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Effects of Radiation on DMSO Solutions with Polymeric Binders and HMX High Explosive[†]

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Abstract

A newly developed dismantlement process uses dimethyl sulfoxide (DMSO) solvent to remove plastic bonded HMX explosives (PBX-9404 and LX-10) from bonded nuclear assemblies. It is remotely possible in this process that DMSO solutions of the explosives will come into prolonged contact with plutonium. This paper addresses the anticipated chemistry and consequences of this contact.

Experimental

Model calculations were carried out over a range of possible exposure times to quantify the expected products of alpha-induced radiolytic decomposition. Only radiation via alpha-particle decay (2.37 MeV, 5.4 MeV max) was considered. The calculations were based on published radiolysis data for DMSO and the anticipated levels of radiation from ^{239}Pu (3.80×10^{13} eV/s/cm²). As some water is expected to be present in the solutions, owing to the hygroscopic nature of DMSO, the oxidation of the Pu by trace water also was calculated ($\text{Pu} + 2\text{H}_2\text{O} \rightarrow \text{PuO}_2 + 2\text{H}_2$). Calculations for ternary solutions of DMSO+HMX+H₂O could not be made due that lack of available data; however, it was noted that these solutions might exhibit more complicated behavior. For example, the presence of water might increase the radiolytic production of CH₄ via CH₃ radicals scavenging H or OH from H₂O highly ionized by alpha radiation. Also, the presence of NO₂ groups on HMX might lead to the formation of HNO₃ by hydrogen abstraction from H₂O.

In preliminary experimental studies, anhydrous DMSO solutions of PBX-9404 were prepared and to some precisely controlled amounts of water were added. These solutions were exposed to clean alpha-plutonium coupons for 21 days (504 hours) under an inert argon atmosphere in specially designed pressure-monitored, temperature-controlled, irradiation vessels. The exposures were carried out at 120°F. Radiolysis and oxidation reactions were tracked by measuring the increase in pressure over time. Termination gas analyses were performed. Changes in the weight and appearance of the Pu coupons were noted.

Preliminary Results and Conclusions

The calculated products of radiolytic decomposition of the DMSO were trivial compared to the quantity of reaction products calculated for the oxidation of plutonium by trace water. Preliminary results from laboratory experiments confirm these calculations. In the experiments, only those solutions containing water exhibited significant reaction (increase in pressure) over the 504 hour exposure period (see Figure 1). A significant pressure rise was not observed, however, until 100+ hours of exposure time had passed. The water containing solutions also showed high hydrogen (H₂) in the termination gas analysis, as well as elevated levels of nitrous oxide N₂O (indicating advanced decomposition of HMX and/or binder). Due to unanticipated condensation of DMSO vapor in the apparatus, the Pu coupons became partially exposed to the vessel atmosphere during the test and direct hydriding followed. The effects of water on Pu corrosion was pronounced: the Pu coupon exposed to the dry solution lost only ~0.2% of its starting mass, whereas those exposed to the wet solutions lost 3.2-4.2% of their mass.

[†] Work performed under the auspices of the U. S. Department of Energy by Lawrence Livermore National Laboratory under contract Number W-7405-ENG-48.

The preliminary conclusions are: 1) the amount of water in the DMSO should be maintained as low as reasonably achievable, and 2) a safety window of about 100 hours may be available to rectify a situation where HE/DMSO solution becomes exposed to plutonium.

Figure 1. Radiolysis vessel pressure vs. exposure time. a) anhydrous PBX-9404(25%)/DMSO solution, b) PBX-9404(25%)/DMSO solution + H₂O (2 wt%), c) Duplicate of b).

Figure 2. Termination gas analyses. a) anhydrous PBX-9404(25%)/DMSO solution, b) PBX-9404(25%)/DMSO solution + H₂O (2 wt%), c) Duplicate of b).

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