CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES

TRIP REPORT

SUBJECT: Attendance at the MRS 2000 Spring Meeting
Charge Number 20.01402.571

DATE/PLACE: April 24–26, 2000, San Francisco, CA

AUTHORS: Osvaldo Pensado (CNWRA)

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PERSONS PRESENT: Osvaldo Pensado (CNWRA)

BACKGROUND AND PURPOSE OF TRIP:

The purpose of the trip was to attend to the symposium on *Corrosion of Metals and Alloys* and gather technical information that would be useful for the Center for Nuclear Waste Regulatory Analyses (CNWRA) technical assistance activities in the Nuclear Regulatory Commission (NRC) high-level waste (HLW) program. Staff also presented the paper *Modeling of the Passive Film on Nickel-Chromium-Molybdenum Alloys*, coauthored by Osvaldo Pensado, Darrell Dunn, Gustavo Cragnolino and Narasi Sridhar. Comments on this paper are discussed later.

SUMMARY OF PERTINENT POINTS:

The MRS 2000 Meeting included 34 technical symposia covering a wide range of Materials Science topics, poster sessions, an exhibit, five tutorials, and special presentations. Staff primarily attended the Symposium H on *Corrosion of Metals and Alloys.* Abstracts of the conference are available online at http://www.mrs.org/meetings/spring2000/abstracts. No proceedings volume is scheduled for Symposium H. A copy of the abstracts volume is available from the author. Comments on relevant papers are provided next.

Nancy Missert (paper H1.4) from Sandia National Labs discussed the use of thin film technology to study the preferential dissolution of Al in Al-Cu alloys. Oxide films of precise properties, ranging from conducting CuO to insulating Al_2O_3 were deposited on substrates of Cu or Al to study ionic transport properties under well controlled conditions. A similar technique could be used to study dissolution phenomena in Alloy 22.

Peter Bedrossian (paper H2.3) from the Lawrence Livermore National Lab discussed the use of Atomic Force Microscopy to study the dissolution of Alloy 22. Some reactions were proposed as mechanism for the alloy dissolution. Some electrochemical measurements, not at steady-state, were presented. No satisfactory answer was given when asked the link between his study and the evaluation of the long-term performance of Alloy 22. Outside of his talk Bedrossian mentioned that support from DOE to continue with his experimental program has been withdrawn. The main interest by DOE is to continue with the weight-loss measurement program to determine dissolution rates.

Henry S. White (paper H3.1) from the University of Utah discussed the use of scanning electrochemical microscopy (SECM) to determine electrochemically active sites on the surface of native oxides that can be correlated to the position of film breakdown. A microscopic tip is employed to scan the surface searching for electronic currents produced by redox reactions and tunneling. In some cases the active sites were shown to correspond to sites of film disruption. The diameter of the active sites ranges from 1 to 50 μm. The author claims that despite these large sizes, which may suggest the presence of particles on the oxide surface, no particles have been found by analytical techniques. The strength of the technique is that it seems to pinpoint candidate locations for film breakdown before its actual occurrence; however, the fact that only a minute surface can be studied makes it difficult to employ SECM as a sensor for detecting the onset of localized corrosion.

K. Sieradzki (paper H6.1) from the Arizona State University discussed a model based on percolation theory to explain the porosity patterns on Au-Ag alloys that appear after active de-alloying of silver. There was no consideration of the oxide film in this model. In a related topic, Jonah D. Erlebacher (paper H6.2) from the Harvard University presented a 3D model to explain the same porosity patterns in Au-Ag alloys. Simple rules that describe the motion of the atoms, considered as hard spheres, are employed to model the evolution of the system. From the number of atoms that *jump* into the solution, current densities are computed that are in fair agreement with experiments. Also in this paper, no consideration was given to the oxide film.

CNWRA staff, Osvaldo Pensado, presented a paper on the modeling of the dissolution of alloys similar to Alloy 22. According to this study, the utilization of two commonly used hypotheses to describe the dissolution of Alloy 22 may not be appropriate: congruent for stoichiometric dissolution and spatially uniform dissolution. The study indicates that preferential dissolution of nickel may occur. Peter Bedrossian mentioned that this seems to be consistent with the long term experiments at LLNL. Therefore, as dissolution proceeds, the composition of the alloy may change with time, having an effect on its mechanical properties. In the uniform dissolution hypothesis (used in the TPA code corrosion model) it is considered that the alloy thickness decreases uniformly in proportion to the amount of material released into the electrolyte. This hypothesis does not consider the formation of vacancies in the bulk of the alloy resulting from the ejection of atoms or ions into the electrolyte. The model, with its estimates of the physical parameters, suggests that abundant vacancies may be created as dissolution proceeds. These vacancies could coalesce along grain boundaries making the material porous, and without a significant change in the total thickness of the alloy wall. Thus, it may not be appropriate to consider total depletion of the alloy components as failure criterion, but just a fraction enough to guarantee that the electrolyte will penetrate via the porous channels or to cause fracture by residual stress. The fact that porosity formation is readily observed in other systems that display non-stoichiometric dissolution, such as those studied by Sieradzki and Erlebacher (mentioned above), suggest that porosity formation should not be disregarded as a feasible mechanism for failure of Alloy 22. It is possible that in the systems studied by Sieradzki and Erlebacher differ from Alloy 22 in that the oxide film may not play any role in the former and that active dissolution is needed to generate the porous structure; however, both systems should be similar in the solid-state transport. Furthermore, some old metal devices (from the Iron Age) are known to become porous oxides that can be made dust by just rubbing. Roger Newman presented one of such oxide samples in his talk, *Some Unsolved Problems in Corrosion Science* (paper H2.10).

SUMMARY OF ACTIVITIES:

Staff attended the Symposium H Corrosion of Metals and Alloys during the MRS 2000 spring meeting and presented a talk modeling of the passive film on nickel-chromium-molybdenum alloys.

CONCLUSIONS:

This is the first time that a corrosion symposium is organized within the MRS conference. Approximately forty researchers attended the symposium, most of them from universities and national laboratories. Participation by students was limited. Interaction with peers was facilitated by the structure of the symposium. It is estimated that more than a thousand people attended the overall meeting.

PROBLEMS ENCOUNTERED:

None.

PENDING ACTIONS:

None.

RECOMMENDATIONS:

Continued participation in the Corrosion of Metals and Alloys symposium at the MRS conference is recommended if organized again in the future. Further exploration of the *porosity formation* idea is also recommended since this mechanism could lead to shorter lifetimes of the waste package materials.

SIGNATURES:

Osvaldo Pensado
Research Engineer

5/15/00
Date

CONCURRENCE:

Narasi Sridhar, Manager
Corrosion Science & Process Engineering, Element

5/15/00
Date

for B. Sagar

Budhi Sagar
Technical Director

5/16/2000
Date

OP:jg