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2

The attached comments have arisen from recent informal discussions on the possibility of the application of the principles outlined by J. H. O. Varley¹² to interpretation of the radiation damage to thorium oxide particles. The Varley hypothesis is compared with results of previous irradiations of solid thorium oxide at ORNL, and for completeness, speculations are included concerning damage to the immediate environment of the particles (i.e., the double layer) as well as to the particles themselves.

The HRP Slurry Program Committee may wish to consider whether the suggested supplementary tests, not presently being carried out in the slurry irradiation program, might be included in subsequent work by the Chemical Technology Division or the Solids States Division.

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3

RADIATION DAMAGE TO AQUEOUS THORIUM OXIDE SLURRIES

I. SUMMARY

Radiation damage to aqueous thorium oxide slurries can be expected to occur through absorption of energy either in the electrical double layer surrounding the slurry particles or in the slurry particles themselves:

1. Radiation effects on the double layer may be either reversible or irreversible. Reversible changes could be dependent upon the presence of radiation and thus require in-pile measurement. Irreversible changes should persist out of pile at room temperature and be susceptible to measurement under these conditions.
2. Radiation damage to the particles may result in their degradation and thus affect the abrasion characteristics, the settling rate and the degree of flocculation.

The objectives of this memo are to examine those changes which could be caused by reactor radiation, to discuss their effects on the operation of a slurry system, and to propose several tests to evaluate the magnitude of these effects.

II. EFFECT OF RADIATION ON DOUBLE LAYER OF SLURRY PARTICLES

It may be convenient to review the characteristics of two types of slurry, flocculated and deflocculated. In a flocculated slurry the particles stick together in the form of loose irregular clusters (flocs), in which the original particles can still be recognized. A flocculated slurry is characterized by:²

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- a. zero or very small charge on the particle,
- b. a rapid settling rate due to aggregate formation,
- c. a large sedimentation volume because the particles preserve the haphazard positions in which they touched each other initially,
- d. a sedimented bed exhibiting a yield stress,
- e. a sedimented bed having a viscosity dependent on the rate of shear (because the rate of aggregation is so large that even in flow the aggregates do not completely disappear).

In a deflocculated slurry the particles repel each other and the original particles are not in contact. A deflocculated slurry is characterized by:²

- a. large charge on the particles,
- b. a slow settling rate due to the small size of the independent discrete particles,
- c. a small sedimentation volume (because the large charge creates a repulsive force allowing the particles to roll over one another to positions of minimum potential energy giving a densely packed bed, but one in which the particles still do not touch each other because of their large charge),
- d. a sedimented bed which does not exhibit a yield stress,
- e. a sedimented bed having a very high Newtonian viscosity so that even for large stresses the flow remains exceedingly slow,
- f. being more difficult to prepare than a flocculated slurry.

A. Reversible Radiation Effects
on the Double Layer

Reversible radiation effects on slurry properties depend on the continued presence of radiation, i.e., in the presence of radiation the double layer may collapse due to ionization, particularly the fission product

ionization. If the reversible effects of radiation were sufficiently large the slurry might actually change from a flocculated to a deflocculated condition upon entering or leaving the core, reverting to the opposite condition while in the remainder of the system. This would not be particularly serious if the system were designed to handle slurry of all degrees of flocculation. However, effects sufficiently large to change the degree of flocculation markedly could not be tolerated in a system designed to handle slurry of only one degree of flocculation.

B. Irreversible Radiation Effects
on the Double Layer

Irreversible radiation effects on the slurry properties via changes in the double layer will be associated with oxidation, reduction, and decomposition of those additives, corrosion products and fission products that form the double layer. It is believed that radiation cannot produce any effects more extreme than those which have already been observed out of pile with various concentrations of different additives, e.g., the settled bed concentration has been found to vary from 1500 to 2400 g Th/liter as the sulfate concentration was varied from ⁰~~2500~~ to 10,000 mg/kg ThO₂; ³ and the apparent viscosity at low rates of shear has been found to decrease from 40,000 to ~10 centipoise upon the addition of 20 millimoles oxalic acid per mole of ThO₂ to a slurry having a concentration of 730 g Th/kg H₂O. ⁴ If such is the case then two types of reactor systems can be considered: (1) a system which is capable of handling slurry of any degree of flocculation or (2) a system specifically designed to handle a slurry whose degree of flocculation is

controlled. In the first case any irreversible radiation effect should be of little consequence, since the system is designed to handle slurry of any degree of flocculation. In the second case, in which the degree of flocculation is controlled by an additive, the major difficulty is the selection of an additive whose decomposition rate is low enough that its continued addition will not poison the system or whose decomposition products are continuously removable. Thus, the problem in determining the importance of irreversible radiation damage to a slurry system need not be connected directly to slurry properties but rather to the rate of decomposition, oxidation, reduction, or removal of additives, fission products, and corrosion products and their effects on slurry properties. These effects can be determined on slurry systems after removal from the pile. It would not be necessary to conduct these tests at the maximum slurry concentration required for reactor operation for at least two reasons:

1. Thorium oxide may act as a catalyst in the decomposition, oxidation or reduction of additives fission products and corrosion products. (It has been shown in room temperature, out of pile studies that thorium oxide serves as a catalyst in the decomposition of sodium pyrophosphate to sodium phosphate.⁵) As a catalyst it is only the presence, not the concentration, of thorium oxide that is important.
2. The techniques of surface chemistry used to study the changes in the properties of the surface due to the changes in fission products, corrosion products and additives and their effect on the double layer should not be dependent on the concentration of slurry irradiated.

C. Proposed Tests to Determine Effect
of Radiation on the Double Layer

1. Test Conditions Common to a Study of Both
Reversible and Irreversible Effects

In a study of effects of radiation on the double layer, the stability of electrolyte additives must be determined for reactor conditions. In addition the oxidation state of corrosion and fission products must be determined under these same conditions. Suggested additives to be studied are thorium sulfate, sulfuric acid, sodium pyrophosphate and sodium silicate. The first two give acidic slurries which should be very stable against irreversible radiation effects since sulfate has been found to be very stable against radiation. Experience has shown that an oxygen atmosphere is desirable in systems in which aqueous thorium oxide is to be circulated under acidic conditions. The latter two additives give basic slurries and their stability under radiation is unknown. It is not clear whether an oxygen or a hydrogen atmosphere would be most suitable for these tests. It has been shown that corrosive attack of basic-aqueous systems is reduced in a hydrogen atmosphere,²⁵ however, tests with no indication of extraordinary attack¹⁰ have been made using aqueous thorium oxide slurries having a pH as high as 9.2 (at room temperature) in systems having an oxygen atmosphere. Both atmospheres should be used with basic slurries.

2. Test for Reversible Radiation Effects

Reversible radiation effects on the double layer will depend on the continued presence of radiation and will appear as changes in the degree of flocculation. The characteristics of the settled-bed are altered

greatly by changes in the degree of flocculation. The settled bed in a deflocculated system has a high bulk density and is an extremely viscous Newtonian material. The settled bed in a flocculated system has a relatively low bulk density and may be extremely non-Newtonian.

One measure of the settled bed characteristics is the force required to lift a rod around which slurry has been allowed to settle.^a Because of the different characteristics of Newtonian and non-Newtonian systems the force-time relationship for slurries having different degrees of flocculation should be determined. In Newtonian systems the time required to pull the rod from the settled bed should be inversely proportional to the lifting force. In a non-Newtonian system this would be true only up to the apparent yield point, for forces greater than the yield point the rise time would be very small. Once these relationships have been ascertained, such tests could be made in a modified dash-pot-stirred autoclave with a more powerful solenoid coil and would not require that the slurry be stirred during irradiation. Comparison with out of pile tests would show clearly whether there were any reversible radiation effects dependent on the presence of radiation; a series of such tests for different radiation times would also indicate any irreversible radiation effects. In addition, the out of pile tests at elevated temperatures could be related to room temperature dynamic resuspension tests by comparing the results of room-temperature force-time tests on settled

^a Tests of this nature were observed by H. F. McDuffie on a visit to the Westinghouse Electric Corporation, Pennsylvania Advanced Reactor Project, April 18, 1956. Variations in the nature of the slurry were said to be accompanied by large variations in the force required for withdrawal of the rod.

beds with transport velocity measurements²³ made on the same slurry.

It is important to use thorium oxide concentrations which are representative of the actual reactor systems.

3. Test for Irreversible Radiation Effects.

A test for irreversible radiation effects would be to allow a deflocculated slurry to settle into a very dense bed in a simple stainless steel autoclave, irradiation of the settled slurry with a high intensity γ -source could conceivably destroy the highly charged double layer, allowing the short range attractive forces to predominate. In the extreme case this would change the settled bed from a highly viscous Newtonian fluid to a hard cake having a brittle fracture, less extreme effects would require some measurement of the settled bed viscosity; a test similar to the one described above might prove satisfactory.

A second test for irreversible effects would be the determination of the stability of additives and the oxidative state of corrosion and fission products under reactor conditions. Since thorium oxide may play the role of a catalyst in the reactions being studied, it must be present in the bombs, however, as a catalyst it should not be necessary to have concentrations greater than 100 grams Th/kg H₂O. Stirring problems with slurries of this low concentration should be greatly reduced, compared to those now encountered. Conduction of the tests with thorium oxide containing 0-0.5% ^{highly} enriched uranium should distinguish any marked differences between gamma ray and fission product effects. Remote analysis of the supernatant fluid might be used to determine the amount of additive decomposed. Spectrophotometric, differential infrared absorption, or polarographic methods should be suitable.

III. EFFECT OF RADIATION ON SLURRY PARTICLES

A. Definition of the Thorium-Oxide Particle

Thorium oxide may be produced in many different ways, each one giving a different kind of particle. Methods that have been, or are being, used include: precipitation of the oxalate or formate, followed either by a carefully controlled calcination or by pyrohydrolysis, and spray burning of an acetone-water solution of thorium nitrate.

Precipitation of the oxalate, followed by a double or triple calcination, is the method currently being used for production of large quantities of oxide. This process yields a pseudomorphic particle having a mean diameter, on a weight basis, of 1-5 microns. This pseudomorphic particle is composed of crystallites from 150-1000 Angstrom on a side, their size depending primarily on the maximum calcination temperature.⁶ The crystallite size of any particular slurry remains unchanged after prolonged exposure to radiation or circulation in the loops, whereas the pseudomorphic particles are markedly degraded in size by circulation in the loops.⁶

Precipitation of the formate, followed by calcination, yields a particle having a mean diameter from 1-5 microns, on a weight basis, which was reported⁷ to have no pseudomorphic structure but which has an X-ray crystallite size of from 200-1000 Å. Pumping in the loops substantially disintegrated those particles above 1 micron⁸ which suggests that some pseudomorphic structure may have been present in the original calcined material.

Pyrohydrolysis of either thorium oxalate or thorium formate produces particles having a mean diameter, on a weight basis, of from 0.5-2 microns

and an X-ray crystallite size of from 150-300 Å. Circulation in the loops gives very little particle degradation for particles less than 1 micron in diameter.⁶

No treatment of thorium oxide particles has been reported to produce an increase in X-ray crystallite sizes once the thorium oxide has received its final calcination.²⁴

Flame burning of acetone-water solution of thorium nitrate produced an oxide whose particles appear spherical upon examination with an electron microscope. These particles were extremely fine and gave a stable suspension in water.⁹ No information is available on the amount of degradation produced during circulation in a loop.

In order to minimize erosive attack on system components, the thorium oxide particles should be 95% less than 0.5 micron on a weight basis.¹⁰ Other reasons for desiring particles of this size include:

1. Mechanical agitation tends to reduce the mean particle diameter on a weight basis to the order of 0.3-0.5 micron; and in doing so, damages the system so that it is less effective for measurement. Introduction of particles of this size into the system would minimize changes in properties accompanying and due to the particle size degradation.
2. The effect of additives would be more predictable since internal surface would be minimized, thus minimizing the quantity of electrolyte required to produce a given degree of flocculation (because only the electrolyte on external surfaces can contribute to double layer forces due to their short range nature).
3. The fission product escape probability is unity for particles less than 5 microns (see Appendix A), therefore all fission products can be expected to be adsorbed on the particle or be driven into the interior from the exterior surface. It does not seem unreasonable to anticipate a chemical processing scheme in which the fission products can be removed without dissolving the thorium oxide particles, this process being considerably easier if it is necessary to deal only with external surface, or pits accessible from the surface.

B. Mechanism of Radiation Damage to Thorium
Oxide Particles

It has been suggested¹¹ that the interpretation of radiation-induced disintegration of a uranium alloy matrix proposed by J. H. O. Varley¹² might be applied to the estimation of the effects of radiation on the thorium oxide particles of a slurry blanket. Varley proposed a relationship between the mechanical properties and the fraction of interstitial atoms to cause fracture or disintegration.^b The results of the calculation of the fraction of interstitial atoms at fracture for a series of values of the ultimate tensile strength and breaking strain, are presented in Table I. The minimum and maximum atomic percentages of interstitials at rupture are 0.12 and 3.0 respectively, for the range of strengths covered.

TABLE I
ATOMIC PERCENTAGE OF INTERSTITIALS BEFORE FRACTURE

Breaking Strain	Ultimate Tensile Strength, p.s.i.					
	2.5×10^4	5×10^4	10^5	2×10^5	4×10^5	8×10^5
10^{-2}	0.53	0.75	1.1	1.5	2.1	3.0
5×10^{-3}	0.37	0.53	0.75	1.1	1.5	2.1
10^{-3}	0.17	0.24	0.33	0.47	0.67	0.94
5×10^{-4}	0.12	0.17	0.24	0.33	0.47	0.67

^b A detailed summary of Varley's hypothesis has been omitted to avoid classification.

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If it is assumed that the excitation energy due to neutron capture in thorium is dissipated by the emission of γ -rays¹⁶ and that the recoil energy is sufficient to rupture chemical bonds and move the recoil nucleus to an interstitial position but not to allow the recoil nucleus to escape from the particle, an atomic percentage of interstitials of 0.12 and 3.0 correspond to 1.7×10^{20} and 4.3×10^{21} nvt, respectively. The assumption that the recoils do not escape from the particle is supported by an experimental irradiation of a U_3O_8 slurry.²¹ A dilute suspension (< 1 gram/liter) of U_3O_8 in gelatin was irradiated for about one minute in the central portion of the Brookhaven reactor. The U_3O_8 particles were less than 1 micron in diameter whereas the minimum fission product range in U_3O_8 was calculated to be 8 microns. As predicted (see Appendix A) more than 99% of the fission product activity was found in the aqueous phase, (after the gelatin was liquified by heating), while the U^{239} and Np^{239} (recoil particles) remained in the uranium oxide particles.

Detailed calculation of the required nvt is difficult as shown by the experiments of Harrison and Seitz¹⁷ in which the observed damage was 1/5 that calculated using simple displacement theory. One explanation of this discrepancy has been advanced by Sampson, Hurwitz and Clancy¹⁸ who made a study of the effect of energy of the colliding atom on the displacement probability. Their results indicated a reduction in the number of displacements over that calculated from the simple theory.

The disruptive action of high energy fission fragments produced by the fission of accumulated uranium would mean that a smaller value of the nvt would be required to break the equivalent number of bonds. For a steady state

concentration of 3 grams U^{233} /liter and a thorium concentration of 1000 grams per liter, the total nvt might be 1/5 to 1/10 the values given above to produce the same damage estimated for the recoil interstitials.

An irradiation of thorium oxide pellets of unknown physical properties, to an nvt of 10^{19} in the Graphite Reactor led to inconclusive results although six of the sixty-seven specimens had shattered.¹⁹ Two sintered thorium oxide rods were irradiated to an nvt of $1.7-2.4 \times 10^{20}$, with no dimensional changes,²⁰ however one rod was broken either due to radiation effects, thermal stresses or when clamped in a vise. The remaining rod was broken in a modulus of rupture test, giving a value of 2000 psi compared to an expected value of 5000 psi.

C. Proposed Tests to Determine Effect of Radiation on Thorium Oxide Particles

In order to test the applicability of Varley's hypothesis on radiation damage to brittle non-metallic materials, samples of dry thorium oxide should be irradiated for nvt up to 10^{22} , preferably in as high flux as possible in order to minimize the effect of U^{233} buildup and consequent fission product damage. The thorium oxide should have crystallites of very closely controlled size and crystallite sizes of 200, 400, 800 and 1600 Å should be irradiated. The ThO_2 should be irradiated in the MTR for periods of 10, 30 and 60 days. X-ray crystallite measurements before and after irradiation should detect any appreciable amount of damage. Radiation damage due to fission fragment emission should be evaluated with coprecipitated thorium and uranium oxides, or by irradiating pure thorium oxide in the MTR, measuring the X-ray crystallite size before and after irradiation, then after allowing all the Pa to decay,

inserting the sample again for a brief period to fission the U^{233} present and again measuring crystallite size. It would be desirable for the crystallite size to be the same size as the individual particles, such particles might be produced by pyrohydrolysis of either thorium formate or thorium oxalate or by the flame-spray method previously mentioned.⁹ In addition, a "standard" thorium oxide should be irradiated to provide a comparison with present results.

If X-ray crystallite sizes are smaller after irradiation then tests must be made to determine the rate of degradation as a function of the mechanical work done on the slurry. During the degradation measurements the erosiveness as well as the degree of flocculation should also be determined. Comparison should be made with unirradiated material.

IV. SUMMARY OF TESTS

A. Effect of Radiation on Double Layer of Slurry Particles

1. Test for Reversible Effects

The test consists of the measurement of settled bed characteristics in the presence and absence of radiation by the amount of force required to lift a rod around which slurry has been allowed to settle. In Newtonian systems the time required to pull the rod from the settled bed should be inversely proportional to the lifting force. In a non-Newtonian system this would be true only up to the apparent yield point, for forces greater than the yield point the rise time would be extremely short.

2. Tests for Irreversible Radiation Effects

- a. The test consists of allowing a deflocculated slurry to settle into a dense bed and then irradiating the material with a high intensity γ -source. This could conceivably destroy the highly charged double layer, allowing short range attractive forces to predominate and in the extreme case changing the settled bed from a highly viscous Newtonian fluid to a hard cake having a brittle fracture.
- b. A second test would be the determination of the stability of additives and the oxidation state of corrosion and fission products under reactor conditions. Remote analysis of the supernatant fluid might be used to determine the amount of radiation damage. Spectrophotometric, differential infra-red adsorption, or polarographic methods should be suitable. Techniques developed in surface chemistry studies might be adopted to changes in the nature of adsorbed additives and fission and corrosion products.

B. Effect of Radiation on Thorium Oxide Particles

Dry thorium oxide having crystallite sizes of 200, 400, 800 and 1600 Å should be irradiated, in the presence and absence of U^{235} , for an nvt of 10^{22} . X-ray crystallite measurements before and after irradiation should detect any appreciable amount of damage.

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V. APPENDIX A

FISSION PRODUCT EMISSION FROM A
SMALL SPHERICAL PARTICLE²²

In order to determine the distribution of fission products, the following model was assumed for fission product emission from a small spherical particle:

where

R = radius of particle in which fissions are occurring

P = point of fission occurrence

L = average range of fission fragment

A = angle at which fission fragment is emitted

r = radius of point P.

The probability that a fragment emitted isotropically from point P has the angle $A = 1/2 dA$ is

$$\frac{d(\cos A)}{\cos A} \Big|_0^\pi$$

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The fraction of the fragments emitted from P that travel beyond the surface $4\pi R^2$ is

$$-\int_0^{A_{\max}} \frac{1}{2} d(\cos A) = \frac{1}{2} \left[1 - \frac{R^2 - r^2 - L^2}{2Lr} \right]$$

where

$A_{\max}$ = maximum angle of fission fragment emission that will still allow the fragment to escape from the particle.

$$\cos A_{\max} = \frac{R^2 - r^2 - L^2}{2Lr}$$

The fraction of the fragments emitted from uniform sources throughout the volume $4\pi R^3/3$ is

$$(3/4\pi R^3) \int_{R-L}^R \frac{1}{2} \left[1 - \frac{R^2 - r^2 - L^2}{2Lr} \right] 4\pi r^2 dr$$

which equals

$$\frac{3}{4} \cdot \frac{L}{R} - \frac{1}{16} \cdot \frac{L^3}{R^3} \text{ when } L \leq 2R \text{ or } 1 \text{ when } L \geq 2R$$

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