95E+04	7.54E+03
10	3.06E+04	1.05E+01	1.11E+05	8.02E+03
11	3.06E+04	1.05E+01	1.22E+05	8.49E+03
12	3.06E+04	1.05E+01	1.33E+05	8.95E+03
13	3.06E+04	1.05E+01	1.44E+05	9.40E+03
14	3.06E+04	1.05E+01	1.55E+05	9.83E+03
15	3.06E+04	1.05E+01	1.66E+05	1.03E+04
16	3.06E+04	1.05E+01	1.77E+05	1.07E+04
17	3.06E+04	1.05E+01	1.88E+05	1.11E+04
18	3.06E+04	1.05E+01	1.99E+05	1.15E+04
19	3.06E+04	1.05E+01	2.10E+05	1.19E+04
20	3.06E+04	1.05E+01	2.21E+05	1.23E+04
21	3.06E+04	1.05E+01	2.32E+05	1.27E+04
22	3.06E+04	1.05E+01	2.43E+05	1.31E+04
23	3.06E+04	1.05E+01	2.54E+05	1.35E+04
24	3.06E+04	1.05E+01	2.65E+05	1.39E+04
25	3.06E+04	1.05E+01	2.77E+05	1.43E+04
26	3.06E+04	1.05E+01	2.88E+05	1.47E+04
27	3.06E+04	1.05E+01	2.99E+05	1.51E+04
28	3.06E+04	1.05E+01	3.10E+05	1.55E+04
29	3.06E+04	1.05E+01	3.21E+05	1.59E+04
30	3.06E+04	1.05E+01	3.32E+05	1.62E+04
31	3.06E+04	1.05E+01	3.43E+05	1.66E+04
32	3.06E+04	1.05E+01	3.54E+05	1.70E+04
33	3.06E+04	1.05E+01	3.65E+05	1.74E+04
34	3.06E+04	1.05E+01	3.76E+05	1.77E+04
35	3.06E+04	1.05E+01	3.87E+05	1.81E+04
36	3.06E+04	1.05E+01	3.98E+05	1.85E+04
37	3.06E+04	1.05E+01	4.09E+05	1.89E+04
38	3.06E+04	1.05E+01	4.20E+05	1.92E+04
39	3.06E+04	1.05E+01	4.31E+05	1.96E+04
40	3.06E+04	1.05E+01	4.42E+05	2.00E+04
41	3.06E+04	1.05E+01	4.53E+05	2.03E+04
42	3.06E+04	1.05E+01	4.65E+05	2.07E+04
43	3.06E+04	1.05E+01	4.76E+05	2.11E+04
44	3.06E+04	1.05E+01	4.87E+05	2.14E+04
45	3.06E+04	1.05E+01	4.98E+05	2.18E+04
46	3.06E+04	1.05E+01	5.09E+05	2.21E+04
47	3.06E+04	1.05E+01	5.20E+05	2.25E+04
48	3.06E+04	1.05E+01	5.31E+05	2.29E+04
49	3.06E+04	1.05E+01	5.42E+05	2.32E+04
50	3.06E+04	1.05E+01	5.53E+05	2.36E+04

Appendix C: Executable file and output for water vapor-filled staging regime

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$FREEFORM

!PROGRAM NAME VAP+RAD+HYD7-19.F90 MODIFIED TO ADD HYDRIDE ANALYSIS
!PROGRAM NAME VAP+RAD7-13.MODIFIED 7-13 TO INCUDE RADIOLYSIS
!
!FOR FUEL IN WATER VAPOR CALCULATES CONSUMPTION OF VAPOR AND OXYGEN,PRODUCTION OF H2
!EACH TIME STEP IS 1HOUR DUE TO UNITS OF REACTION RATE EQUATION

  INTEGER I,J       !1=FUEL IMMERSED IN H2O,2=FUEL IN VAPOR,3=DRYAIR
  INTEGER MO,DAY,YR,STEPS
  DOUBLE PRECISION TEMP,SIGTEMP      !TEMPERATURE IN KELVIN,STD DEV
  DOUBLE PRECISION SAEXP,SIGSAEXP    !TOTAL EXPOSED SURFACE AREA
  DOUBLE PRECISION UCORR,SIGUCORR    !TOTAL CORROSION IN MG
  DOUBLE PRECISION PO2,SIGPO2        !PARTIAL PRESSURE OF OXYGEN,ATM
  DOUBLE PRECISION PH2O,SIGPH2O      !PARTIAL PRESSURE OF WATER VAPOR,Pa
  DOUBLE PRECISION TOTPRES,SIGTOTPRES       !TOTAL SYSTEM PRESSURE, Pa
  DOUBLE PRECISION MOLESH2O,SIGMOLESH2O !MOLES OF H2O BOUND,FREE,CHEMISORBED
  DOUBLE PRECISION RADH2O,SIGRADH2O !MOLES OF WATER AVAILABLE FOR RADIOLYSIS
  DOUBLE PRECISION DECAY,SIGDECAY    !DECAY HEAT, WATTS
  DOUBLE PRECISION H2OVAP,SIGH2OVAP !MOLES OF WATER VAPOR
  DOUBLE PRECISION FREEVOL           !FREE VOLUME IN M^3
  DOUBLE PRECISION R                 !GAS CONSTANT IN J/MOL*K
  DOUBLE PRECISION PO2LIM            !PRESSURE LIM,Pa, FOR OXYGEN FREE ATMOSPHERE
  DOUBLE PRECISION MOLESH2,SIGMOLESH2   !TOTAL MOLES OF H2, ADDED TO AT END OF EACH LCOP
  DOUBLE PRECISION COEF1,COEF2
  DOUBLE PRECISION TSAT(100),PSAT(100) !SATURATION TEMPERATURES AND PRESSURES
  DOUBLE PRECISION MOLESO2,SIGMOLESO2,MOLESO2LIM, SIGMOLESO2LIM !MOLES OF O2 IN WATER VAPOR
                           !LIMIT IS THE LIMIT FOR WHICH O2 CORROSION OCCURS
  DOUBLE PRECISION SLUDGE,SIGSLUDGE !SLUDGE, KGS
  DOUBLE PRECISION FREEH2O,SIGFREEH2O   !FREE WATER IN KGS, NOT IN TOUCH WITH FUEL
  DOUBLE PRECISION NEWO2,SIGNEWO2
  DOUBLE PRECISION H2OCONS,SIGH2OCONS   !MOLES OF H2O CONSUMED IN CORROSION
  DOUBLE PRECISION TIMESTEP             !IN HOURS, INPUT TOTAL LENGTH OF TIMESTEP
  DOUBLE PRECISION PH2,SIGPH2   !HYDROGEN PARTIAL PRESSURE IN Pa
  DOUBLE PRECISION H2EQPP           !EQ. H2 PARTIAL PRESS BELOW WHICH HYDRIDING DOES NOT OCCUR
  DOUBLE PRECISION O2CORRCONS,SIGO2CORRCONS,H2CORRPROD,SIGH2CORRPROD,H2HYDCONS,SIGH2HYDCONS,O2RADPRO
D
  DOUBLE PRECISION SIGO2RADPROD,H2ORADCONS,SIGH2ORADCONS,NEWH2,SIGNEWH2,NEWH2OVAP,SIGNEWH2OVAP
  DOUBLE PRECISION H2OCORRCONS,SIGH2OCORRCONS,H2RADPROD,SIGH2RADPROD
  DOUBLE PRECISION NEWRADH2O,SIGNEWRADH2O
  DOUBLE PRECISION TEMP1,SIGTEMP1,TEMP2,SIGTEMP2,TEMP3,SIGTEMP3,PH2MIN,SIGPH2MIN,MOLESH2MIN,SIGMOLES
H2MIN

OPEN(1,FILE='c:\users\lisas\tbl_tc.prn')
OPEN(2,FILE='c:\users\lisas\tbl_pm.prn')
OPEN(3,FILE='c:\users\lisas\OUTPUT\VAPOR\VAP.txt')

!READ IN SATURATION T AND PRES
DO 5 I=1,50
    READ(1,*)TSAT(I)
    READ(2,*)PSAT(I)
    TSAT(I)=TSAT(I)+273 !CONVERT TO KELVIN
5 CONTINUE
CLOSE(1)
CLOSE(2)

CALL GETDAT(YR,MO,DAY)
WRITE(3,*)'FILE GENERATED ON ',MO,'-',DAY,'-',YR

!SET CONSTANTS
FREEVOL=0.6D0
TEMP=298.D0
SIGTEMP=10.D0
SAEXP=3000.D0
SIGSAEXP=500.D0
R=8.314D0
PO2LIM=10.D0        !FROM APPENDIX G OF TAG REPORT
SLUDGE=10.D0
SIGSLUDGE=3.D0
FREEH2O=0.01D0
SIGFREEH2O=0.001D0
DECAY=550.D0
SIGDECAY=100.D0
TIMESTEP=1.D0
STEPS=50

!INITIALIZE VARIABLES
PO2=0.D0
SIGPO2=0.D0
PH2O=1.013D5
```

```
SIGPH2O=10.D0
H2OCONS=0.D0
SIGH2OCONS=0.D0

!CALCULATE MIN H2 PARTIAL PRESSURE
CALL H2MIN(TEMP,SIGTEMP,PH2MIN,SIGPH2MIN)
!CALCULATE MIN MOLES OF H2
CALL MOLECONV(PH2MIN,SIGPH2MIN,FREEVOL,TEMP,SIGTEMP,MOLESH2MIN,SIGMOLESH2MIN)
!INITIAL H2 IS MINIMUM
PH2=PH2MIN
SIGPH2=SIGPH2MIN
MOLESH2=MOLESH2MIN
SIGMOLESH2=SIGMOLESH2MIN

!CALCULATE INITIAL TOTAL PRESSURE
TEMP1=PH2+PO2
CALL STATSUM(PH2,1.D0,SIGPH2,PO2,1.D0,SIGPO2,0.D0,TEMP1,SIGTEMP1)
TOTPRES=PH2O+TEMP1
CALL STATSUM(PH2O,1.D0,SIGPH2O,TEMP1,1.D0,SIGTEMP1,0.D0,TOTPRES,SIGTOTPRES)

!CALCULATE INITIAL MOLES OF O2,AND MOLES PO2LIM. STD DEV ASSUMES NO CORRELATION BETWEEN
!TEMPERATURE AND PRESSURE, ASSUMES FILLED AT 25C
CALL MOLECONV(PO2,SIGPO2,FREEVOL,298.D0,10.D0,MOLESO2,SIGMOLESO2)
CALL MOLECONV(PO2LIM,0.D0,FREEVOL,298.D0,10.D0,MOLESO2LIM,SIGMOLESO2LIM)

!CALCULATE INITIAL MOLES OF H2O VAPOR FROM PH2O,FREEVOLUME AND TEMP
CALL MOLECONV(PH2O,SIGPH2O,FREEVOL,298.D0,10.D0,H2OVAP,SIGH2OVAP)

!CALCULATE MOLES OF H2O FREE,BOUND,CHEMISORBED, TOTAL MOLES OF WATER
!FREE,BOUND,CHEMISORBED,VAPOR, ARE AVAILABLE FOR RADIOLYSIS,--RADH2O
CALL WATER(SLUDGE,SIGSLUDGE,FREEH2O,SIGFREEH2O,MOLESH2O,SIGMOLESH2O)
RADH2O=H2OVAP+MOLESH2O
CALL STATSUM(H2OVAP,1.0D0,SIGH2OVAP,MOLESH2O,1.0D0,SIGMOLESH2O,0.D0,RADH2O,SIGRADH2O)

!WRITE(3,*)'TEMP= ',TEMP,' INITAL PRES= ',TOTPRES
!WRITE(3,*)'TIMESTEP= ',TIMESTEP,'HOURS'
!WRITE(3,*)' J      PH2O        SIGPH2O      PH2        SIGPH2       PO2       SIGPO2        TOTPRES    SIG
TOTPRES'


DO 10 J=1,STEPS

    !WITH O2:
        !O2 CONSUMED BY CORROSION, PRODUCED BY RADIOLYSIS
        !H2 PRODUCED BY RADIOLYSIS, CONSUMED BY HYDRIDING
        !H2O CONSUMED BY RADIOLYSIS
    !WITHOUT O2
        !O2 PRODUCED BY RADIOLYSIS
        !H2 PRODUCED BY RADIOLYSIS AND CORROSION, CONSUMED BY HYDRIDING
        !H2O CONSUMED BY RADIOLYSIS AND CORROSION
    CALL UVAPOR(TSAT,PSAT,TEMP,SIGTEMP,TIMESTEP,PH2O,SIGPH2O,MOLESO2,MOLESO2LIM,TOTPRES,SAEXP,SIGSAE
XP,UCORR,SIGUCORR,H2OCORRCONS,SIGH2OCORRCONS,O2CORRCONS,SIGO2CORRCONS,H2CORRPROD,SIGH2CORRPROD)

    !CHECK TO SEE IF PARTIAL PRESS OF H2 IS ABOVE EQ PRESSURE OVER HYDRIDE.
    !IF SO, CALCULATE THE H2 CONSUMED IN HYDRIDING, OTHERWISE NO H2 CONSUMED
    IF (PH2 .GT. H2EQPP(TEMP))THEN
        CALL HYDRIDE(TEMP,PH2,TIMESTEP,SAEXP,SIGSAEXP,H2HYDCONS,SIGH2HYDCONS)
    ELSE
        H2HYDCONS=0.D0
        SIGH2HYDCONS=0.D0
    ENDIF


!CHECK THAT H2CONS IS NOT LOWER THAN EQUILIBRIUM LIMIT
    TEMP1=MOLESH2-H2HYDCONS
    IF ((TEMP1 .LE. MOLESH2MIN).AND.(H2HYDCONS.GT.0.D0))THEN
        H2HYDCONS=MOLESH2-MOLESH2MIN
        CALL STATSUM(MOLESH2,1.D0,SIGMOLESH2,MOLESH2MIN,-1.D0,SIGMOLESH2MIN,0.D0,H2HYDCONS,SIGH2HYDC
ONS)
    ENDIF

    CALL RADIOL(DECAY,SIGDECAY,TIMESTEP,H2RADPROD,SIGH2RADPROD,O2RADPROD,SIGO2RADPROD,H2ORADCONS,SIG
H2ORADCONS)

    !CALCULATE NEW MOLES OF H2,O2,H2O AND RADH2O

    IF (H2OCORRCONS .LT. H2OVAP)THEN
        NEWH2OVAP=H2OVAP-H2OCORRCONS
        CALL STATSUM(H2OVAP,1.D0,SIGH2OVAP,H2OCORRCONS,-1.D0,SIGH2OCORRCONS,0.D0,NEWH2OVAP,SIGNEWH2O
VAP)
```

```
        H2OVAP=NEWH2OVAP
        SIGH2OVAP=SIGNEWH2OVAP
    ELSE
        H2OCORRCONS=H2OVAP
        SIGH2OCORRCONS=SIGH2OVAP
        !IF CORROSION IS BY WATER, THEN AMOUNT OF H2 PRODUCED IS CHANGED,
        IF (H2CORRPROD .GT. 0.D0)THEN
        H2CORRPROD=H2OCORRCONS
        SIGH2CORRPROD=SIGH2OCORRCONS
        ENDIF
        !STD DEV ON H2OVAP DOES NOT CHANGE
        H2OVAP=0.D0
    ENDIF

    !CALCULATE NEW H2--IF H2HYDCONS WAS ALTERED ABOVE, STDDEV MUST BE CALCULATED DIFFERENTLY
    !TO AVOID INCORPORATING SIGMOLESH2 TWICE
    TEMP2=H2CORRPROD+H2RADPROD
    CALL STATSUM(H2CORRPROD,1.D0,SIGH2CORRPROD,H2RADPROD,1.D0,SIGH2RADPROD,0.D0,TEMP1,SIGTEMP1)
    IF (TEMP1.LE.MOLESH2MIN)THEN
        NEWH2=TEMP2+MOLESH2MIN
        CALL STATSUM(TEMP2,1.D0,SIGTEMP2,MOLESH2MIN,1.D0,SIGMOLESH2MIN,0.D0,NEWH2,SIGNEWH2)
    ELSE
        TEMP3=TEMP2-H2HYDCONS
        CALL STATSUM(TEMP2,1.D0,SIGTEMP2,H2HYDCONS,-1.D0,SIGH2HYDCONS,0.D0,TEMP3,SIGTEMP3)
        NEWH2=MOLESH2+TEMP3
        CALL STATSUM(MOLESH2,1.D0,SIGMOLESH2,TEMP3,1.D0,SIGTEMP3,0.D0,NEWH2,SIGNEWH2)
    ENDIF
        MOLESH2=NEWH2
        SIGMOLESH2=SIGNEWH2

    !CALCULATE NEW O2
    IF (O2CORRCONS .LT. MOLESO2)THEN
        TEMP1=O2RADPROD-O2CORRCONS
        CALL STATSUM(O2RADPROD,1.D0,SIGO2RADPROD,O2CORRCONS,-1.D0,SIGO2CORRCONS,0.D0,TEMP1,SIGTEMP1)
        NEWO2=MOLESO2+TEMP1
        CALL STATSUM(MOLESO2,1.D0,SIGMOLESO2,TEMP1,1.D0,SIGTEMP1,0.D0,NEWO2,SIGNEWO2)
        MOLESO2=NEWO2
        SIGMOLESO2=SIGNEWO2
    ELSE
        MOLESO2=O2RADPROD
        SIGMOLESO2=SIGO2RADPROD
    ENDIF


    TEMP1=H2OCORRCONS+H2ORADCONS
    CALL STATSUM(H2OCORRCONS,1.D0,SIGH2OCORRCONS,H2ORADCONS,1.D0,SIGH2ORADCONS,0.D0,TEMP1,SIGTEMP1)
    NEWRADH2O=RADH2O-TEMP1
    CALL STATSUM(RADH2O,1.D0,SIGRADH2O,TEMP1,-1.D0,SIGTEMP1,0.D0,NEWRADH2O,SIGNEWRADH2O)
    RADH2O=NEWRADH2O
    SIGRADH2O=SIGNEWRADH2O

    CALL PRESSURE(H2OVAP,SIGH2OVAP,TEMP,SIGTEMP,FREEVOL,PH2O,SIGPH2O)
    CALL PRESSURE(MOLESH2,SIGMOLESH2,TEMP,SIGTEMP,FREEVOL,PH2,SIGPH2)
    CALL PRESSURE(MOLESO2,SIGMOLESO2,TEMP,SIGTEMP,FREEVOL,PO2,SIGPO2)

    TEMP1=PH2O+PH2
    CALL STATSUM(PH2O,1.D0,SIGPH2O,PH2,1.D0,SIGPH2,0.D0,TEMP1,SIGTEMP1)
    TOTPRES=TEMP1+PO2
    CALL STATSUM(TEMP1,1.D0,SIGTEMP1,PO2,1.D0,SIGPO2,0.D0,TOTPRES,SIGTOTPRES)



    PRINT*,J,PH2O,SIGPH2O,PH2,SIGPH2,PO2,SIGPO2,TOTPRES,SIGTOTPRES
    PRINT*
    WRITE(3,50)J,PH2O,SIGPH2O,PH2,SIGPH2,PO2,SIGPO2,TOTPRES,SIGTOTPRES


    !IF ALL OF THE VAPOR HAS BEEN CONSUMED THEN EXIT LOOP--WILL GO TO DRY OXYGEN/AIR
    IF (H2OVAP .EQ. 0.0D0)THEN
        PRINT*,'ALL WATER VAPOR CONSUMED IN ',J,' HOURS'
        WRITE(3,*)'ALL WATER VAPOR CONSUMED IN ',J,' HOURS'
        EXIT
    ENDIF

10 CONTINUE

50 FORMAT(I3,6(2X,E10.4),2(2X,E12.7))

END
```

```
SUBROUTINE UVAPOR(TSAT,PSAT,TEMP,SIGTEMP,TIME,PH2O,SIGPH2O,MOLESO2,MOLESO2LIM,TOTPRES,SAEXP,SIGSAEXP
,UCORR,SIGUCORR,H2OCONS,SIGH2OCONS,O2CONS,SIGO2CONS,H2PROD,SIGH2PROD)

!THIS SUBROUTINE CALCULATES THE REACTION RATE, mg O/cm^2/hr (WEIGHT GAIN), FOR
!URANIUM IN WATER VAPOR USING PEARCE RELATIONS FROM "A REVEIW OF THE RATES OF
!REACTION OF UNIRRADIATED URANIUM IN GASEOUS ATMOSPHERES" CONVERTS TO MG BY MULTIPLYING
!BY SURFACE AREA AND TIMESTEP

DOUBLE PRECISION TEMP,SIGTEMP   !TEMPERATURE IN KELVIN CALLED FROM MAIN
DOUBLE PRECISION TIME           !TIME STEP IN HOURS
DOUBLE PRECISION MOLESO2 !MOLES OF OXYGEN, CALLED FROM MAIN
DOUBLE PRECISION PH2O,SIGPH2O       !VAPOR PARTIAL PRESSURE, IN ATM, CALLED FROM MAIN
DOUBLE PRECISION PRESSAT            !VAPOR SATURATION PRESSURE AT TEMP, ATM
DOUBLE PRECISION TOTPRES        !TOTAL SYSTEM PRESSURE, Pa, CALLED FROM MAIN
DOUBLE PRECISION PRES,SIGPRES           !TOTAL PRESSURE,kPA,USED IN CALCULATION
DOUBLE PRECISION MOLESO2LIM         !PRESSURE LIMIT,MOLES, FOR O2 FREE REGIME
DOUBLE PRECISION SAEXP,SIGSAEXP !TOTAL EXPOSED SURFACE AREA CALLED FROM MAIN
DOUBLE PRECISION UCORR,SIGUCORR     !KP*SAEXP
DOUBLE PRECISION TSAT(100),PSAT(100) !SATURATION TEMP AND PRES READ FROM FILES,T IN CELSIUS,
DOUBLE PRECISION M,SIGM
DOUBLE PRECISION B,SIGB
DOUBLE PRECISION ADD,SIGADD
DOUBLE PRECISION H2OCONS,SIGH2OCONS,O2CONS,SIGO2CONS,H2PROD,SIGH2PROD
INTEGER J


!CHECK TO SEE IF TEMPERATURE FALLS IN FIRST REGION, IF YES, INTERPOLATE FOR PRESSURE
IF (TEMP .EQ. TSAT(1))THEN
    PRESSAT=PSAT(1)
    ELSEIF(TEMP .LT. TSAT(1))THEN
    PRESSAT=0.0D0+((TEMP-0.0D0)/(TSAT(1)-0.0D0))*(PSAT(1)-0.0D0)
ENDIF

!FIND RANGE THAT TEMP FALLS IN AND INTERPOLATE TO FIND SATURATION PRESSURE
DO 20 J=1,49
    IF (TEMP .EQ. TSAT(J+1))THEN
    PRESSAT=PSAT(J+1)
    ELSEIF ((TEMP .GT. TSAT(J)) .AND. (TEMP .LT. TSAT(J+1)))THEN
    PRESSAT=PSAT(J)+((TEMP-TSAT(J))/(TSAT(J+1)-TSAT(J)))*(PSAT(J+1)-PSAT(J))
    ENDIF
20  CONTINUE

PRESSAT=PRESSAT*1E6 !CHANGE MPa TO Pa

!CHANGE Pa TO kPA
PRES=PH2O/1.D3
SIGPRES=SIGPH2O/1.D3

IF (MOLESO2 .LT. MOLESO2LIM)THEN
    M=-2274.D0              !SLOPE AND INTERCEPT VALUES FOR O2 FREE ATMOSPHERE
    !SIGM=0.0D0
    SIGM=42.7D0             !WATER CONSUMED, H2 GENERATED
    B=4.59D0
    SIGB=0.12D0
    ADD=0.5D0*LOG10(PRES)
    CALL STATLOG10(0.5D0,1.D0,PRES,SIGPRES,ADD,SIGADD)
    CALL CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,SIGADD,UCORR,SIGUCORR)
    CALL H2PRES(UCORR,SIGUCORR,H2PROD,SIGH2PROD)
    CALL H2OPRES(H2PROD,SIGH2PROD,H2OCONS,SIGH2OCONS)
    O2CONS=0.D0
    SIGO2CONS=0.D0
    ELSEIF ((TEMP .LT. 373.D0) .AND. (PH2O .GE. PRESSAT))THEN   !OTHERWISE, MOIST AIR CALCS
    M=-3797.D0                  !IF UNDER 100c, CHECK RH
    SIGM=345.D0
    !SIGM=0.0D0                 !O2 CONSUMED
    B=8.54D0
    !SIGB=1.0D0
    SIGB=0.0D0
    ADD=0.D0
    SIGADD=0.D0
    CALL CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,SIGADD,UCORR,SIGUCORR)
    CALL O2PRES(UCORR,SIGUCORR,O2CONS,SIGO2CONS)
    H2PROD=0.D0
    SIGH2PROD=0.D0
    ELSEIF(TEMP .LT. 465)THEN
    M=-3720.D0
    SIGM=93.6D0
    !SIGM=0.0D0
    B=7.50D0
```

```
    !SIGB=0.24D0
    SIGB=0.0D0
    ADD=0.3D0*LOG10(PRES)
    CALL STATLOG10(0.3D0,1.D0,PRES,SIGPRES,ADD,SIGADD)
    CALL CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,SIGADD,UCORR,SIGUCORR)
    CALL O2PRES(UCORR,SIGUCORR,O2CONS,SIGO2CONS)
    H2PROD=0.D0
    SIGH2PROD=0.D0
    ELSE
    M=-2963.D0
    SIGM=100.D0
    !SIGM=0.0D0
    B=6.19D0
    !SIGB=0.02D0
    SIGB=0.0D0
    ADD=0.3D0*LOG10(PRES)
    CALL STATLOG10(0.3D0,1.D0,PRES,SIGPRES,ADD,SIGADD)
    CALL CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,SIGADD,UCORR,SIGUCORR)
    CALL O2PRES(UCORR,SIGUCORR,O2CONS,SIGO2CONS)
    H2PROD=0.D0
    SIGH2PROD=0.D0
    ENDIF


END SUBROUTINE UVAPOR



SUBROUTINE CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,SIGADD,UCORR,SIGUCORR)

DOUBLE PRECISION M,SIGM      !SLOPE AND STD DEV
DOUBLE PRECISION B,SIGB      !INTERCEPT AND STD DEV
DOUBLE PRECISION SAEXP,SIGSAEXP !EXPOSED FUEL AREA,CM^2
DOUBLE PRECISION TEMP,SIGTEMP   !TEMPERATURE IN KELVIN
DOUBLE PRECISION TIME           !TIME STEP IN HOURS
DOUBLE PRECISION ADD,SIGADD
DOUBLE PRECISION KP,SIGKP       !REACTION RATE IN MG/HR/CM^2
DOUBLE PRECISION UCORR,SIGUCORR     !REACTION  IN MG
DOUBLE PRECISION TERM1,SIGTERM1,TERM2,SIGTERM2,EXPO,SIGEXPO

!CALC REACTION RATE AND VARIANCE
TERM1=B+ADD
CALL STATSUM(B,1.D0,SIGB,ADD,1.D0,SIGADD,0.D0,TERM1,SIGTERM1)
TERM2=M/TEMP
CALL STATDIV(M,1.D0,SIGM,TEMP,SIGTEMP,0.D0,TERM2,SIGTERM2)
EXPO=TERM1+TERM2
CALL STATSUM(TERM1,1.D0,SIGTERM1,TERM2,1.D0,SIGTERM2,0.D0,EXPO,SIGEXPO)
KP=10**EXPO
CALL STATPOW10(EXPO,SIGEXPO,1.D0,1.D0,KP,SIGKP)

!MULTIPLY BY SURFACE AREA, STD DEV ASSUMES NO CORRELATION
UCORR=TIME*KP*SAEXP
CALL STATPROD(KP,TIME,SIGKP,SAEXP,SIGSAEXP,0.D0,UCORR,SIGUCORR)

END SUBROUTINE CORRODE


SUBROUTINE H2PRES(UCORR,SIGUCORR,H2PROD,SIGH2PROD)

DOUBLE PRECISION UCORR,SIGUCORR     !REACTION RATE IN MG O
DOUBLE PRECISION H2PROD,SIGH2PROD

!CALC MOLES OF H2 PRODUCED, 7.347 CONVERTS FROM MG O TO MG U
H2PROD=UCORR*7.437D0*2.D0/2.38D5
SIGH2PROD=SIGUCORR*7.437D0*2.D0/2.38D5

END SUBROUTINE H2PRES


SUBROUTINE H2OPRES(H2PROD,SIGH2PROD,H2OCONS,SIGH2OCONS)

DOUBLE PRECISION H2PROD,SIGH2PROD
DOUBLE PRECISION H2OCONS,SIGH2OCONS

!NUMBER OF MOLES OF WATER CONSUMED EQUALS NUMBER MOLES OF H2 PRODUCED
H2OCONS=H2PROD
SIGH2OCONS=SIGH2PROD
```

```
END SUBROUTINE H2OPRES


SUBROUTINE O2PRES(UCORR,SIGUCORR,O2CONS,SIGO2CONS)

DOUBLE PRECISION UCORR,SIGUCORR      !REACTION RATE IN MG O
DOUBLE PRECISION O2CONS,SIGO2CONS

!CALC MOLES OF O2 CONSUMED
O2CONS=UCORR*1.D-3/32.D0
SIGO2CONS=SIGUCORR*1.D-3/32.D0

END SUBROUTINE O2PRES



SUBROUTINE STATDIV(VAR1,COEF1,SIG1,VAR2,SIG2,SIG12,X,SIGX)

!USE TO CALCULATE THE STD DEV OF X IN EQUATION X=COEF1*(VAR1/VAR2) WITH
!STD DEVS OF VAR1 AND VAR2 EQUAL TO SIG1 AND SIG2, RESPECTIVELY, AND
!COVARIANCE SIG12

DOUBLE PRECISION VAR1,VAR2
DOUBLE PRECISION COEF1
DOUBLE PRECISION SIG1,SIG2,SIG12
DOUBLE PRECISION X,SIGX

SIGX=SQRT(X**2*((SIG1/VAR1)**2+(SIG2/VAR2)**2-2*SIG12**2/(VAR1*VAR2)))

END SUBROUTINE STATDIV


SUBROUTINE STATSUM(VAR1,COEF1,SIG1,VAR2,COEF2,SIG2,SIG12,X,SIGX)

!USE TO CALCULATE STD DEV OF X IN EQUATION X=COEF1*VAR1+COEF2*VAR2 WITH
!STD DEVS OF VAR1 AND VAR2 EQUAL TO SIG1 AND SIG2, RESPECTIVELY AND
!COVARIANCE SIG12

DOUBLE PRECISION VAR1,VAR2
DOUBLE PRECISION COEF1,COEF2
DOUBLE PRECISION SIG1,SIG2,SIG12
DOUBLE PRECISION X,SIGX

SIGX=SQRT(COEF1**2*SIG1**2+COEF2**2*SIG2**2+2*COEF1*COEF2*SIG12**2)

END SUBROUTINE STATSUM


SUBROUTINE STATPROD(VAR1,COEF1,SIG1,VAR2,SIG2,SIG12,X,SIGX)

!USE TO CALCULATE THE STD DEV OF X IN EQUATION X=COEF1*VAR1*VAR2 WITH
!STD DEVS OF VAR1 AND VAR2 EQUAL TO SIG1 AND SIG2, RESPECTIVELY AND
!COVARIANCE SIG12

DOUBLE PRECISION VAR1,VAR2
DOUBLE PRECISION COEF1
DOUBLE PRECISION SIG1,SIG2,SIG12
DOUBLE PRECISION X,SIGX

!GET FLOATING POINT ERROR WHEN MOLESH2O=0
!!!***CHECK SOLUTION***!!!

IF((VAR1 .EQ. 0.D0) .OR. (VAR2 .EQ. 0.D0) .OR. (X .EQ. 0.D0))THEN
    SIGX=0.D0
ELSE

    SIGX=SQRT(X**2*((SIG1/VAR1)**2+(SIG2/VAR2)**2+2*SIG12**2/(VAR1*VAR2)))

ENDIF

END SUBROUTINE STATPROD


SUBROUTINE STATPOW10(VAR,SIG,COEF1,COEF2,X,SIGX)
```

```
!USED TO CALCULATE STD DEV OF X IN EQUATION X=COEF1*10**(COEF2*VAR)
!WITH STD DEV OF VAR EQUAL TO SIQ

DOUBLE PRECISION VAR,SIG,COEF1,COEF2,X,SIGX

SIGX=X*2.303D0*COEF2*SIG

END SUBROUTINE STATPOW10


SUBROUTINE STATLOG10(COEF1,COEF2,VAR,SIGVAR,X,SIGX)

!CALCULATE SIG X WITH X FORM OF X=COEF1*LOG10(COEF2*VAR)

DOUBLE PRECISION COEF1,COEF2,VAR,SIGVAR,X,SIGX

SIGX=COEF1*SIGVAR/(2.303*VAR)

END SUBROUTINE



SUBROUTINE WATER(SLUDGE,SIGSLUDGE,FREEH2O,SIGFREEH2O,MOLESH2O,SIGMOLESH2O)

!CALCULATES THE MOLES OF WATER (FREE,BOUND,CHEMISORBED) AVAILABLE FOR RADIOLYSIS

DOUBLE PRECISION SLUDGE,SIGSLUDGE   !KGS OF SLUDGE IN THE Mco
DOUBLE PRECISION FREEH2O,SIGFREEH2O !KGS OF FREE WATER
DOUBLE PRECISION BOUNDFRAC,SIGBOUNDFRAC !FRACTION OF SLUDGE THAT IS BOUND WATER
DOUBLE PRECISION CHEMFRAC,SIGCHEMFRAC   !FRACTION OF SLUDGE THAT IS CHEMISORBED WATER
DOUBLE PRECISION BOUNDKG,SIGBOUNDKG,CHEMKG,SIGCHEMKG,ABSKG,SIGABSKG
DOUBLE PRECISION MOLESH2O,SIGMOLESH2O !TOTAL INITAL MOLES OF WATER IN MCO

!PHYSICAL PARAMETERS
BOUNDFRAC=0.05D0
SIGBOUNDFRAC=0.01D0
CHEMFRAC=0.015D0
SIGCHEMFRAC=0.003D0

!CALCULATE KGS OF BOUND AND CHEMISORBED WATER
BOUNDKG=SLUDGE*BOUNDFRAC
CALL STATPROD(SLUDGE,1.0D0,SIGSLUDGE,BOUNDFRAC,SIGBOUNDFRAC,0.D0,BOUNDKG,SIGBOUNDKG)
CHEMKG=SLUDGE*CHEMFRAC
CALL STATPROD(SLUDGE,1.0D0,SIGSLUDGE,CHEMFRAC,SIGCHEMFRAC,0.D0,CHEMKG,SIGCHEMKG)
!ADD BOUND AND CHEMISORBED WATER
ABSKG=BOUNDKG+CHEMKG
CALL STATSUM(BOUNDKG,1.0D0,SIGBOUNDKG,CHEMKG,1.0D0,SIGCHEMKG,0.0D0,ABSKG,SIGABSKG)

!ADD TOTAL TO FREE WATER AND CONVERT TO MOLES WITH 1/0.018 MOLES/KG
MOLESH2O=55.56D0*(FREEH2O+ABSKG)
CALL STATSUM(FREEH2O,55.56D0,SIGFREEH2O,ABSKG,55.56D0,SIGABSKG,0.0D0,MOLESH2O,SIGMOLESH2O)

!PRINT*,MOLESH2O,' MOLES OF H2O +-',SIGMOLESH2O

END SUBROUTINE WATER


SUBROUTINE RADIOL(DECAY,SIGDECAY,TIMESTEP,RADH2,SIGRADH2,RADO2,SIGRADO2,H2OCONS,SIGH2OCONS)

DOUBLE PRECISION GR !MOLES OF HYDROGEN PRODUCED PER SECOND PER WATT OF E ABSORBED, FROM RADIOL CODE
DOUBLE PRECISION RADFRAC,SIGRADFRAC !FRAC OF ENERGY DEPOSITED OUTSIDE FUEL
DOUBLE PRECISION ABSFRAC,SIGABSFRAC !FRAC OF RADFRAC DEPOSITED IN WATER
DOUBLE PRECISION DECAY,SIGDECAY     !DECAY POWER IN WATTS
DOUBLE PRECISION FRAC,SIGFRAC,MOLRATE,SIGMOLRATE
DOUBLE PRECISION RADH2,SIGRADH2,RADO2,SIGRADO2  !MOLES OF H2 AND O2 PRODUCED BY RADIOLYSIS
DOUBLE PRECISION H2OCONS,SIGH2OCONS
DOUBLE PRECISION TIME               !TIME IN SECONDS
DOUBLE PRECISION TIMESTEP

TIME=TIMESTEP*3600
!PHYSICAL PARAMETERS

GR=1.04D-7
RADFRAC=0.01D0
SIGRADFRAC=0.001D0
ABSFRAC=0.5D0
SIGABSFRAC=0.0D0
```

```
!CALCULATE FRACTION OF ENERGY DEPOSITED IN WATER
FRAC=ABSFRAC*RADFRAC
CALL STATPROD(ABSFRAC,1.0D0,SIGABSFRAC,RADFRAC,SIGRADFRAC,0.D0,FRAC,SIGFRAC)

!CALCULATE MOLES OF H2 PRODUCED
MOLRATE=GR*DECAY
CALL STATPROD(GF,1.0D0,0.D0,DECAY,SIGDECAY,0.D0,MOLRATE,SIGMOLRATE)

RADH2=MOLRATE*TIME*FRAC
CALL STATPROD(MOLRATE,TIME,SIGMOLRATE,FRAC,SIGFRAC,0.0D0,RADH2,SIGRADH2)

!0.5 MOLES OF O2 PRODUCED PER MOLE OF H2 PRODUCED
RADO2=0.5D0*RADH2
SIGRADO2=0.5D0*SIGRADH2

H2OCONS=RADH2
SIGH2OCONS=SIGRADH2

END SUBROUTINE RADIOL



SUBROUTINE HYDRIDE(TEMP,PRES,TIMESTEP,SAEXP,SIGSAEXP,H2CONS,SIGH2CONS)

!CALCULATES THE WEIGHT GAIN AND SPALL FRONT VELOCITY IN U HYDRIDING.
!MODEL TAKEN FROM KIRKPATRICK AND CONDON, THE LINEAR SOLUTION FOR HYDRIDING OF U

!EXTERNALS RTSEC,BRACK TAKEN FROM NUMERICAL RECIPES FOR FORTRAN

DOUBLE PRECISION TEMP        !TEMPERATURE, K
DOUBLE PRECISION PRES        !HYDROGEN PARTIAL PRESSURE, PA,
DOUBLE PRECISION TIME,TIMESTEP  !TIME IN SECONDS,CALLED FROM MAIN AS HOURS
DOUBLE PRECISION TI             !TIME FOR FIRST SPALL, S
DOUBLE PRECISION DE             !GENERALIZED EFFECTIVE DIFFUSIVITY, M2/S
DOUBLE PRECISION VS             !SPALL FRONT VELOCITY, M/S
DOUBLE PRECISION S              !STOICHIOMETRY TERM, FOR UH3, S=3
DOUBLE PRECISION CO             !SOLUBILITY MOLAR FRACTION FOR FREE HYDROGEN IN U
DOUBLE PRECISION WG             !WEIGHT GAIN IN MOLE U M/S
DOUBLE PRECISION UC             !U MOLAR FRACTION FOR SPALL
DOUBLE PRECISION K1             !HYDRIDING RATE CONSANT,S^-1
DOUBLE PRECISION KP,KPH2,SIGKPH2    !WEIGHT GAIN MOLE U/M^2/S,H2 CONSUMED
DOUBLE PRECISION X1,X2          !BRACKETS FOR FINDING ROOT USTOP
DOUBLE PRECISION XACC           !ACCURACY IN ROOT FINDING FOR USTOP
DOUBLE PRECISION USTOP          !U MOLAR FRACTION FOR WHICH HYDRIDING REACTION RATE
                    !BECOMES ZERO BECAUSE HYDRIDING AN DEHYDRIDING RATES
                    !ARE IN BALANCE
DOUBLE PRECISION NN,N           !MAXIMUM AMOUNT OF FREE DIFFUSING H2 THAT THE URANIUM MATRIX CAN HOL
D
LOGICAL SUCCES
INTEGER I
DOUBLE PRECISION SAEXP,SIGSAEXP
DOUBLE PRECISION H2CONS,SIGH2CONS   !MOLES OF H2 CONSUMED IN TIMESTEP
DOUBLE PRECISION RTSEC

    !CONVERT FROM ATM TO PA
    S=3.D0
    TIME=3600.D0*TIMESTEP
    X1=0.00001D0
    X2=1.D0
    XACC=1.D-4

    NN=N(TEMP)

    CALL CZERO(TEMP,PRES,CO)
    !CALCULATE TI FOR TEMP.
    CALL TIS(TEMP,CO,TI)
    TI=ABS(TI)

    CALL DEQ(TEMP,PRES,NN,DE)
    CALL VSS(TEMP,TI,DE,S,VS)

    !GET BRACKETS TO FIND ROOTS FOR USTOP
    CALL zbrac(TEMP,CO,x1,x2,succes)
    USTOP=rtsec(TEMP,CO,x1,x2,xacc)

    !PLOT=VS*(1-USTOP)
    WG=S*VS*(1-USTOP)+CO*VS  !MOLE U M/S
    KP=8.D4*WG  !MOLE U/M^2/S
```

```
    !1.5 MOLES OF H2 CONSUMED FOR EACH MOLE OF U,CONVERT TO CM^2
    KPH2=1.5D0*KP/1.D4    !MOLES OF H2/CM^2/SEC
    SIGKPH2=0.75D0*KPH2

    !MULTIPLY BY TIME AND SAEXP
    H2CONS=TIME*SAEXP*KPH2
    CALL STATPROD(SAEXP,TIME,SIGSAEXP,KPH2,SIGKPH2,0.D0,H2CONS,SIGH2CONS)

    !1 MOLE OF H2 IS 2 GRAMS OR 2000MG
    !KPH2=KPH2*2000/1E4    !mG/M^2/S

END SUBROUTINE HYDRIDE


SUBROUTINE TIS(TEMP,CO,TI)

DOUBLE PRECISION TEMP,CO,TI,UC,K1

TI=-LOG(UC(TEMP))/(K1(TEMP)*CO)

END SUBROUTINE TIS

SUBROUTINE VSS(TEMP,TI,DE,S,VS)

DOUBLE PRECISION TEMP,TI,DE,S,K1,UE,VS

VS=(1/TI)*(DE/(S*K1(TEMP)*UE(TEMP)))**0.5

END SUBROUTINE VSS


SUBROUTINE FUNCTI(TEMP,PRES,U,FUNC)

DOUBLE PRECISION K1,K2,TEMP,U,FUNC,PRES,CO

CALL CZERO(TEMP,PRES,CO)

FUNC=-1.D0/(K1(TEMP)*CO*U+K2(TEMP)*(1.D0-U)**(1.D0/3.))

END SUBROUTINE FUNCTI


FUNCTION K1(TEMP)

DOUBLE PRECISION TEMP,K1
K1=10.4D0*EXP(1592.D0/TEMP)

END FUNCTION K1


FUNCTION K2(TEMP)

DOUBLE PRECISION TEMP,K2
K2=137.5D0*EXP(-4900.D0/TEMP)

END FUNCTION K2


SUBROUTINE CZERO(TEMP,PRES,CO)

DOUBLE PRECISION N,TEMP,PRES,CO,DENOM1,DENOM2,DENOM,NUM1,NUM

DENOM1=4.183D-6*PRES**0.5
DENOM2=DENOM1*EXP(-894.D0/TEMP)
DENOM=N(TEMP)+DENOM2

NUM1=DENOM2
NUM=N(TEMP)*NUM1

CO=NUM/DENOM

END SUBROUTINE CZERO
```

```
FUNCTION N(TEMP)

DOUBLE PRECISION N,TEMP

N=EXP(-2.362D0-2305.D0/TEMP)

END FUNCTION


FUNCTION D(TEMP)

DOUBLE PRECISION TEMP,D

D=1.9D-6*EXP(-5820.D0/TEMP)

END FUNCTION D


SUBROUTINE DEQ(TEMP,PRES,N,DE)

DOUBLE PRECISION TEMP,PRES,DE,N,D,CO

CALL CZERO(TEMP,PRES,CO)
DE=D(TEMP)*(N/(N-CO))**0.5
!PRINT*,'DE=',DE

END SUBROUTINE DEQ


FUNCTION UC(TEMP)

DOUBLE PRECISION TEMP,UC

UC=1.D0-(1.D0-0.989D0)**(0.5D0-0.5D0*TANH(-2950.D0*((1/TEMP)-0.00146D0)))

END FUNCTION UC

FUNCTION UE(TEMP)

DOUBLE PRECISION TEMP,UE,UC

UE=(UC(TEMP))**0.5

END FUNCTION UE


SUBROUTINE FUNCUSTOP(TEMP,CO,X,FUNCX)

DOUBLE PRECISION TEMP,CO,X,FUNCX,K2,K1

FUNCX=(K2(TEMP)*(1-X)**(1.D0/3.D0))-(K1(TEMP)*CO*X)

END SUBROUTINE FUNCUSTOP


 SUBROUTINE zbrac(TEMP,CO,x1,x2,succes)
      INTEGER NTRY
      DOUBLE PRECISION x1,x2,FACTOR,TEMP,CO
      PARAMETER (FACTOR=1.6,NTRY=50)
      INTEGER j
      DOUBLE PRECISION f1,f2
      LOGICAL succes
      if(x1.eq.x2)pause 'you have to guess an initial range in zbrac'
      CALL FUNCUSTOP(TEMP,CO,X1,F1)
      CALL FUNCUSTOP(TEMP,CO,X2,F2)
            succes=.true.
      do 11 j=1,NTRY
        if(f1*f2.lt.0.D0)return
        if(abs(f1).lt.abs(f2))then
          x1=x1+FACTOR*(x1-x2)
          CALL FUNCUSTOP(TEMP,CO,X1,F1)
        else
          x2=x2+FACTOR*(x2-x1)
          CALL FUNCUSTOP(TEMP,CO,X2,F2)
```

```
        endif
11    continue
      succes=.false.
      return
      END


        FUNCTION rtsec(TEMP,CO,x1,x2,xacc)
      INTEGER MAXIT
      DOUBLE PRECISION rtsec,x1,x2,xacc,TEMP,CO
      PARAMETER (MAXIT=30)
      INTEGER j
      DOUBLE PRECISION dx,f,fl,swap,xl
      CALL FUNCUSTOP(TEMP,CO,X1,FL)
      CALL FUNCUSTOP(TEMP,CO,X2,F)
       if(abs(fl).lt.abs(f))then
        rtsec=xl
        xl=x2
        swap=fl
        fl=f
        f=swap
      else
        xl=x1
        rtsec=x2
      endif
      do 11 j=1,MAXIT
        dx=(xl-rtsec)*f/(f-fl)
        xl=rtsec
        fl=f
        rtsec=rtsec+dx
        CALL FUNCUSTOP(TEMP,CO,RTSEC,F)
        if(abs(dx).lt.xacc.or.f.eq.0.)return

11    continue
      pause 'rtsec exceed maximum iterations'
      END FUNCTION RTSEC


      FUNCTION H2EQPP(TEMP)

      DOUBLE PRECISION TEMP,H2EQPP,PTORR

      !CALCULATE PRESSURE IN TORR
      PTORR=10**(-4590.D0/TEMP+9.39D0)

      !CONVERT TO PA
      H2EQPP=133.29D0*PTORR

      END FUNCTION H2EQPP



      SUBROUTINE PRESSURE(MOLES,SIGMOLES,TEMP,SIGTEMP,FREEVOL,PRES,SIGPRES)

      DOUBLE PRECISION MOLES,SIGMOLES
      DOUBLE PRECISION TEMP,SIGTEMP
      DOUBLE PRECISION FREEVOL
      DOUBLE PRECISION PRES,SIGPRES
      DOUBLE PRECISION COEF

      COEF=8.314/FREEVOL
      PRES=COEF*MOLES*TEMP
      CALL STATPROD(MOLES,COEF,SIGMOLES,TEMP,SIGTEMP,0.D0,PRES,SIGPRES)

      END SUBROUTINE PRESSURE


      SUBROUTINE MOLECONV(PRES,SIGPRES,FREEVOL,TEMP,SIGTEMP,MOLES,SIGMOLES)

      DOUBLE PRECISION MOLES,SIGMOLES
      DOUBLE PRECISION TEMP,SIGTEMP
      DOUBLE PRECISION FREEVOL
      DOUBLE PRECISION PRES,SIGPRES
      DOUBLE PRECISION COEF

      IF (PRES .GT. 0.D0)THEN
      COEF=FREEVOL/8.314
      MOLES=COEF*PRES/TEMP
      CALL STATDIV(PRES,COEF,SIGPRES,TEMP,SIGTEMP,0.D0,MOLES,SIGMOLES)
      ELSE
```

```
  MOLES=0.D0
  SIGMOLES=0.D0
  ENDIF
  END SUBROUTINE MOLECONV


SUBROUTINE H2MIN(TEMP,SIGTEMP,PH2MIN,SIGPH2MIN)

  DOUBLE PRECISION TEMP,SIGTEMP,PTORR,EXPO,SIGEXPO,SIGPTORR,PH2MIN,SIGPH2MIN
  DOUBLE PRECISION TERM1,SIGTERM1
  !CALCULATE PRESSURE IN TORR
  TERM1=-4590.D0/TEMP
  CALL STATDIV (-4590.D0,1.D0,0.D0,TEMP,SIGTEMP,0.D0,TERM1,SIGTERM1)
  EXPO=TERM1+9.39D0
  CALL STATSUM(TERM1,1.D0,SIGTERM1,9.39D0,1.D0,0.D0,0.D0,EXPO,SIGEXPO)

  PTORR=10**(EXPO)

  CALL STATPOW10(EXPO,SIGEXPO,1.D0,1.D0,PTORR,SIGPTORR)

  !CONVERT TO PA
  PH2MIN=133.29D0*PTORR
  SIGPH2MIN=133.29D0*SIGPTORR

  END
```

1	1.01E+05	4.81E+03	1.13E+01	3.81E-01	2.13E+00	4.47E-01	1.01E+05	4.81E+03
2	1.01E+05	4.81E+03	1.13E+01	3.81E-01	4.25E+00	6.40E-01	1.01E+05	4.81E+03
3	1.01E+05	4.81E+03	1.13E+01	3.81E-01	6.38E+00	7.93E-01	1.01E+05	4.81E+03
4	1.01E+05	4.81E+03	1.13E+01	3.81E-01	8.50E+00	9.27E-01	1.01E+05	4.81E+03
5	1.01E+05	4.81E+03	1.13E+01	3.81E-01	1.06E+01	1.05E+00	1.01E+05	4.81E+03
6	1.01E+05	4.81E+03	4.25E+00	1.43E-01	1.27E+01	1.16E+00	1.01E+05	4.81E+03
7	1.01E+05	4.81E+03	4.25E+00	1.43E-01	1.48E+01	1.27E+00	1.01E+05	4.81E+03
8	1.01E+05	4.81E+03	4.25E+00	1.43E-01	1.69E+01	1.38E+00	1.01E+05	4.81E+03
9	1.01E+05	4.81E+03	4.25E+00	1.43E-01	1.90E+01	1.48E+00	1.01E+05	4.81E+03
10	1.01E+05	4.81E+03	4.25E+00	1.43E-01	2.11E+01	1.57E+00	1.01E+05	4.81E+03
11	1.01E+05	4.81E+03	4.25E+00	1.43E-01	2.32E+01	1.67E+00	1.01E+05	4.81E+03
12	1.01E+05	4.81E+03	4.25E+00	1.43E-01	2.53E+01	1.76E+00	1.01E+05	4.81E+03
13	1.01E+05	4.81E+03	4.25E+00	1.43E-01	2.74E+01	1.85E+00	1.01E+05	4.81E+03
14	1.01E+05	4.81E+03	4.25E+00	1.43E-01	2.95E+01	1.94E+00	1.01E+05	4.81E+03
15	1.01E+05	4.81E+03	4.25E+00	1.43E-01	3.16E+01	2.02E+00	1.01E+05	4.80E+03
16	1.01E+05	4.81E+03	4.25E+00	1.43E-01	3.37E+01	2.11E+00	1.01E+05	4.80E+03
17	1.01E+05	4.81E+03	4.25E+00	1.43E-01	3.59E+01	2.19E+00	1.01E+05	4.80E+03
18	1.01E+05	4.80E+03	4.25E+00	1.43E-01	3.80E+01	2.28E+00	1.01E+05	4.80E+03
19	1.01E+05	4.80E+03	4.25E+00	1.43E-01	4.01E+01	2.36E+00	1.01E+05	4.80E+03
20	1.01E+05	4.80E+03	4.25E+00	1.43E-01	4.22E+01	2.44E+00	1.01E+05	4.80E+03
21	1.01E+05	4.80E+03	4.25E+00	1.43E-01	4.43E+01	2.52E+00	1.01E+05	4.80E+03
22	1.01E+05	4.80E+03	4.25E+00	1.43E-01	4.64E+01	2.60E+00	1.01E+05	4.80E+03
23	1.01E+05	4.80E+03	4.25E+00	1.43E-01	4.85E+01	2.68E+00	1.01E+05	4.80E+03
24	1.01E+05	4.80E+03	4.25E+00	1.43E-01	5.06E+01	2.76E+00	1.01E+05	4.80E+03
25	1.01E+05	4.80E+03	4.25E+00	1.43E-01	5.27E+01	2.84E+00	1.01E+05	4.80E+03
26	1.01E+05	4.80E+03	4.25E+00	1.43E-01	5.48E+01	2.92E+00	1.01E+05	4.80E+03
27	1.01E+05	4.80E+03	4.25E+00	1.43E-01	5.69E+01	3.00E+00	1.01E+05	4.80E+03
28	1.01E+05	4.80E+03	4.25E+00	1.43E-01	5.90E+01	3.08E+00	1.01E+05	4.80E+03
29	1.01E+05	4.80E+03	4.25E+00	1.43E-01	6.11E+01	3.16E+00	1.01E+05	4.80E+03
30	1.01E+05	4.80E+03	4.25E+00	1.43E-01	6.32E+01	3.23E+00	1.01E+05	4.80E+03
31	1.01E+05	4.80E+03	4.25E+00	1.43E-01	6.53E+01	3.31E+00	1.01E+05	4.80E+03
32	1.01E+05	4.80E+03	4.25E+00	1.43E-01	6.74E+01	3.39E+00	1.01E+05	4.80E+03
33	1.01E+05	4.80E+03	4.25E+00	1.43E-01	6.95E+01	3.46E+00	1.01E+05	4.80E+03
34	1.01E+05	4.80E+03	4.25E+00	1.43E-01	7.16E+01	3.54E+00	1.01E+05	4.80E+03
35	1.01E+05	4.80E+03	4.25E+00	1.43E-01	7.37E+01	3.62E+00	1.01E+05	4.80E+03
36	1.01E+05	4.80E+03	4.25E+00	1.43E-01	7.58E+01	3.69E+00	1.01E+05	4.80E+03

37	1.01E+05	4.80E+03	4.25E+00	1.43E-01	7.79E+01	3.77E+00	1.01E+05	4.80E+03
38	1.01E+05	4.80E+03	4.25E+00	1.43E-01	8.00E+01	3.84E+00	1.01E+05	4.80E+03
39	1.01E+05	4.80E+03	4.25E+00	1.43E-01	8.21E+01	3.92E+00	1.01E+05	4.80E+03
40	1.01E+05	4.80E+03	4.25E+00	1.43E-01	8.42E+01	3.99E+00	1.01E+05	4.80E+03
41	1.01E+05	4.80E+03	4.25E+00	1.43E-01	8.63E+01	4.07E+00	1.01E+05	4.80E+03
42	1.01E+05	4.80E+03	4.25E+00	1.43E-01	8.84E+01	4.14E+00	1.01E+05	4.80E+03
43	1.01E+05	4.80E+03	4.25E+00	1.43E-01	9.05E+01	4.22E+00	1.01E+05	4.80E+03
44	1.01E+05	4.80E+03	4.25E+00	1.43E-01	9.26E+01	4.29E+00	1.01E+05	4.80E+03
45	1.01E+05	4.80E+03	4.25E+00	1.43E-01	9.47E+01	4.36E+00	1.01E+05	4.80E+03
46	1.01E+05	4.80E+03	4.25E+00	1.43E-01	9.68E+01	4.44E+00	1.01E+05	4.80E+03
47	1.01E+05	4.80E+03	4.25E+00	1.43E-01	9.89E+01	4.51E+00	1.01E+05	4.80E+03
48	1.01E+05	4.80E+03	4.25E+00	1.43E-01	1.01E+02	4.59E+00	1.01E+05	4.80E+03
49	1.01E+05	4.80E+03	4.25E+00	1.43E-01	1.03E+02	4.66E+00	1.01E+05	4.80E+03
50	1.01E+05	4.80E+03	4.25E+00	1.43E-01	1.05E+02	4.73E+00	1.01E+05	4.80E+03

Appendix D: Executable file and output for dry interim storage regime

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$FREEFORM

!PROGRAM NAME DRYO+RAD+HYD7-19.F90
!PROGRAM NAME DRYO+RAD+HYD7-17.F90
!MODIFIED 7/9/97,7/13 TO ADD RADIOLYSIS, 7/17 TO ADD HYDRIDE MODEL
!CALCULATES CORROSION RATE AND OXYGEN CONSUMPTIONI IN DRY AIR OR OXYGEN
!INCLUDES RADIOLYSIS PRODUCTION

  INTEGER I,K,Z,J,YR,MO,DAY,STEPS   !EACH LOOP IS 1HOUR TIME STEP DUE TO REACTION RATE UNITS
  DOUBLE PRECISION TEMP,SIGTEMP     !TEMPERATURE IN KELVIN,STD DEV
  DOUBLE PRECISION SAEXP,SIGSAEXP   !TOTAL EXPOSED SURFACE AREA
  DOUBLE PRECISION UCORR,SIGUCORR   !TOTAL CORROSION IN MG U
  DOUBLE PRECISION PO2,SIGPO2               !PARTIAL PRESSURE OF OXYGEN,PA
  DOUBLE PRECISION TOTPRES,SIGTOTPRES   !TOTAL SYSTEM PRESSURE, PA,INCLUDES OTHER GASES
  DOUBLE PRECISION MOLESTOT,SIGMOLESTOT !TOTAL MOLES OF GAS
  DOUBLE PRECISION MOLESIN,SIGMOLESIN   !TOTAL MOLES OF INERT GAS
  DOUBLE PRECISION FREEVOL          !FREE VOLUME IN M^3
  DOUBLE PRECISION MOLESO2,SIGMOLESO2 !MOLES OF O2 IN FREE VOLUME
  DOUBLE PRECISION SLUDGE,SIGSLUDGE !AMOUNT OF SLUDGE, KG
  DOUBLE PRECISION MOLESH2O,SIGMOLESH2O !MOLES OF BOUND AND CHEMISORBED WATER FOR RADIOLYSIS
  DOUBLE PRECISION DECAY,SIGDECAY   !DECAY POWER FOR RADIOLYSIS, W
  DOUBLE PRECISION MOLESH2,SIGMOLESH2 !MOLES OF H2 IN FREE VOLUME
  DOUBLE PRECISION O2RADPROD,SIGO2RADPROD,H2RADPROD,SIGH2RADPROD !H2 AND O2 PRODUCED IN RADIOLYSIS
  DOUBLE PRECISION H2ORADCONS,SIGH2ORADCONS !MOLES H2O CONSUMED IN RADIOLYSIS
  DOUBLE PRECISION NEWO2,SIGNEWO2,NEWH2,SIGNEWH2,NEWH2O,SIGNEWH2O,NEWTOT,SIGNEWTOT
  DOUBLE PRECISION TIMESTEP          !INPUT LENGTH OF TIMESTEP IN HOURS
  DOUBLE PRECISION PH2,SIGPH2   !HYDROGEN PARTIAL PRESSURE IN Pa
  DOUBLE PRECISION H2EQPP           !EQUILIBRIUM H2 PARTIAL PRES BELOW WHICH HYDRIDING DOES NOT OCCU
R
  DOUBLE PRECISION H2HYDCONS,SIGH2HYDCONS   !MOLES OF H2 CONS IN HYDRIDING
  DOUBLE PRECISION O2CORRCONS,SIGO2CORRCONS !MOLES OF O2 CONSUMED IN CORROSION
  DOUBLE PRECISION TEMP1,SIGTEMP1,TEMP2,SIGTEMP2
  DOUBLE PRECISION FREEH2O,SIGFREEH2O,PH2MIN,SIGPH2MIN,MOLESH2MIN,SIGMOLESH2MIN

  !OPEN(1,FILE='c:\users\lisas\OUTPUT\DRY\DRYOUT25.TXT')
  !OPEN(1,FILE='c:\users\lisas\OUTPUT\\DRY\DRY722.TXT')
  !OPEN(2,FILE='c:\users\lisas\OUTPUT\DRY\DRYOUT50.TXT')
  !OPEN(3,FILE='C:\USERS\LISAS\OUTPUT\DRY\DRYOUT75.TXT')
  !OPEN(4,FILE='C:\USERS\LISAS\OUTPUT\DRY\DRYOUT100.TXT')
  OPEN(1,FILE='C:\USERS\LISAS\OUTPUT\DRY\DRYOUT.TXT')

  CALL GETDAT(YR,MO,DAY)
  WRITE(1,*)'FILE GENERATED ON ',MO,'-',DAY,'-',YR
  PRINT*,'FILE'

!SET PARAMETERS
TEMP=343.D0
SIGTEMP=1.D0
PO2=0.D0
SIGPO2=0.D0
TOTPRES=1.013E5
SIGTOTPRES=100.D0
FREEVOL=0.6D0
SLUDGE=50.D0
SIGSLUDGE=50.D0
FREEH2O=0.0D0
SIGFREEH2O=0.0D0
DECAY=550.D0
SIGDECAY=100.D0
TIMESTEP=1.D0
STEPS=50
SAEXP=60000.D0
SIGSAEXP=10000.D0

!CALCULATE MINIMUM H2 PARTIAL PRESSURE
CALL H2MIN(TEMP,SIGTEMP,PH2MIN,SIGPH2MIN)

!CALCULATE MIN MOLES OF H2
CALL MOLECONV(PH2MIN,SIGPH2MIN,FREEVOL,TEMP,SIGTEMP,MOLESH2MIN,SIGMOLESH2MIN)

!CALCULATE AMOUNT OF WATER IN SLUDGE
CALL WATER(SLUDGE,SIGSLUDGE,FREEH2O,SIGFREEH2O,MOLESH2O,SIGMOLESH2O)

!CALCULATE INITIAL MOLES OF GAS AT FILL ASSUMING FILLED AT 25C
CALL MOLECONV(TOTPRES,SIGTOTPRES,FREEVOL,298.D0,1.D0,MOLESTOT,SIGMOLESTOT)

!CALCULATE INITIAL MOLES OF O2 AND H2,STD DEV ASSUMES COVARIANCE 1. FOR T AND PO2
IF(PO2.GT.0.D0)THEN
    CALL MOLECONV(PO2,SIGPO2,FREEVOL,298.D0,1.D0,MOLESO2,SIGMOLESO2)
ELSE
```

```
    MOLESO2=0.D0
    SIGMOLESO2=0.D0
ENDIF
!INITIAL AMOUNT OF H2 IS MINIMUM
PH2=PH2MIN
SIGPH2=SIGPH2MIN
MOLESH2=MOLESH2MIN
SIGMOLESH2=SIGMOLESH2MIN

!CALCULATE MOLES OF INERT GAS
TEMP1=MOLESO2+MOLESH2
CALL STATSUM(MOLESO2,1.D0,SIGMOLESO2,MOLESH2,1.D0,SIGMOLESH2,0.D0,TEMP1,SIGTEMP1)

MOLESIN=MOLESTOT-TEMP1
CALL STATSUM(MOLESTOT,1.D0,SIGMOLESTOT,TEMP1,-1.D0,TEMP1,0.D0,MOLESIN,SIGMOLESIN)


DO 10 I=1,STEPS

    !CALCULATE CORROSION RATE IN MG
    !O2 PRODUCED BY RADIOLYSIS, CONSUMED BY CORROSION
    !WATER CONSUMED BY RADIOLYSIS
    !H2 PRODUCED BY RADIOLYSIS, CONSUMED BY HYDRIDING
    CALL UDRYAIR(TEMP,SIGTEMP,TIMESTEP,SAEXP,SIGSAEXP,UCORR,SIGUCORR)

    !CALCULATE THE O2 CONSUMED IN CORROSION
    CALL O2PRES(UCORR,SIGUCORR,O2CORRCONS,SIGO2CORRCONS)
    !PRINT*,'NEWO2= ',NEWO2

    !CHECK TO SEE IF PARTIAL PRESS OF H2 IS ABOVE EQ PRESSURE OVER HYDRIDE.
    !IF SO, CALCULATE THE H2 CONSUMED IN HYDRIDING, OTHERWISE NO H2 CONSUMED
    IF (PH2 .GT. H2EQPP(TEMP))THEN
        CALL HYDRIDE(TEMP,PH2,TIMESTEP,SAEXP,SIGSAEXP,H2HYDCONS,SIGH2HYDCONS)
    ELSE
        H2HYDCONS=0.D0
        SIGH2HYDCONS=0.D0
    ENDIF


!CHECK THAT H2CONS IS NOT LOWER THAN EQUILIBRIUM LIMIT
    TEMP1=MOLESH2-H2HYDCONS
    IF ((TEMP1 .LE. MOLESH2MIN).AND.(H2HYDCONS.GT.0.D0))THEN
        H2HYDCONS=MOLESH2-MOLESH2MIN
        CALL STATSUM(MOLESH2,1.D0,SIGMOLESH2,MOLESH2MIN,-1.D0,SIGMOLESH2MIN,0.D0,H2HYDCONS,SIGH2HYDC
ONS)
    ENDIF

    !IF BOUND WATER IS PRESENT, CALL RADIOL TO FIND GENERATED O2 AND H2 AND CONSUMED WATER.
    IF (MOLESH2O .GT. 0.D0)THEN

    CALL RADIOL(SLUDGE,SIGSLUDGE,DECAY,SIGDECAY,TIMESTEP,H2RADPROD,SIGH2RADPROD,O2RADPROD,SIGO2RADPR
OD,H2ORADCONS,SIGH2ORADCONS)

    IF(H2ORADCONS .LT. MOLESH2O)THEN
        NEWH2O=MOLESH2O-H2ORADCONS
        CALL STATSUM(MOLESH2O,1.D0,0.D0,H2ORADCONS,-1.D0,0.D0,0.D0,NEWH2O,SIGNEWH2O)
        MOLESH2O=NEWH2O
        SIGMOLESH2O=SIGNEWH2O
    ELSE
        H2RADPROD=2.D0/3.D0*MOLESH2O
        SIGH2RADPROD=2.D0/3.D0*SIGMOLESH2O
        O2RADPROD=0.5D0*H2RADPROD
        SIGO2RADPROD=0.5D0*SIGH2RADPROD
        MOLESH2O=0.D0
        SIGMOLESH2O=1.D-6
    ENDIF

    !CALCULATE NEW AMOUNT OF HYDROGEN--IF H2HYDCONS WAS ALTERED ABOVE, STDDEV MUST BE CALCULATED
    !DIFFERENTLY TO AVOID INCORPORATING SIGMOLESH2 TWICE
    IF (TEMP1 .LE. MOLESH2MIN)THEN
        NEWH2=H2RADPROD+MOLESH2MIN
        CALL STATSUM(H2RADPROD,1.D0,SIGH2RADPROD,MOLESH2MIN,1.D0,SIGMOLESH2MIN,0.D0,NEWH2,SIGNEWH2)
        MOLESH2=NEWH2
        SIGMOLESH2=SIGNEWH2
    ELSE
        TEMP2=H2RADPROD-H2HYDCONS
        CALL STATSUM(H2RADPROD,1.D0,SIGH2RADPROD,H2HYDCONS,-1.D0,SIGH2HYDCONS,0.D0,TEMP2,SIGTEMP2)
        NEWH2=MOLESH2+TEMP2
        CALL STATSUM(MOLESH2,1.D0,SIGMOLESH2,TEMP2,1.D0,SIGTEMP2,0.D0,NEWH2,SIGNEWH2)
        MOLESH2=NEWH2
```

```
        SIGMOLESH2=SIGNEWH2
    ENDIF

    !CALCULATE NEW AMOUNT OF OXYGEN
    IF(O2CORRCONS .LT. MOLESO2)THEN
        TEMP1=O2RADPROD-O2CORRCONS
        CALL STATSUM(O2RADPROD,1.D0,SIGO2RADPROD,O2CORRCONS,-1.D0,SIGO2CORRCONS,0.D0,TEMP1,SIGTEMP1)
        NEWO2=MOLESO2+TEMP1
        CALL STATSUM(MOLESO2,1.D0,SIGMOLESO2,TEMP1,1.D0,SIGTEMP1,0.D0,NEWO2,SIGNEWO2)
        MOLESO2=NEWO2
        SIGMOLESO2=SIGNEWO2
    ELSE
        MOLESO2=O2RADPROD
        SIGMOLESO2=SIGO2RADPROD
    ENDIF


    !IF NO WATER IS PRESENT, NO H2 OR O2 GENERATED, ONLY O2 CONSUMED BY CORROSION,AND H2
    !CONSUMED BY HYDRIDING
    ELSE
        NEWO2=MOLESO2-O2CORRCONS
        CALL STATSUM(MOLESO2,1.D0,SIGMOLESO2,O2CORRCONS,-1.D0,SIGO2CORRCONS,0.D0,NEWO2,SIGNEWO2)
        NEWH2=MOLESH2-H2HYDCONS
        CALL STATSUM(MOLESH2,1.D0,SIGMOLESH2,H2HYDCONS,-1.D0,SIGH2HYDCONS,0.D0,NEWH2,SIGNEWH2)
        MOLESH2=NEWH2
        SIGMOLESH2=SIGNEWH2
        MOLESO2=NEWO2
        SIGMOLESO2=SIGNEWO2
    ENDIF

    !CALCULATE TOTAL PRESSURE
    TEMP1=MOLESO2+MOLESH2
    CALL STATSUM(MOLESO2,1.D0,SIGMOLESO2,MOLESH2,1.D0,SIGMOLESH2,0.D0,TEMP1,SIGTEMP1)
    NEWTOT=MOLESIN+TEMP1
    CALL STATSUM(MOLESIN,1.D0,SIGMOLESIN,TEMP1,1.D0,SIGTEMP1,0.D0,NEWTOT,SIGNEWTOT)
    MOLESTOT=NEWTOT
    SIGMOLESTOT=SIGNEWTOT

    CALL PRESSURE(MOLESH2,SIGMOLESH2,TEMP,SIGTEMP,FREEVOL,PH2,SIGPH2)
    CALL PRESSURE(MOLESO2,SIGMOLESO2,TEMP,SIGTEMP,FREEVOL,PO2,SIGPO2)
    CALL PRESSURE(MOLESTOT,SIGMOLESTOT,TEMP,SIGTEMP,FREEVOL,TOTPRES,SIGTOTPRES)

    WRITE(1,50)I,PH2,SIGPH2,PO2,SIGPO2,TOTPRES,SIGTOTPRES

    !USE THIS LOOP TO PRINT OUT PRESSURE JUST AT YEARLY INTERVALS
    !DO 20 K=1,50
    !Z=K*8760
    !IF (I.EQ.Z)THEN
    !WRITE(1,50)K,PH2,SIGPH2,PO2,SIGPO2,TOTPRES,SIGTOTPRES
    !ENDIF
!20 !CONTINUE
!   IF(MOLESO2 .EQ. 0.D0)THEN
!       PRINT*,'ALL O2 CONSUMED IN ',I,'HOURS'
!       EXIT
!   ENDIF

10 CONTINUE

5  CONTINUE

50 FORMAT(I3,6(2X,E16.8))

END


SUBROUTINE UDRYAIR(TEMP,SIGTEMP,TIME,SAEXP,SIGSAEXP,UCORR,SIGUCORR)

!THIS SUBROUTINE CALCULATES THE REACTION RATE IN MG U/HR FOR
!A DRY AIR REACTION USING THE RELATIONSHIP GENERATED FROM
!DATA FROM PEARCE, 10/89.

DOUBLE PRECISION TEMP,SIGTEMP   !TEMPERATURE IN KELVIN, CALLED FROM MAIN        !REACTION RATE IN mg
 U/cm^2/hr
DOUBLE PRECISION SAEXP,SIGSAEXP !TOTAL EXPOSED FUEL AREA, CALLED FROM MAIN
DOUBLE PRECISION UCORR,SIGUCORR !TOTAL WEIGHT GAIN, MG/HR,
DOUBLE PRECISION M,SIGM         !SLOPE AND STD DEV
DOUBLE PRECISION B,SIGB         !INTERCEPT
DOUBLE PRECISION ADD
DOUBLE PRECISION TIME           !TIMESTEP IN HOURS
```

```
M=-3520.D0
SIGM=100.D0
B=6.71D0
SIGB=0.02D0
ADD=0.D0

CALL CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,UCORR,SIGUCORR)

END SUBROUTINE



SUBROUTINE CORRODE(M,SIGM,B,SIGB,SAEXP,SIGSAEXP,TEMP,SIGTEMP,TIME,ADD,UCORR,SIGUCORR)

DOUBLE PRECISION M,SIGM      !SLOPE AND STD DEV
DOUBLE PRECISION B,SIGB      !INTERCEPT AND STD DEV
DOUBLE PRECISION SAEXP,SIGSAEXP !EXPOSED FUEL AREA,CM^2
DOUBLE PRECISION TEMP,SIGTEMP   !TEMPERATURE IN KELVIN
DOUBLE PRECISION TIME           !TIME STEP IN HOURS
DOUBLE PRECISION ADD
DOUBLE PRECISION KP,SIGKP       !REACTION RATE IN MG/HR/CM^2
DOUBLE PRECISION UCORR,SIGUCORR     !REACTION RATE IN MG/HR
DOUBLE PRECISION EXP1,EXP2,EXP3,SIGEXP1,SIGEXP2,SIGEXP3

EXP1=M/TEMP
CALL STATDIV(M,1.0D0,SIGM,TEMP,SIGTEMP,0.D0,EXP1,SIGEXP1)

EXP2=B+EXP1
CALL STATSUM(B,1.D0,SIGB,EXP1,1.0D0,SIGEXP1,0.D0,EXP2,SIGEXP2)

EXP3=EXP2+ADD
CALL STATSUM(EXP2,1.D0,SIGEXP2,ADD,1.0D0,0.D0,0.D0,EXP3,SIGEXP3)

!CALC REACTION RATE AND VARIANCE
KP=10**EXP3
CALL STATPOW10(EXP3,SIGEXP3,1.D0,1.D0,KP,SIGKP)

!MULTIPLY BY SURFACE AREA, STD DEV ASSUMES NO CORRELATION
UCORR=TIME*KP*SAEXP
CALL STATPROD(KP,TIME,SIGKP,SAEXP,SIGSAEXP,0.D0,UCORR,SIGUCORR)

END SUBROUTINE CORRODE


SUBROUTINE O2PRES(UCORR,SIGUCORR,O2CONS,SIGO2CONS)

DOUBLE PRECISION UCORR,SIGUCORR     !REACTION RATE IN MG O
DOUBLE PRECISION O2CONS,SIGO2CONS

!CALC MOLES OF O2 CONSUMED
O2CONS=UCORR*1E-3/32.D0
SIGO2CONS=SIGUCORR*1E-3/32.D0

END SUBROUTINE O2PRES



SUBROUTINE STATSUM(VAR1,COEF1,SIG1,VAR2,COEF2,SIG2,SIG12,X,SIGX)

!USE TO CALCULATE STD DEV OF X IN EQUATION X=COEF1*VAR1+COEF2*VAR2 WITH
!STD DEVS OF VAR1 AND VAR2 EQUAL TO SIG1 AND SIG2, RESPECTIVELY AND
'COVARIANCE SIG12

DOUBLE PRECISION VAR1,VAR2
DOUBLE PRECISION COEF1,COEF2
DOUBLE PRECISION SIG1,SIG2,SIG12
DOUBLE PRECISION X,SIGX

SIGX=SQRT(COEF1**2*SIG1**2+COEF2**2*SIG2**2+2.0D0*COEF1*COEF2*SIG12**2)

END SUBROUTINE STATSUM



SUBROUTINE STATPROD(VAR1,COEF1,SIG1,VAR2,SIG2,SIG12,X,SIGX)

!USE TO CALCULATE THE STD DEV OF X IN EQUATION X=COEF1*VAR1*VAR2 WITH
!STD DEVS OF VAR1 AND VAR2 EQUAL TO SIG1 AND SIG2, RESPECTIVELY AND
```

```
!COVARIANCE SIG12

DOUBLE PRECISION VAR1,VAR2
DOUBLE PRECISION COEF1
DOUBLE PRECISION SIG1,SIG2
DOUBLE PRECISION X,SIGX,SIG12

SIGX=SQRT(X**2*((SIG1/VAR1)**2+(SIG2/VAR2)**2+2.0D0*SIG12**2.0D0/(VAR1*VAR2)))

END SUBROUTINE STATPROD


SUBROUTINE STATDIV(VAR1,COEF1,SIG1,VAR2,SIG2,SIG12,X,SIGX)

!USE TO CALCULATE THE STD DEV OF X IN EQUATION X=COEF1*(VAR1/VAR2) WITH
!STD DEVS OF VAR1 AND VAR2 EQUAL TO SIG1 AND SIG2, RESPECTIVELY, AND
!COVARIANCE SIG12

DOUBLE PRECISION VAR1,VAR2
DOUBLE PRECISION COEF1
DOUBLE PRECISION SIG1,SIG2,SIG12
DOUBLE PRECISION X,SIGX

SIGX=SQRT(ABS(X**2*((SIG1/VAR1)**2+(SIG2/VAR2)**2-2.0D0*SIG12**2.0D0/(VAR1*VAR2))))

END SUBROUTINE STATDIV


SUBROUTINE STATPOW(VAR,SIG,COEF,EXPO,X,SIGX)

!USE TO CALCULATE STD DEV OF X IN EQUATION: X=COEF*VAR**EXPO WITH STD OF
!VAR EQUAL TO SIG

DOUBLE PRECISION VAR,SIG,COEF,EXPO,X,SIGX

SIGX=X*EXPO*SIG/VAR

END SUBROUTINE STATPOW


SUBROUTINE STATEXPO(VAR,SIG,COEF1,COEF2,X,SIGX)

!USED TO CALCULATE STD DEV OF X IN EQUATION X=COEF1*e**(COEF2*VAR)
!WITH STD DEV OF VAR EQUAL TO SIG

DOUBLE PRECISION VAR,SIG,COEF1,COEF2,X,SIGX

SIGX=X*COEF2*SIG

END SUBROUTINE STATEXPO


SUBROUTINE STATPOW10(VAR,SIG,COEF1,COEF2,X,SIGX)

!USED TO CALCULATE STD DEV OF X IN EQUATION X=COEF1*10**(COEF2*VAR)
!WITH STD DEV OF VAR EQUAL TO SIQ

DOUBLE PRECISION VAR,SIG,COEF1,COEF2,X,SIGX

SIGX=ABS(X*2.303D0*COEF2*SIG)

END SUBROUTINE STATPOW10


SUBROUTINE WATER(SLUDGE,SIGSLUDGE,FREEH2O,SIGFREEH2O,MOLESH2O,SIGMOLESH2O)

!CALCULATES THE MOLES OF WATER (FREE,BOUND,CHEMISORBED) AVAILABLE FOR RADIOLYSIS

DOUBLE PRECISION SLUDGE,SIGSLUDGE   !KGS OF SLUDGE IN THE Mco
DOUBLE PRECISION FREEH2O,SIGFREEH2O !KGS OF FREE WATER
DOUBLE PRECISION BOUNDFRAC,SIGBOUNDFRAC !FRACTION OF SLUDGE THAT IS BOUND WATER
DOUBLE PRECISION CHEMFRAC,SIGCHEMFRAC   !FRACTION OF SLUDGE THAT IS CHEMISORBED WATER
DOUBLE PRECISION BOUNDKG,SIGBOUNDKG,CHEMKG,SIGCHEMKG,ABSKG,SIGABSKG
DOUBLE PRECISION MOLESH2O,SIGMOLESH2O !TOTAL INITAL MOLES OF WATER IN MCO

!PHYSICAL PARAMETERS
BOUNDFRAC=0.0D0     !NO BOUND WATER IN THIS CASE
```

```
SIGBOUNDFRAC=0.01D0
CHEMFRAC=0.015D0
SIGCHEMFRAC=0.003D0

!CALCULATE KGS OF BOUND AND CHEMISORBED WATER
BOUNDKG=0.0D0
SIGBOUNDKG=0.0D0
!CALL STATPROD(SLUDGE,1.0D0,SIGSLUDGE,BOUNDFRAC,SIGBOUNDFRAC,0.D0,BOUNDKG,SIGBOUNDKG)
CHEMKG=SLUDGE*CHEMFRAC
CALL STATPROD(SLUDGE,1.0D0,SIGSLUDGE,CHEMFRAC,SIGCHEMFRAC,0.D0,CHEMKG,SIGCHEMKG)
!ADD BOUND AND CHEMISORBED WATER
ABSKG=BOUNDKG+CHEMKG
CALL STATSUM(BOUNDKG,1.0D0,SIGBOUNDKG,CHEMKG,1.0D0,SIGCHEMKG,0.D0,ABSKG,SIGABSKG)

!ADD TOTAL TO FREE WATER AND CONVERT TO MOLES WITH 1/0.018 MOLES/KG
MOLESH2O=55.56D0*(FREEH2O+ABSKG)
CALL STATSUM(FREEH2O,55.56D0,SIGFREEH2O,ABSKG,55.56D0,SIGABSKG,0.D0,MOLESH2O,SIGMOLESH2O)

!PRINT*,MOLESH2O,' MOLES OF H2O +-',SIGMOLESH2O

END SUBROUTINE WATER


SUBROUTINE RADIOL(SLUDGE,SIGSLUDGE,DECAY,SIGDECAY,TIMESTEP,RADH2,SIGRADH2,RADO2,SIGRADO2,H2OCONS,SIG
H2OCONS)

!CALCULATES H2 AND O2 PRODUCED AND H2O CONSUMED.  G VALUE FOR SLUDGE IS 0.001 MOLECULES/100 Ev OR
!1.04E-10 MOLES/W/S
DOUBLE PRECISION GR !MOLES OF HYDROGEN PRODUCED PER SECOND PER WATT OF E ABSORBED,
DOUBLE PRECISION DECAY,SIGDECAY      !DECAY POWER IN WATTS
DOUBLE PRECISION FRAC,SIGFRAC
DOUBLE PRECISION RADH2,SIGRADH2,RADO2,SIGRADO2  !MOLES OF H2 AND O2 PRODUCED BY RADIOLYSIS
DOUBLE PRECISION H2OCONS,SIGH2OCONS,SLUDGE,SIGSLUDGE
DOUBLE PRECISION TIME                !TIME IN SECONDS
DOUBLE PRECISION TIMESTEP

TIME=TIMESTEP*3600
!PHYSICAL PARAMETERS

GR=1.04D-10

!CALCULATE FRACTION OF ENERGY DEPOSITED IN SLUDGE
!FRACTION--
FRAC=9.75D-5*SLUDGE*DECAY
CALL STATPROD(SLUDGE,9.75D-5,SIGSLUDGE,DECAY,SIGDECAY,0.0D0,FRAC,SIGFRAC)

RADH2=GR*TIME*FRAC
SIGRADH2=GR*TIME*SIGFRAC

!0.5 MOLES OF O2 PRODUCED PER MOLE OF H2 PRODUCED
RADO2=0.5D0*RADH2
SIGRADO2=0.5D0*SIGRADH2

H2OCONS=RADH2
SIGH2OCONS=SIGRADH2

END SUBROUTINE RADIOL


SUBROUTINE HYDRIDE(TEMP,PRES,TIMESTEP,SAEXP,SIGSAEXP,H2CONS,SIGH2CONS)


!CALCULATES THE WEIGHT GAIN AND SPALL FRONT VELOCITY IN U HYDRIDING.
!MODEL TAKEN FROM KIRKPATRICK AND CONDON, THE LINEAR SOLUTION FOR HYDRIDING OF U

!EXTERNALS QGAUSS,RTSEC,BRACK TAKEN FROM NUMERICAL RECIPES FOR FORTRAN

DOUBLE PRECISION TEMP        !TEMPERATURE, K
DOUBLE PRECISION PRES,TOTPRES   !HYDROGEN PARTIAL PRESSURE, PA,CALLED FROM MAIN AS ATM
DOUBLE PRECISION TIME,TIMESTEP  !TIME IN SECONDS,CALLED FROM MAIN AS HOURS
DOUBLE PRECISION TI             !TIME FOR FIRST SPALL, S
DOUBLE PRECISION DE             !GENERALIZED EFFECTIVE DIFFUSIVITY, M2/S
DOUBLE PRECISION VS             !SPALL FRONT VELOCITY, M/S
DOUBLE PRECISION S              !STOICHIOMETRY TERM, FOR UH3, S=3
DOUBLE PRECISION CO             !SOLUBILITY MOLAR FRACTION FOR FREE HYDROGEN IN U
DOUBLE PRECISION WG             !WEIGHT GAIN IN MOLE U M/S
DOUBLE PRECISION UC             !U MOLAR FRACTION FOR SPALL
DOUBLE PRECISION K1             !HYDRIDING RATE CONSANT,S^-1
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DOUBLE PRECISION KP,KPH2,SIGKPH2     !WEIGHT GAIN MOLE U/M^2/S,H2 CONSUMED
DOUBLE PRECISION X1,X2               !BRACKETS FOR FINDING ROOT USTOP
DOUBLE PRECISION XACC                !ACCURACY IN ROOT FINDING FOR USTOP
DOUBLE PRECISION USTOP               !U MOLAR FRACTION FOR WHICH HYDRIDING REACTION RATE
                    !BECOMES ZERO BECAUSE HYDRIDING AN DEHYDRIDING RATES
                    !ARE IN BALANCE
DOUBLE PRECISION NN,N                !MAXIMUM AMOUNT OF FREE DIFFUSING H2 THAT THE URANIUM MATRIX CAN HOL
D
LOGICAL SUCCES
INTEGER I
DOUBLE PRECISION SAEXP,SIGSAEXP
DOUBLE PRECISION H2CONS,SIGH2CONS    !MOLES OF H2 CONSUMED IN TIMESTEP
DOUBLE PRECISION RTSEC

    !CONVERT FROM ATM TO PA
    S=3.D0
    TIME=3600.D0*TIMESTEP
    X1=0.00001D0
    X2=1.D0
    XACC=1.D-4

    NN=N(TEMP)

    CALL CZERO(TEMP,PRES,CO)
    !CALCULATE TI FOR TEMP.
    CALL TIS(TEMP,CO,TI)
    TI=ABS(TI)

    CALL DEQ(TEMP,PRES,NN,DE)
    CALL VSS(TEMP,TI,DE,S,VS)

    !GET BRACKETS TO FIND ROOTS FOR USTOP
    CALL zbrac(TEMP,CO,x1,x2,succes)
    USTOP=rtsec(TEMP,CO,x1,x2,xacc)

    !PLOT=VS*(1-USTOP)
    WG=S*VS*(1-USTOP)+CO*VS  !MOLE U M/S
    KP=8.D4*WG  !MOLE U/M^2/S

    !1.5 MOLES OF H2 CONSUMED FOR EACH MOLE OF U,CONVERT TO CM^2
    KPH2=1.5D0*KP/1.D4    !MOLES OF H2/CM^2/SEC
    SIGKPH2=0.75D0*KPH2

    !MULTIPLY BY TIME AND SAEXP,FACTOR OF 20 TO MATCH INDUSTRIAL DATA
    H2CONS=TIME*SAEXP*KPH2/20.D0
    CALL STATPROD(SAEXP,TIME/10.D0,SIGSAEXP,KPH2,SIGKPH2,0.D0,H2CONS,SIGH2CONS)
    !1 MOLE OF H2 IS 2 GRAMS OR 2000MG
    !KPH2=KPH2*2000/1E4    !mG/M^2/S


END SUBROUTINE HYDRIDE


SUBROUTINE TIS(TEMP,CO,TI)

DOUBLE PRECISION TEMP,CO,TI,UC,K1

TI=-LOG(UC(TEMP))/(K1(TEMP)*CO)

END SUBROUTINE TIS


SUBROUTINE VSS(TEMP,TI,DE,S,VS)

DOUBLE PRECISION TEMP,TI,DE,S,K1,UE,VS

VS=(1/TI)*(DE/(S*K1(TEMP)*UE(TEMP)))**0.5

END SUBROUTINE VSS


SUBROUTINE FUNCTI(TEMP,PRES,U,FUNC)

DOUBLE PRECISION K1,K2,TEMP,U,FUNC,PRES,CO

CALL CZERO(TEMP,PRES,CO)

FUNC=-1.D0/(K1(TEMP)*CO*U+K2(TEMP)*(1.D0-U)**(1.D0/3.))
```

```
END SUBROUTINE FUNCTI


FUNCTION K1(TEMP)

DOUBLE PRECISION TEMP,K1
K1=10.4D0*EXP(1592.D0/TEMP)

END FUNCTION K1


FUNCTION K2(TEMP)

DOUBLE PRECISION TEMP,K2
K2=137.5D0*EXP(-4900.D0/TEMP)

END FUNCTION K2


SUBROUTINE CZERO(TEMP,PRES,CO)

DOUBLE PRECISION N,TEMP,PRES,CO,DENOM1,DENOM2,DENOM,NUM1,NUM

DENOM1=4.183D-6*PRES**0.5
DENOM2=DENOM1*EXP(-894.D0/TEMP)
DENOM=N(TEMP)+DENOM2

NUM1=DENOM2
NUM=N(TEMP)*NUM1

CO=NUM/DENOM

END SUBROUTINE CZERO


FUNCTION N(TEMP)

DOUBLE PRECISION N,TEMP

N=EXP(-2.362D0-2305.D0/TEMP)

END FUNCTION


FUNCTION D(TEMP)

DOUBLE PRECISION TEMP,D

D=1.9D-6*EXP(-5820.D0/TEMP)

END FUNCTION D


SUBROUTINE DEQ(TEMP,PRES,N,DE)

DOUBLE PRECISION TEMP,PRES,DE,N,D,CO

CALL CZERO(TEMP,PRES,CO)
DE=D(TEMP)*(N/(N-CO))**0.5
!PRINT*,'DE=',DE

END SUBROUTINE DEQ


FUNCTION UC(TEMP)

DOUBLE PRECISION TEMP,UC

UC=1.D0-(1.D0-0.989D0)**(0.5D0-0.5D0*TANH(-2950.D0*((1/TEMP)-0.00146D0)))

END FUNCTION UC
```

```
FUNCTION UE(TEMP)

DOUBLE PRECISION TEMP,UE,UC

UE=(UC(TEMP))**0.5

END FUNCTION UE


SUBROUTINE FUNCUSTOP(TEMP,CO,X,FUNCX)

DOUBLE PRECISION TEMP,CO,X,FUNCX,K2,K1

FUNCX=(K2(TEMP)*(1-X)**(1.D0/3.D0))-(K1(TEMP)*CO*X)

END SUBROUTINE FUNCUSTOP


 SUBROUTINE zbrac(TEMP,CO,x1,x2,succes)
      INTEGER NTRY
      DOUBLE PRECISION x1,x2,FACTOR,TEMP,CO
      PARAMETER (FACTOR=1.6,NTRY=50)
      INTEGER j
      DOUBLE PRECISION f1,f2
      LOGICAL succes
      if(x1.eq.x2)pause 'you have to guess an initial range in zbrac'
      CALL FUNCUSTOP(TEMP,CO,X1,F1)
      CALL FUNCUSTOP(TEMP,CO,X2,F2)
            succes=.true.
      do 11 j=1,NTRY
        if(f1*f2.lt.0.D0)return
        if(abs(f1).lt.abs(f2))then
          x1=x1+FACTOR*(x1-x2)
          CALL FUNCUSTOP.TEMP,CO,X1,F1)
        else
          x2=x2+FACTOR*(x2-x1)
          CALL FUNCUSTOP(TEMP,CO,X2,F2)
        endif
11    continue
      succes=.false.
      return
      END


        FUNCTION rtsec(TEMP,CO,x1,x2,xacc)
      INTEGER MAXIT
      DOUBLE PRECISION rtsec,x1,x2,xacc,TEMP,CO
      PARAMETER (MAXIT=30)
      INTEGER j
      DOUBLE PRECISION dx,f,fl,swap,xl
      CALL FUNCUSTOP(TEMP,CO,X1,FL)
      CALL FUNCUSTOP(TEMP,CO,X2,F)
       if(abs(fl).lt.abs(f))then
        rtsec=x1
        xl=x2
        swap=fl
        fl=f
        f=swap
      else
        xl=x1
        rtsec=x2
      endif
      do 11 j=1,MAXIT
        dx=(xl-rtsec)*f/(f-fl)
        xl=rtsec
        fl=f
        rtsec=rtsec+dx
        CALL FUNCUSTOP(TEMP,CO,RTSEC,F)
        if(abs(dx).lt.xacc.or.f.eq.0.)return

11    continue
      pause 'rtsec exceed maximum iterations'
      END FUNCTION RTSEC


      FUNCTION H2EQPP(TEMP)

      DOUBLE PRECISION TEMP,H2EQPP,PTORR
```

```
!CALCULATE PRESSURE IN TORR
PTORR=10**(-4590.D0/TEMP+9.39D0)

!CONVERT TO PA
H2EQPP=133.29D0*PTORR

END FUNCTION H2EQPP


SUBROUTINE H2MIN(TEMP,SIGTEMP,PH2MIN,SIGPH2MIN)

DOUBLE PRECISION TEMP,SIGTEMP,PTORR,EXPO,SIGEXPO,SIGPTORR,PH2MIN,SIGPH2MIN
DOUBLE PRECISION TERM1,SIGTERM1
!CALCULATE PRESSURE IN TORR
TERM1=-4590.D0/TEMP
CALL STATDIV (-4590.D0,1.D0,0.D0,TEMP,SIGTEMP,0.D0,TERM1,SIGTERM1)
EXPO=TERM1+9.39D0
CALL STATSUM(TERM1,1.D0,SIGTERM1,9.39D0,1.D0,0.D0,0.D0,EXPO,SIGEXPO)

PTORR=10**(EXPO)

CALL STATPOW10(EXPO,SIGEXPO,1.D0,1.D0,PTORR,SIGPTORR)

!CONVERT TO PA
PH2MIN=133.29D0*PTORR
SIGPH2MIN=133.29D0*SIGPTORR

END



SUBROUTINE PRESSURE(MOLES,SIGMOLES,TEMP,SIGTEMP,FREEVOL,PRES,SIGPRES)

DOUBLE PRECISION MOLES,SIGMOLES
DOUBLE PRECISION TEMP,SIGTEMP
DOUBLE PRECISION FREEVOL
DOUBLE PRECISION PRES,SIGPRES
DOUBLE PRECISION COEF

COEF=8.314D0/FREEVOL

IF (MOLES.GT.0.D0)THEN
PRES=COEF*MOLES*TEMP
CALL STATPROD(MOLES,COEF,SIGMOLES,TEMP,SIGTEMP,0.D0,PRES,SIGPRES)
ELSEIF(SIGMOLES .GT. 0.D0)THEN
PRES=0.D0
SIGPRES=COEF*SIGMOLES*TEMP
ELSE
PRES=0.D0
SIGPRES=0.D0
ENDIF

END SUBROUTINE PRESSURE


SUBROUTINE MOLECONV(PRES,SIGPRES,FREEVOL,TEMP,SIGTEMP,MOLES,SIGMOLES)

DOUBLE PRECISION MOLES,SIGMOLES
DOUBLE PRECISION TEMP,SIGTEMP
DOUBLE PRECISION FREEVOL
DOUBLE PRECISION PRES,SIGPRES
DOUBLE PRECISION COEF

 COEF=FREEVOL/8.314D0
MOLES=COEF*PRES/TEMP
CALL STATDIV(PRES,COEF,SIGPRES,TEMP,SIGTEMP,0.D0,MOLES,SIGMOLES)


END SUBROUTINE MOLECONV
```

HOURS	PH2	SIGPH2	PO2	SIGPO2	PTOT	SIGPTOT
1	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
2	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
3	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
4	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
5	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
6	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
7	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
8	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
9	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
10	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
11	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
12	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
13	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
14	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
15	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
16	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
17	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
18	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
19	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
20	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
21	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
22	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
23	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
24	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
25	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
26	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
27	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
28	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
29	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
30	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
31	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
32	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
33	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
34	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
35	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
36	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
37	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
38	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
39	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
40	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
41	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
42	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
43	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
44	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
45	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
46	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
47	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
48	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
49	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02
50	1.84E-02	5.00E-03	2.39E-03	2.42E-03	1.17E+05	5.31E+02