

FACTORS INFLUENCING LEACHABILITY IN A PELLETIZED WASTE FORM

MASTER

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Burnable wastes at Mound Facility which contain <10 nCi radioisotope/g waste are burned in the experimental cyclone incinerator. This is a volume reduction process resulting in a volume reduction of ~43:1. Radioactive ash and sludge (byproduct from the incinerator scrubbing system) are produced and these wastes are immobilized in a pelletized waste/cement matrix. Pellets of low porosity and high density are manufactured by pressing at 25,000 psi a mixture of calcined waste, water, and Portland cement. Sample pellets are tested for leachability, radiolytic gas generation, thermal stability, and compressive strength.

All Mound leachability testing has been performed using plutonium-238 oxide "spiked" high-level pellets. These pellets contain ~0.3 wt % PuO₂ and the data presented is preliminary in nature. The waste/cement weight ratios are 65%/35% for the ash/cement pellets and 50%/50% for the sludge solids/cement pellets. Leachability testing has been in progress for 15 months and the factors studied include temperature, leachant, test procedure, and pellet manufacturing pressure (density - porosity). Incremental leach rates have been calculated using the expression in Equation 1:

$$r_n = \left(\frac{a_n}{A_0} \right) \left(\frac{M}{S} \right) \left(\frac{1}{t_n} \right)$$

where:

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r_n = leach rate = (fraction leached) (g) (cm)⁻² (day)⁻¹,

a_n = grams of isotope lost during leaching period n,

A_0 = initial grams of isotope in leach specimen,

M = mass of specimen in grams,

S = exposed (geometrical) surface area of specimen in cm²,

t_n = duration of leachant renewal period in days.

If results are expressed graphically, the leach rate r_n should be plotted against $(t-t_n/2)$, the time represented by the middle of the leaching period.

In the experiments designed to determine the effects of temperature on the leach rate, two temperatures were studied, namely ambient (~22°C) and 70°C. Duplicate "spiked" sludge/cement and ash/cement pellets were leached in saturated brine solution using the Interim TRU Leach Procedure (Keith Johnstone, Sandia Labs). As can be seen in Figure 1, test results indicate that temperature has little effect on the leach rate of sludge/cement pellets (maximum difference one order of magnitude). However, the leach test results of ash/cement pellets proved to be inconclusive because of the wide scattering of data points for the 70°C tests.

Spiked ash/cement pellets were leached in two leachants (saturated brine and deionized water) to study the effect of leachant composition upon the leach rate. These tests were conducted at ambient temperature using the Interim TRU Leach Procedure. As can be seen in Figure 2, the leach rate of duplicate pellets in deionized water after four months of leaching was ~5 orders of magnitude greater.

Leachability tests were also conducted on ash/cement pellets using two

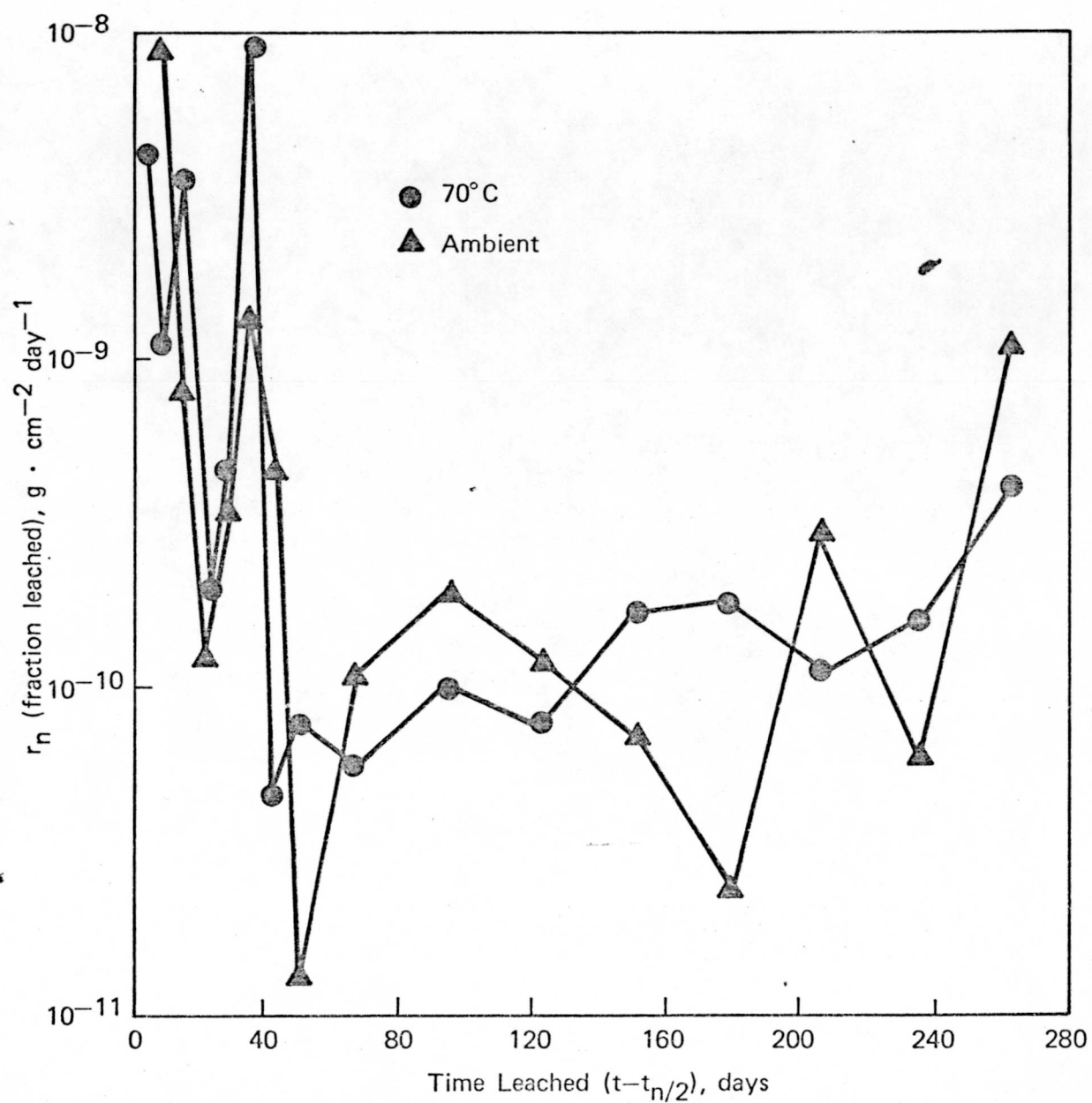


FIGURE 1 - Comparison of leach rates of sludge/cement pellets at ambient temperature and 70°C.

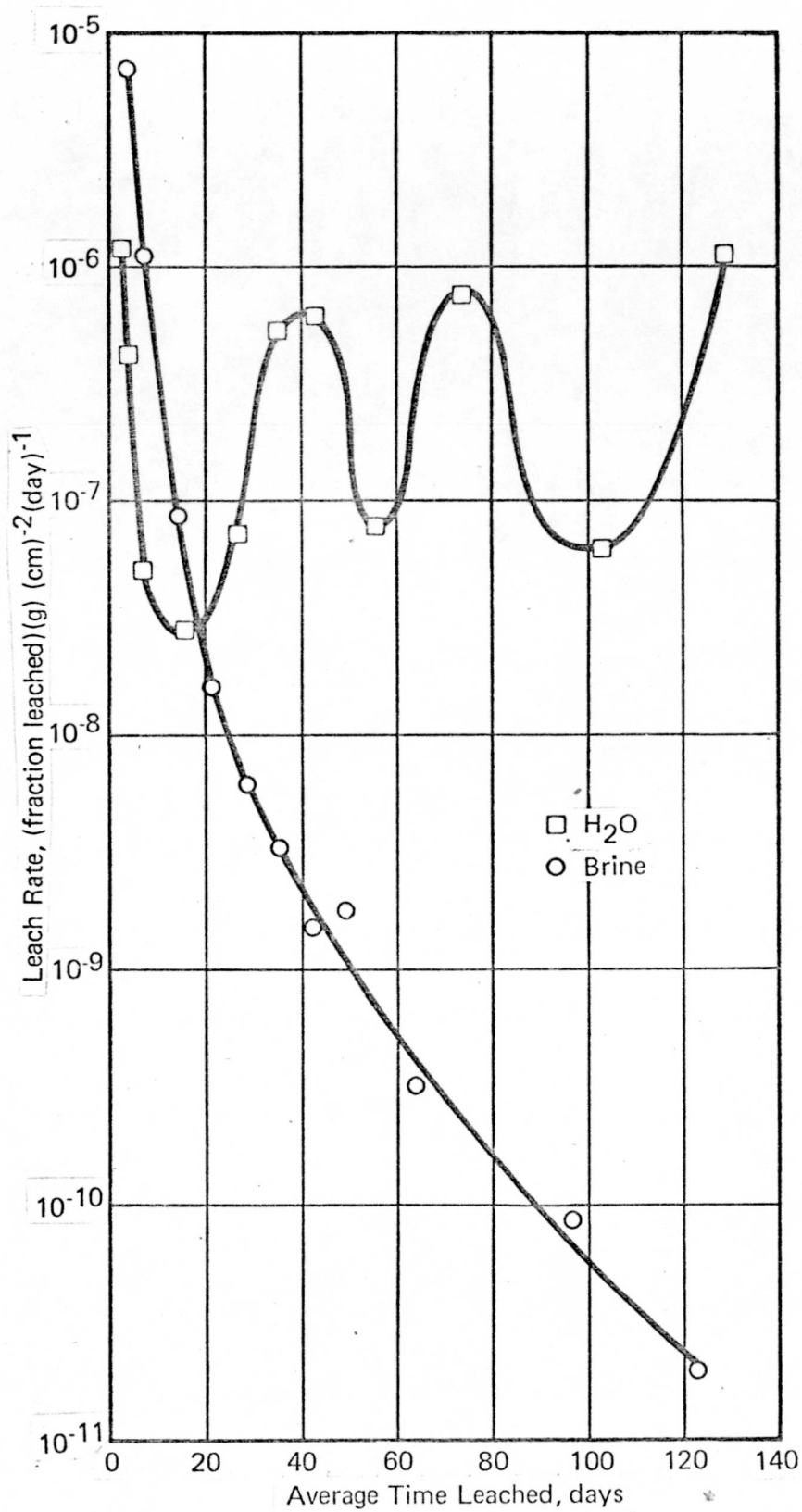


FIGURE 2 - Comparison of leach rates of ash/cement pellets in saturated brine solution and deionized water.

different leach procedures, the TRU and the ANS 16.1 (Herschel Godbee, ORNL). These experiments were done in deionized water at ambient temperature. Test results for the first 180 days of testing indicate that both procedures produce approximately identical results [leach rates in the 10^{-6} to 10^{-7} (fraction leached) (g) (cm) $^{-2}$ (day) $^{-1}$ range].

A designed leachability experiment has been prepared which may be used later to gather much more data. This experiment studies the effects of seven variables upon the leach rate. This test matrix is of a two-level saturated factorial design.