

TABLE I
RESONANCE DETECTORS

| Detector | Resonance energy (ev) | Foil material | Thickness (cm) | γ -activity counted | Corresponding theoretical group (ev) |
|-----------------------|-----------------------|-----------------------------|----------------|----------------------------|--------------------------------------|
| Indium | 1.46 | 99.8% Pb-0.2% In | 0.01 | 54 min In ¹¹⁶ | 1.1-3.0 |
| Gold | 4.9 | 100% Au | 0.0025 | 2.70 day Au ¹⁹⁸ | 3.0-8.3 |
| Uranium | 6.7 & up | 80% Al-20% U ²³⁸ | 0.005 | 2.33 day Np ²³⁹ | |
| Manganese subtraction | 337 | 89% Mn-11% Ni | 0.013 | 2.58 hr Mn ⁵⁶ | 170-450 |

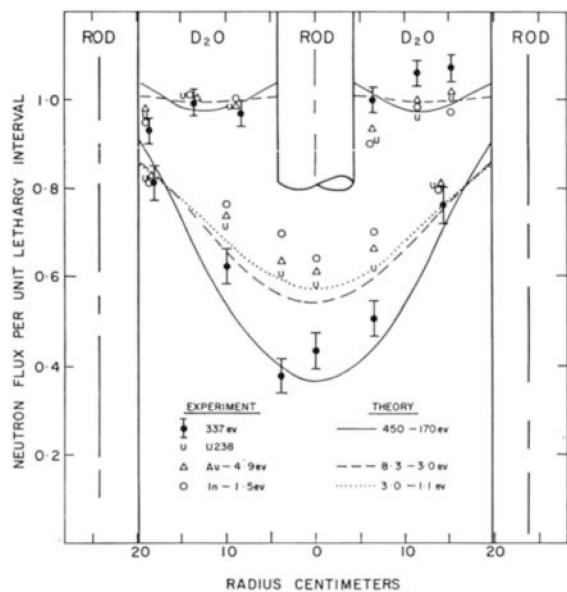


FIG. 1. Epithermal neutron distributions measured in the central cell of ZEEP at four different energies with (upper points) and without (lower points) the central rod in place. The curves are the predictions from age diffusion theory for energy groups corresponding to the experimental points. Where points are grouped together they were all obtained at the same radius.

near the rod because of resonance capture in the rod, and this was confirmed by Fig. 1 and other experiments. Since resonance capture was omitted from the theory the normalization in these cases was done at a point midway between rods.

The agreement between experiment and theory is considered satisfactory. The asymmetries in the experimental points with the rod in were probably caused by misalignment of the central rod and foil holders. The theory appears to overestimate the depression of the flux with the rod out in the region below 8.3 eV. This discrepancy is in the same direction and of similar magnitude to that expected to be caused by neglecting resonance capture in the calculation.

The results show that large distortions may occur in the slowing-down spectrum. Substantial errors can result from using empty lattice or reflector positions to measure resonance integrals in the region of a few hundred eV, the resonance integral being underestimated if the gold resonance integral is used as a standard. The same error occurs in the measurement of the Doppler effect in cadmium-covered

uranium since the fission sources are removed. The error in this case is large since the Doppler region extends up to several kev.

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Some Studies of Aqueous Uranium Oxide Slurries¹

Aqueous slurries of uranium compounds offer some promise as reactor fuels; the purpose of this letter is to point out that the preparation of such slurries in concentrations sufficiently great and of sufficient stability to be of interest as fuels offers an excellent illustration of colloid principles.

The sedimentation velocity of spheres, of radius r , density d in a solvent of viscosity η , density d_s under gravity, is given by the classical sedimentation velocity equation

$$V(\text{cm/sec}) = \frac{2}{9} r^2 \frac{(d - d_s)g}{\eta}$$

¹ Contribution No. 779. Work was performed in the Ames Laboratory of the U.S. Atomic Energy Commission during the period 1950-1952; this work was earlier summarized in two AEC reports, namely, ISC-145 (1951) and ISC-194 (1952), classified "Secret" at dates of issue. This area has been recently declassified.

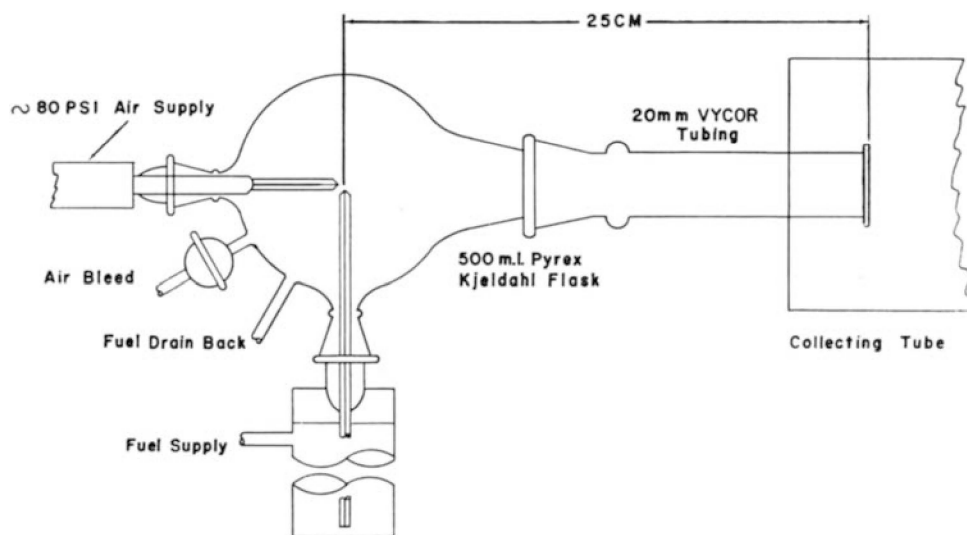


FIG. 1. Combustion apparatus: atomizer and burner

The density of U_3O_8 , for example, is 7.31 gm/cc. Thus, to obtain a sufficiently small sedimentation velocity to be readily compensated by random convection currents, about 10^{-6} cm/sec, (1) spheres of U_3O_8 dispersed in water should have a radius less than 27 μ at 25°C. It is therefore necessary to prepare primary particles with this radius or less, and to prevent their agglomeration after dispersion. The rate of agglomeration can be greatly reduced by charging the particle surface and maintaining an electrolyte concentration sufficiently low so that this charge is not neutralized (i.e., the double layer is not collapsed).

Uranium oxide identified by x-ray powder patterns (2) as principally U_2O_5 average particle diameter 60 μ (from BET nitrogen adsorption surface area and electron photomicrographs), was prepared by combustion of a spray of a solution of 125 gm $UO_2(NO_3)_2 \cdot 6H_2O$ per liter of acetone. The combustion apparatus is shown in Fig. 1; the air blast nozzle was a 2 mm inner diameter capillary drawn down at the tip to about 0.7 mm i.d., the fuel intake capillary was 0.5 mm i.d., and it is believed that other features in Fig. 1 are self-explanatory.

Thorium oxide powders of average particle diameter 38.5 μ were similarly prepared; in this case the spray solution was prepared by heating 100 gm $Th(NO_3)_4 \cdot 4H_2O$ with 150 ml ethanol until a clear solution resulted (about 1 hr), and diluting this solution with 1 liter of acetone.

The following slurries were prepared from the U_2O_5 powder obtained by spray combustion:

Slurry I.—200 gm U_2O_5 were ground into hot (70°C) 6N NaOH in a Charlotte colloid mill, Model ND1, the material being recirculated through the mill at constant temperature for 25 min. The solid settled immediately from the mother liquor and was washed with distilled water by centrifugation and decantation. The solid was then slurried with 1200 ml distilled water, the slurry purified by electro-dialysis, and finally again circulated through the colloid mill for $\frac{1}{2}$ hr at room temperature.

Slurry II.—50 gm U_2O_5 were suspended in 300 ml 0.5M Na_2HPO_4 , and the suspension was boiled for 1 hr. The suspension was allowed to settle, the supernatant liquid was decanted, the solid resuspended in 300 ml distilled

water, and the suspension electro-dialyzed. The resulting slurry was circulated through the colloid mill for $\frac{1}{2}$ hr at room temperature.

Slurry III.—30 gm U_2O_5 were ground with 30 gm $LiNO_3$ and 70 gm $NaNO_3$ using a mortar and pestle. This mixture was then fused for 3 hrs (about 550°C, gentle NO_2 evolution) which converted the U_2O_5 (gray-black) to UO_3 (yellow). The melt was poured into 600 ml distilled water, the UO_3 allowed to settle, and the supernatant liquid decanted. The UO_3 was washed twice with distilled water and was then suspended in 400 ml distilled water. The suspension was electro-dialyzed, circulated through the colloid mill for $\frac{1}{2}$ hr, and finally concentrated to 10% uranium by evaporation of water.

Methods for preparing slurries not depending on U_2O_5 fine particles as starting materials were also investigated, and the following preparations led to reasonably stable slurries:

Slurry IV.—11 gm $UO_2(NO_3)_2 \cdot 6H_2O$, 20 gm Na_2HPO_4 , and 30 gm $NaHCO_2$ (sodium formate) were ground together, and fused (350–400°C) until color changed from green to dark black (about $\frac{1}{2}$ hr.). The melt was cooled, slurried in 200 ml hot water, and the solid separated by centrifuging. The solid was suspended in 50 ml distilled water and the suspension electro-dialyzed.

Slurry V.—Preparation identical to that of slurry IV, except that 20 gm Na_2CO_3 replaced 20 gm Na_2HPO_4 in the initial fusion.

Slurry VI.—Preparation identical to that of slurry IV, except that 20 gm $NaNO_3$ replaced 20 gm Na_2HPO_4 in the initial fusion.

Slurry VII.—28 gm $LiNO_3$, 28 gm $NaNO_3$, 10 gm $NaCl$, and 10 gm Na_2HPO_4 were ground together and fused (250°C). 11 gm $UO_2(NO_3)_2 \cdot 6H_2O$ were stirred in, the temperature being maintained at 250°C until gentle evolution of NO_2 ceased. The melt was cooled, soluble salts extracted with water, the solid residue suspended in 50 ml of distilled water and the suspension electro-dialyzed.

Slurry properties are summarized in Table I.

Flocculation experiments on Slurries I and II indicated that the disperse phase was negatively charged, as might

TABLE I
 SLURRY PROPERTIES

| Slurry number | Solid phase properties | | | Suspension properties | | | | |
|---------------|------------------------------------|--|------|--|---|-------------|---------------------------------------|----------------|
| | Composition ^a | Surface area (m ² /gm ²) ^b | pH | Specific conductance (mho/cm 10 ⁴) | Viscosity × (millipoise, 25°C) ^c | Color | Stability ^d (25°C) (250°C) | |
| I | U ₂ O ₅ | 13.7 | 10.8 | 8.3 | 9.5 | Gray-black | B ⁺ | C |
| II | U ₂ O ₅ | 13.7 | 10.0 | 0.26 | 10.0 | Gray-black | B | B |
| III | UO ₃ | 16.0 | 10.0 | 9.7 | 10.4 | Yellow | B | D ^e |
| IV | UO ₂ | 27.4 | 8.6 | 10.5 | 12.7 | Blue-black | A | C ⁺ |
| V | UO ₂ + higher oxides | 34.9 | 9.4 | 12.0 | 10.9 | Brown-black | B | C |
| VI | UO ₄ ·4H ₂ O | 14.4 | 6.8 | 8.8 | — | Yellow | C | D ^e |
| VII | UO ₃ | 10.5 | 8.5 | — | >1000 | Yellow | C | D ^e |

^a Principal component, from x-ray powder diffraction pattern.

^b By nitrogen adsorption, BET technique.

^c Brookfield Synchro-lectric Viscometer.

^d Stability at 250°C was investigated by sealing 5 ml of slurry in 10-mm Pyrex pressure tubing, placing tube in a metal rack and heating at 250°C in a muffle for 5 hr. About 15% of such tubes exploded during heating. Slurry stabilities are rated on basis of appearance after 5 hr as follows: (A) No residue on tube bottom, no clear portion in liquid; (B) Slight residue easily dispersed, no clear portion in liquid; (C) Appreciable residue, dispersed by shaking; 5-mm clear supernatant; (D) Appreciable residue, dispersed by vigorous shaking; 5-mm clear supernatant liquid.

^e Of 3 samples, one formed a very viscous light yellow mass, presumably due to a hydration reaction of the solid.

be expected from their preparation in solutions of strongly adsorbable anions. Thus, when slurry II was made 0.001M in AlCl₃ or 0.002M in BaCl₂ (or higher concentrations) extensive settling had occurred within 16 hr, 80% of the solvent appearing to be almost clear. Concentrations of at least 0.02M Na₃PO₄, 0.01M Na₂SO₄, and 0.02M NaCl were required to produce similar results, presumably through collapse of the electrical double layer. Slurry pH could be adjusted within the range 6 < pH < 11 without appreciable effect on slurry stability, but rapid flocculation occurred at pH values below 5. Although a concentration of 0.001M AlCl₃ caused rapid flocculation, a concentration of 0.02M AlCl₃ stabilized the slurry, presumably with charge reversal.

ACKNOWLEDGMENTS

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