III. PHYSICAL ELECTRONICS AND SURFACE PHYSICS

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RESEARCH OBJECTIVES AND SUMMARY OF RESEARCH

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The principal objective of this work is to provide experimental and analytical information on the interaction of hydrogen with metals. Absorption, diffusion, and permeation of hydrogen in solids frequently lead to technological problems because hydrogen absorption degrades the mechanical, electrical, and magnetic properties of many materials, and hydrogen permeation results in unwanted losses or gains of gases in electron tubes, vacuum systems, and energy and power systems. Available experimental results indicate that, although the rate-controlling process generally is the diffusion of hydrogen in the bulk of the material, there are many instances in which the rate depends upon surface processes, such as adsorption, desorption, and penetration of the atoms into the lattice. In some cases, unintentional surface contamination causes the surface processes to have anomalously low rates; in other cases, surface coatings are intentionally used to reduce the rate of entry of atoms into materials.

Our work is designed to provide a more detailed understanding of these surface processes, thereby enabling engineers to make more accurate predictions of their influence on the rate at which hydrogen enters or leaves various materials. We have formulated a simple theoretical model of the rates of these processes, and we are now performing various experiments to test the model and to evaluate the unknown parameters of the model.

The first experiment is an investigation of the effects of surface contamination (e.g. , oxygen, sulfur, and carbon) on the rate of absorption, desorption, and permeation of hydrogen through various metals. At present, we are studying niobium, since this material is being used in an increasing number of technological applications, and it has an exceptionally high rate of diffusion and permeation. In the future, we plan to extend this investigation to other materials, especially to steels. The experimental apparatus is a unique combination of a variety of techniques, including the following.

1. Auger electron spectroscopy for determining the chemical composition of the surface of the material (i.e. , to identify the surface contamination).

2. Ion bombardment gun for cleaning the surface by sputtering.

3. Various sources for contaminating the surface in a controlled manner.

4. Rotatable detector for measuring the angular distribution of H_2 desorbed from the surface.

5. Mass spectrometry for determining the partial pressures of hydrogen and the contaminant gases.

We have used this apparatus recently to measure the rate of permeation of hydrogen through niobium for pressures from 1×10^{-4} to 10 Torr and for temperatures up to

~1000°C. The results exhibit deviations from the classical permeation equation, and we are now attempting to explain these deviations in terms of the surface processes. In the coming year we shall study the influence of surface contaminants on the permeation rate.

A second experimental study is designed to provide detailed information on the adsorption and desorption of hydrogen on copper, a material that is frequently used as a coating to reduce the rate of absorption and permeation of hydrogen in various metals. Existing data indicate that the adsorption probability of hydrogen on copper is extremely small. Furthermore, previous studies in our laboratory show that the angular distribution of H_2 desorbed from copper deviates markedly from the commonly assumed form of diffuse emission (i. e., the form cos θ, where θ is the angle measured from the surface normal); specifically, these data show that the angular distribution is strongly peaked in the vicinity of the surface normal. In the present experiment we are employing molecular-beam techniques to measure the adsorption probability as a function of the energy and angle of incidence of the impinging hydrogen molecules. The energy of the molecules is varied by changing the temperature of a tungsten nozzle which serves as the molecular-beam source. A rotatable mass spectrometer is used to measure the angular distributions of the molecules scattered and desorbed from the surface. The chemical composition of the surface is determined by Auger electron spectroscopy, and the surface is cleaned by both sputtering and oxidation. We have obtained results for single-crystal samples of copper having different crystallographic orientations (i. e., exposing different crystallographic planes). These results show that the adsorption probability increases with increasing energy of the impinging molecules and with decreasing angle of incidence, that is, closer to normal incidence. In addition, we observe that the adsorption probability depends on the crystallographic orientation of the surface. These results, together with the measured angular distributions of the desorbed molecules, can be described approximately by a simple model that assumes that there is an activation energy barrier associated with the dissociative adsorption $[H_2(g) \rightarrow 2H(ads)]$ of hydrogen on copper. The experimental results and the model satisfy the principle of detailed balancing. We plan to extend this investigation to other gases and metals.

In the third experiment we use tritium, the radioactive isotope of hydrogen, to obtain data on the concentration of hydrogen in the surface layers of metals, and to measure small rates of permeation of hydrogen through materials covered with permeation-resistant coatings (e. g., oxides). High sensitivity is attained by use of counting techniques. This experiment is designed to supplement the information provided by the preceding studies. We have designed and constructed most of the necessary components, and preliminary experiments will be performed.