# Vapor Hydration Testing of Nuclear Waste Glasses Using D$_2$O and H$_2$O

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Durability is one of the most important properties of glass to be used for the stabilization of nuclear waste. Because projected times of storage exceed practical durations of laboratory testing, several schemes of "accelerated" testing have been devised to aid in the selection of proposed formulations. Even with such tools, the test duration adequate to certify a glass for acceptance is difficult to establish with confidence because the corrosion rates measured on such tests are frequently non-linear and are subject to accelerations even after it appears that a stable corrosion rate has been achieved. A more complete understanding of the corrosion process is necessary to increase confidence in assessments of glass durability made using any means of accelerated testing.

One accelerated test in current use is the Vapor Hydration Test (VHT) in which a glass test coupon is suspended in a pressure bomb in contact with saturated water vapor at elevated temperature (usually 200°C) under conditions designed to minimize or prevent reflux.[1] The corrosion rate is determined by measuring the thickness of the modified layer formed on the coupon after 24 days. We have occasionally observed wide variations in the results of the test run on the same glass under identical conditions. This appears to be due to appearance of a new secondary phase in the modified layer, or a change in the growth rate of a phase already present. Two factors that control the corrosion rate are diffusion of water into the glass matrix and reaction rate of the glass with water. Substituting D$_2$O for H$_2$O provides a basis for distinguishing processes limited by diffusion which should scale as the square root of the mass ratio, $(18/20)^{0.5}$, from those limited by reaction rates. We have run comparative H$_2$O-D$_2$O VHT's of various durations on two typical high-sodium borosilicate glasses, NAE 4 and NAE 5. Preliminary results indicate that rate of reaction is the more significant factor in rate control because the corrosion rates measured in each glass in H$_2$O are usually at least twice as great as those measured in D$_2$O. Comparative plots of layer thickness for NAE 5 and NAE 4 are shown in Figure 1. NAE 4 is lower in sodium than NAE 5 accounting primarily for the lower levels of attack seen in NAE 4 at equivalent conditions. Selected typical layer cross sections are shown in Figures 2 and 3. It is particularly instructive to note the correspondence of variation in layer structure in the 8.5- through 10-day D$_2$O coupons of NAE 5 with the variations in average layer thickness. The 9.5-day coupon exhibits a much thinner layer than either the 8.5- or 9-day coupons, and also has much less structure with an almost complete absence exaggerated crystal growth. The large crystals are analcime. It is clear that growth of a high population of such large crystals is associated with an enhanced rate of corrosion, but causal relationships and mechanism are not immediately evident. Multiple repeats of these tests are now being run to help clarify these issues.

References

[1] W.L. Ebert and J.K. Bates, "The Reaction of Synthetic Nuclear Waste Glass in Steam and Hydrothermal Solution," Mat. Res. Soc. Symp. Proc. 176, (1990) 339-346.
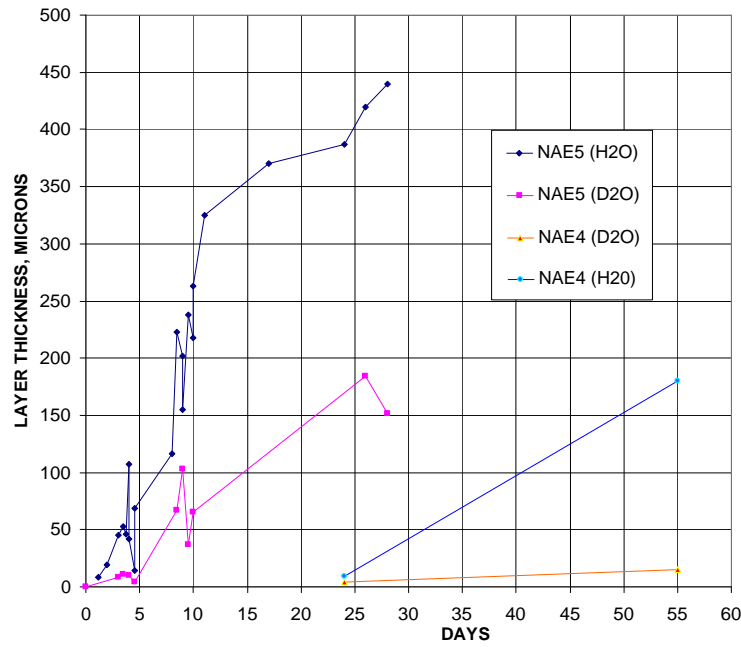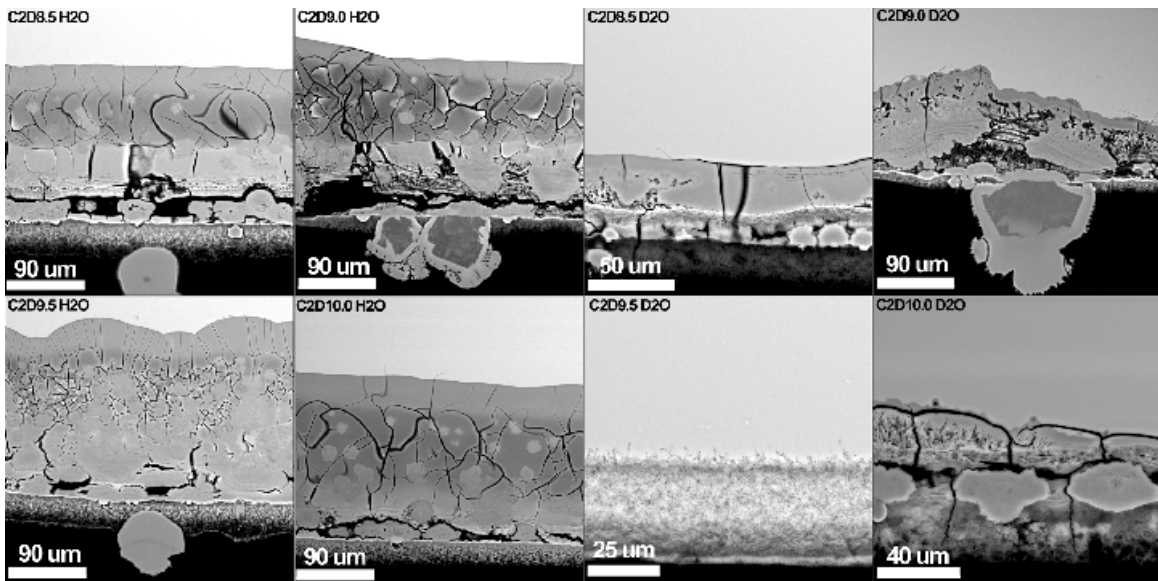
**FIG. 1. NAE4 and 5 corrosion data.**
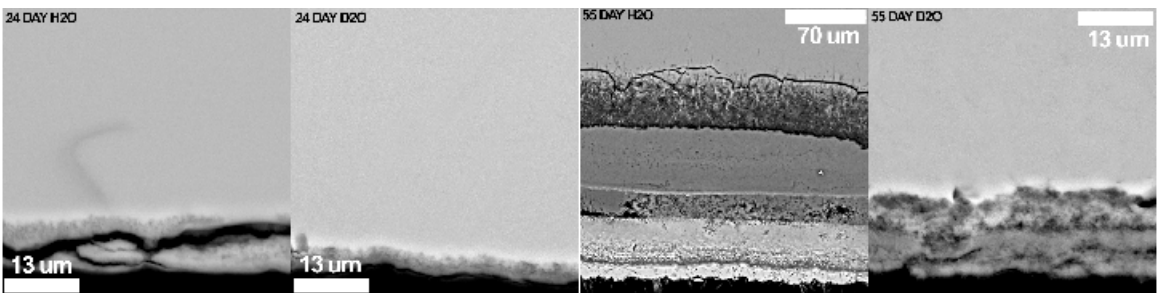


**FIG. 2. Typical layer cross-sections of NAE5.**



**FIG. 3. Typical layer cross-sections of NAE4.**