

Exploring the Influence of Variable Hydrogen Pressure on the Early Stages of the Plutonium–Hydrogen Reaction

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The early stages of the reaction between plutonium and hydrogen were investigated under both constant and variable pressure conditions to better understand factors relevant to the storage of plutonium metal. Interest in the formation of plutonium hydride has increased in recent years due to its relevance for ensuring the safe storage of plutonium metal [1–8]. Hydrogen can accumulate within sealed storage vessels during extended storage and conditions favourable for hydride formation may develop [9]. Hydride formation raises safety concerns due to the pyrophoric nature of plutonium hydride, which poses a risk of dispersing plutonium beyond appropriate containment [10]. The dependence of initiation time, nucleation rates, and radial growth velocities of plutonium hydride corrosion sites on hydrogen pressure have been established [1]. However, unlike controlled laboratory experiments, the pressure of hydrogen during storage would not remain constant and therefore, exploring the reaction under varied pressure regimes is of interest.

Using an *in situ* hot stage microscopy capability, the influence of hydrogen pressure on the initiation stages of the reaction between hydrogen and a plutonium alloy have been examined (Figure 1). A set of standard constant pressure experiments that measured initiation times, nucleation rates, and radial growth of plutonium hydride corrosion are compared to variable pressure experiments conducted using ramped pressure control or by varying the positioning of the gas inlet.

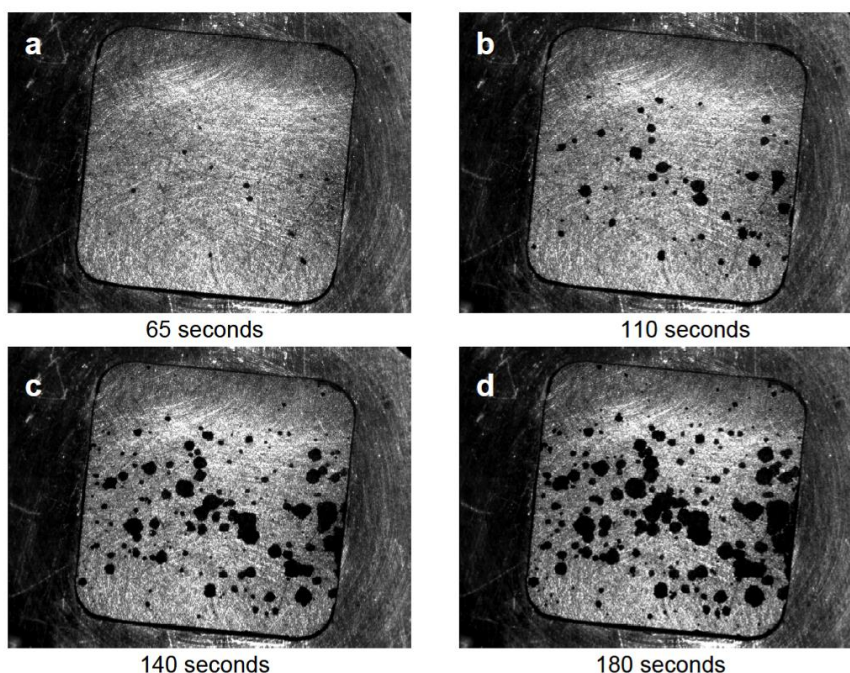


Figure 1 - Time resolved images illustrating the nucleation and growth behaviour of plutonium hydride reaction sites.

Variable hydrogen pressure conditions exert a measurable influence on the initial stages of the plutonium–hydrogen reaction. Although the kinetics observed under constant and ramped pressure conditions differ, they appear to correlate in terms of the hydrogen exposure ($\text{bar}\cdot\text{s}^{-1}$). The positioning of the gas inlet causes short-lived variations in local pressure which influence kinetics (Figure 2).

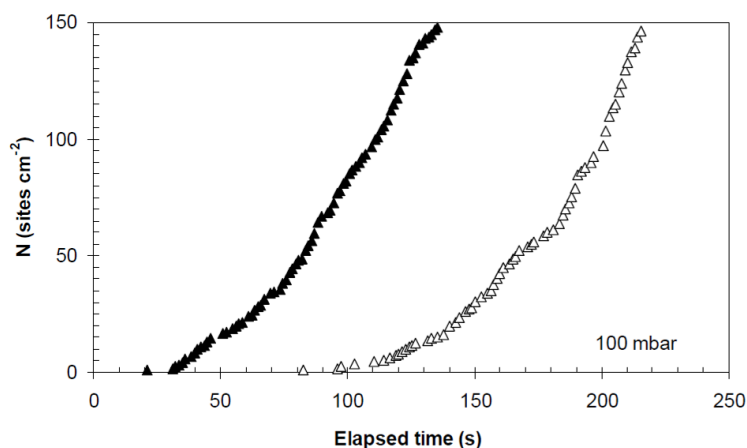


Figure 2 - Nucleation profiles for experiments conducted under 100 mbar of hydrogen. This study made use of two reaction vessels which were identical apart from the location of the hydrogen inlet. When the hydrogen inlet is directed at the sample the nucleation rates are greater.

This study has observed variations in hydrogen pressure, even over short timescales, influencing the initiation behaviour and early kinetics of the plutonium–hydrogen reaction. By comparing constant pressure experiments with those conducted under ramped and spatially varied pressure conditions, it is demonstrated that hydride formation is sensitive not only to the absolute pressure but also to the dynamic nature of hydrogen delivery. These findings highlight the importance of considering realistic, pressure regimes when assessing hydride formation.

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