Zirconium Viscosity

Preliminary Recommendation

The preliminary recommendation for the viscosity of molten zirconium at the melting point is

$$8^{+7}_{-2}\ \text{mPa} \cdot \text{s}$$

This value was reported by Yelvutin et al. [1] from measurements using a graphite crucible. A large positive uncertainty is warranted because this value reported for zirconium differs significantly from the viscosity of Zircaloy-2 measured by Bunnell and Prater [2]. Iida and Guthrie [3] report small differences between viscosities of dilute alloys and pure metals (1-5%). Thus, the viscosity of Zircaloy-2 (98 wt% Zr, 1.2-1.7 wt% Sn, 0.18-0.38 wt% Fe+Cr+Ni) is expected to be similar to that of zirconium. The negative uncertainty is based on differences between the value reported by Yelvutin et al. [1] and viscosities reported in the literature for similar metals.

Discussion

The viscosity of Zircaloy-2 was measured by Bunnell and Prater [2] as a function of temperature from 2075 to 2175 K. They found that, in this temperature range, the viscosity of Zircaloy-2 is a constant equal to 15 mPa · s. Bunnel and Prater comment that the different viscosities obtained for zirconium and Zircaloy-2 may be due either to differences in viscosity of Zircaloy-2 and zirconium or to impurity effects introduced by the crucible used in the measurements. Yelvutin et al. used a graphite crucible whereas Bunnell and Prater used a less reactive thoria crucible. At high temperatures, zirconium reacts with graphite to form ZrC. No data are available on the post-test analysis of the solidified liquid from the viscosity measurements of Yelvutin et al. Thus, it is possible that their reported viscosity is that of a liquid mixture of ZrC + Zr or of zirconium with carbon in solution, not pure zirconium.

In order to rule out contamination of their sample from interaction with the thoria crucible, Bunnell and Prater [2] repeated their measurements after holding the sample at temperature for 2 hr. They obtained the same viscosities. Metallographic examination of the sample after these 2 hr experiments showed metallic thorium precipitates. X-ray fluorescence measurements indicated 2 mol% thorium in the Zircaloy. Bunnel and Prater also measured the viscosity of Zr-UO_2 mixtures containing 70 to 94.9 mol% zirconium. Analysis of the samples of these mixtures after the viscosity measurements showed that thorium contamination was less than 1 mol%. These measurements indicate that the mixture viscosity increases with increasing zirconium content from 10 mPa · s for 70 mol% Zr to 17 mPa · s for 94.9 mol% zirconium. These results are consistent with the viscosity measured for Zircaloy-2 and with other viscosity measurements of UO_2-Zr mixtures.

Although Bunnel and Prater report an abrupt change in viscosity when the sample became molten, it is possible that their measurements were made just as the sample began to flow. Liquidus temperatures of Zircaloy are a function of the amount of oxygen in the Zircaloy and range from 2136 to 2243 K for oxygen atom fractions of 0.007 to 0.19 [4]. The measurement by Yelvutin et al. was most likely made on a completely liquid sample since, unlike the alloy, the pure metal has a sharp melting point.

Based on the above data and considerations, it is clear that additional viscosity measurements are needed under well controlled atmospheres without contamination from containers. Until such data are available, a viscosity of 15 mPa · s is suggested for modeling the beginning of melting of Zircaloy, a Zr-Nb alloy, or zirconium with an oxide coating in an oxidizing atmosphere when the material begins to flow. However, for modeling the viscosity of zirconium in an inert or reducing atmosphere, a viscosity of 8 mPa · s is preferred.

References

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1. V. P. Yelvutin, et al., Chernage Met **7** (128) (1965); as referenced by Bunnell and Prater (Reference 2).

2. L. R. Bunnell, and J. T. Prater, *Viscosity of Zriconium-Uranium Oxide (Zr-UO_2) Mixtures at 1800 to 2100^{o}C*, U.S. Nuclear Regulatory Commission Report **NUREG/CR-4495** (September 1986); *Viscosity of Zirconium Uranium Oxide (Zr-UO_2) Mixtures at 2075 to 2375 K*, Pacific Northwest Laboratory Report **PNL-SA-15644** (February 1988).

3. T. Iida and R. I. L. Guthrie, *The Physical Properties of Liquid Metals*, Clarendon Press, Oxford (1993).

4. D. T. Hagrman, (ed.) SCADAP/RELAP5/MOD3.1 Code Manual MATPRO- A Library of Materials Properties for Light-Water-Reactor Accident Analysis, **NUREG/CR-6150, EGG-2720 Vol. 4** (June 1995), p 4-1 to 4-5.