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Second Quarterly Report, 1956

on

I. PREPARATION OF THORIA SOLS

II. MEASUREMENT OF PHYSICAL PROPERTIES OF SOLS AND SLURRIES

Subcontract 904 under W-7405 eng-26

UNCLASSIFIED

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SUMMARY

This report describes the progress made in the study of thoria sols and related systems and in the development of methods for measuring the physical properties of sols and slurries as required by Subcontract 904 under W-7405 eng-26.

The preparation of thoria sols, by an improved dialysis process, has continued. In the improved process the sols are concentrated to a glass and redispersed in water, repeatedly, at intermediate stages of the dialysis. This results in condensation of the thoria micels and release of nitrate ion which is then readily removed from the system. A total of 19 sol samples containing nine different inorganic stabilizers have been prepared and tested for stability at high temperatures. All of the sols were stable for long periods at 100°C; all decomposed to slurry in sixteen hours or less at 300°C in unstirred bombs as shown by examination at temperature by x-radiography. The presence of nitrate ion may have masked the effect of the additives.

Electrophoresis experiments have shown that a thoria sol, containing less than 1% nitrate on the thoria basis, consists of positively charged micels. X-ray diffraction crystallite size measurements indicate that crystallite growth is proportional to the nitrate to thoria ratio in sol-slurry systems.

Thorium phosphate gel beads, which may be useful in a non-coking "slurry" system, have been made with an average diameter of about 2 mm. A study of the hydrothermal stability and physical properties of these beads is in progress.

Electrophoresis apparatus for studying the electrical properties of micels and particles at elevated temperatures is being designed. Arrangements have been made with Sperry Products, Inc. for laboratory tests of ultrasonic apparatus for measuring slurry settling rates.

INTRODUCTION

This report describes the work done in the second quarter of 1956 on the preparation of thorium sols stable at elevated temperature and on the development of instrumentation for studying the physical properties of sols and slurries of thorium oxide in water.

The preparation of sols by dialysis has continued and high temperature stability tests are in progress. X-ray examination of the stability test bombs has been established on a routine basis.

Correlation of sol stability with the physical and chemical properties of the thorium-water system is being studied. Fruitful results have already been obtained from x-ray diffraction crystallite size measurements, small angle scattering of x-rays and electrophoresis measurements. The ultrasonic method for measuring slurry settling rates will be investigated with Sperry Products, Inc., manufacturers of pulse-type ultrasonic fault locators.

Several methods other than dialysis have been tried for sol preparation. Further work in this field is contemplated, particularly the use of ultra-filtration for separating colloidal particles and ions.

The preparation of thorium and thorium phosphate gels, both to be peptized and for use in the form of beads, in non-coking "slurry" systems, is in progress.

DISCUSSION

Preparation and Stability of Thoria Sols

The details of preparation of thoria sols by dialysis are given in Table I. Many of these sols were prepared with added ions which were expected to increase the amount of charge carried by the thoria micel. Nitrate removal, however, has not been very complete and the effects of the added ions cannot be separately evaluated.

During dialysis osmotic transfer of water to the sols occurred and it was necessary to periodically concentrate them to increase the rate of nitrate ion removal. In the early preparations, care was taken to avoid concentrating the sols much below the initial volume charged to the dialysis cell. It was later found (Table II) that concentrating the sol until a glass was formed, then redispersing this glass in water, gave a measurable increase in the rate of removal of nitrate ion. It is believed that condensation of colloidal micels took place with consequent release of nitrate ion to the solution.

High temperature stability tests have been made on most of the sols listed in Table I. None of the sols were stable for as long as sixteen hours. Some of the tests failed owing to evaporation of water from imperfectly sealed bombs. The separated solids from some of the test bombs have been examined by x-ray diffraction. The results, given in Table III, suggest that the rate of recrystallization of thoria in systems containing nitrate ion is an inverse function of the thoria to nitrate ratio, the greater the ratio, the slower the rate of recrystallization. Other ions which form thorium salts more soluble than the hydroxide would be expected to act similarly. Further studies of this will be made.

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TABLE II

Removal of Nitrate Ion by Dialysis

All sols prepared by mixing additive with 150 ml. 1 M $\text{Th}(\text{NO}_3)_3$ soln. and adjusting pH to ~ 3.6 before dialysis in small continuous dialyzer.

<u>Sample No.</u>	<u>Description</u>	<u>Days of Dialysis</u>	<u>$\text{ThO}_2/\text{NO}_3^-$ Wt. Ratio</u>
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These sols were concentrated to approx. original volume each three to five days.

1139-SR-2	2 Mol % Cr_2O_3 as $\text{Cr}(\text{NO}_3)_3$	43	4.5
1142-SR	1 Mol % Fe_2O_3 as $\text{Fe}(\text{NO}_3)_3$	28	4.0
1144-SR	1 Mol % CrO_4^{2-} as CrO_3	31	3.3

Concentrated to glass after each three to five days of dialysis.

1170-SR	4 Mol % PbO as $\text{Pb}(\text{NO}_3)_2$	34	6.3
1171-SR	4 Mol % Y_2O_3 as $\text{Y}(\text{NO}_3)_3$	29	5.9
1167-SR	4 Mol % La_2O_3 as $\text{La}(\text{NO}_3)_3$	36	13.7
1184-SR	None, continuation of 1138-SR*	33	16.0
1169-SR	4 Mol % Fe_2O_3 as $\text{Fe}(\text{NO}_3)_3$	38	17.6
1187-SR	None, Large dialyzer used. (2 liters charge)	11 35	4.0 14.

*Large scale preparation in large continuous dialyzer. Time in large dialyzer to give 4.5/1 $\text{ThO}_2/\text{NO}_3^-$ included above.

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TABLE IIIX-Ray Crystallite Sizes of Thoria Sols and Slurries

<u>Expt. No.</u>	<u>Description</u>	<u>ThO₂/NO₃⁻ Wt. Ratio</u>	<u>Crystallite Size A</u>
1138-SR	Sol prepared by dialysis without concentration to glassy state.	4.6	Not observable
1184-SR-4	Sol prepared from 1138-SR by further dialysis including conc. to glass at intermediate stages.	16	30
1206-SR-26	Dry residue ex attempted 1 week hydrothermal treat of 1184-SR-4. At least 16 hours in water at 300°C (by x-ray of bomb).	16	70
1212-SR-11	Slurry ex hydrothermal treat 1197-SR-4, sol with slight amount of suspended solid prepared from "pyrolyzed" Th(NO ₃) ₄ (See Table IV)	21	80
1212-SR-17	Slurry ex hydrothermal treat of 1209-SR-4, sol from "pyrolysis" of Th(NO ₃) ₄ (see Table IV)	10.1	180

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TABLE IVProducts of Stepwise Pyrolysis of Thorium Nitrate at Reduced Pressure

<u>Expt. No.</u>	<u>Max. Temp. Hot Plate °F</u>	<u>Hours Heating</u>	<u>ThO₂/NO₃⁻ Wt. Ratio</u>	<u>pH of "dispersion"</u>	<u>Degree of Dispersion</u>
1197-SR-1*	550	1	1.8	-	Incomplete
-2	550	1	6.4	-	"
-3	600	1-1/2	10.6	-	Complete
-4	~ 650	2-1/2	21	3.6	Incomplete
1209-SR-1	550	1	2.0	1.9	Incomplete
-2	550	2	4.1	2.5	"
-3	550	4	6.9	2.4	"
-4	550	15	10.1	2.2	Complete
-5	650	2	13.6	2.6	"

Physical Properties of Thoria Sols

The measurement of thorium oxide crystallite size by x-ray diffraction line broadening has been combined with nitrogen surface area and electron micrographs to characterize the calcined thorium oxide used for slurry experiments. Characterization of the thoria sols prepared by dialysis is in progress using x-ray diffraction and small angle scattering to obtain the relationship between crystallite and micel size and investigate changes in these quantities with treatment and composition.

Two thoria sols prepared by dialysis have been examined by x-ray diffraction using a liquid sample holder with a Mylar film window. The window diffraction spectrum pattern interferes with the thorium oxide spectrum between 3.5 and 2.5 Å spacing but has no effect on the 1.98 Å and 1.67 Å diffraction lines. The sample description and x-ray diffraction results are listed below.

	Wt. Ratio $\text{ThO}_2/\text{NO}_3^-$	Crystallite Size from Broadening	
		1.98 Å line	1.67 Å line
1138-SR - Dialysis of $\text{Th}(\text{NO}_3)_4$ solution adjusted to pH = 3.6, giving clear sol.	4.6	no line	no line
1184-SR-4 - Sol 1138-SR taken to glass stage 3 times, during dialysis. Final sol slightly milky.	16	25 Å	30 Å

The small angle scattering measurements have not yet been made. Further work on thoria sols of different thoria to nitrate ion ratios and with other ions incorporated in the thoria lattice is planned.

The stability of colloidal systems is generally related to the sign and magnitude of the charge on individual particles or micels. Electrophoretic movement of the colloidal particles toward an electrode gives a qualitative indication of the sign of the charge residing on them. A simple U-tube electrophoresis cell with a layer of sol overlaid in with distilled water in each limb was provided with platinum electrodes and connected to a variable voltage DC supply. Two thorium sols were examined (at a potential gradient of 1 volt/cm) with the results set forth below.

<u>Sol</u>	<u>Product at Electrode of Sign</u>	
1110-SR - Thorium without additive $\text{ThO}_2/\text{NO}_3^- = 27$ by wt.	nitrate ion	$\text{ThO}_2 \cdot x \text{H}_2\text{O}$ gel
1144-SR - Thorium with 1 mole % CrO_4^{2-} $\text{ThO}_2/\text{NO}_3^- = 4$ by wt.	chromate ion	$\text{ThO}_2 \cdot x \text{H}_2\text{O}$ with CrO_4^{2-} adsorbed.

The nitrate ion was detected by the diphenylamine color test, the chromate ion by color. Movement of the sol-water boundary and plating out of a gel on the negative electrode showed that the thorium was positively charged. In addition to showing that the thorium micel in these sols is positively charged, the experiments showed that some, at least of the adsorbed anions, are not very strongly held. No test was made for nitrate ion in the gel which plated on the negative electrode.

Further development of this technique is in progress. Apparatus for work at high temperature is being designed. With this apparatus it may be possible to determine whether the inflection points in the high temperature

settling rate curves obtained by Allred for thorium slurries correspond to changes in sign or amount of charge on the thorium particles.

Sperry Products, Inc. has been contacted in connection with the ultrasonic measurement of slurry settling rates. Some preliminary room temperature measurements will be made to determine the feasibility of the technique. The problem of coupling the transducer to very hot systems is thought to be the major problem.

Other Methods for the Preparation of Thorium Sols

Exploratory experiments on other methods of sol preparation were carried out. These included:

1. Reduction of nitrate ion with hydrogen sulfide.
2. Distillation of nitric acid from a saturated solution of thorium nitrate at atmospheric pressure.
3. Pyrolysis of thorium nitrate at reduced pressure.
4. Hydrothermal decomposition of thorium oxalate in the presence of small amounts of sulfate and phosphate.

The reduction of nitrate ion by hydrogen sulfide proceeded readily at 100°C; however, the sulfide was oxidized to sulfate, resulting in no net gain. The distillation of nitric acid out of a saturated solution of thorium nitrate was very slow. This approach was abandoned when the successful super-pressure hydrolysis of thorium nitrate at ORNL was brought to our attention.

The pyrolysis of thorium nitrate at reduced pressure (~50 mm Hg) opens a new approach to the preparation of charge solutions for dialysis or ultra-filtration to separate thorium micels from ionic impurities. The experiments were carried out by repeated evaporation to dryness, heating at a fixed

temperature until no more fumes of NO_2 were observed, cooling, redispersing the residue in water and repeating the process. Two series of such treatments were performed; the results are set out in Table IV. It should be noted that complete dispersion to an opalescent sol was obtained at only one stage in the first experiment. Further exploration of this method for the preparation of samples for dialysis is intended. It has advantages in speed of removal of nitrate and in not requiring initial pH adjustment by the addition of ammonium hydroxide.

Some experiments have been carried out on the hydrothermal decomposition of thorium oxalate in the presence of sulfate and phosphate ions in amounts from 1000 to 10,000 ppm on the thorium. The treatments were made at 300°C for from sixteen to sixty-four hours. No dispersed systems were obtained. No characterization of the product has been done.

Preparation of Thorium and Thorium Phosphate Gels

Initial experiments have been carried out looking toward the preparation of thorium-containing gels to be peptized into sols or formed into beads. Thorium phosphate gels have been prepared, using the jet mixer shown in Figure 1, in both massive and bead form. When a small excess over the stoichiometric amount of phosphoric acid for $\text{Th}_3(\text{PO}_4)_4$ is employed in preparing gels at a product concentration of 1 mole (264 g. ThO_2 equivalent) per liter, the gel is hard, clean and fairly easily washed free of nitric acid without observable peptization. At lower product concentrations and with less phosphoric acid, the gel may be quite soft. When these soft gels are washed, excessive peptization of the thorium phosphate occurs.

Hydrothermal treatments of the high product concentration thorium phosphate gel have been carried out at several pH's. Qualitatively, the clear gel disintegrates to a white powder in suspension. The product from one treatment, at pH = 3.0, has been examined by x-ray diffraction. A strong diffraction pattern with many sharp lines, indicative of large crystallite size, was obtained. The original gel, examined as a faced-off block, had no observable diffraction pattern.

Thorium phosphate beads from 2 to 8 mm in diameter (wet) have been prepared by spraying the mixed thorium nitrate-phosphoric acid stream into Nujol in a two-liter graduate cylinder under the jet mixer. Washing and drying experiments on these beads are underway. It is expected that the calcined beads will range from 0.5 mm to 2 mm in diameter.

The suitability of these beads for use in a moving bed system will be studied.

No usable thorium hydroxide gels have been prepared by direct reaction of ammonium hydroxide and thorium nitrate. The variables investigated were rate of mixing, ratio of thorium nitrate to ammonium hydroxide, initial temperature of solution and product concentration. It was found that clear sols could be obtained at product concentrations as high as 0.5 M in ThO_2 at a pH as high as 4.2. When the pH was increased to about 4.6, only flocculent precipitates were obtained. Further work with this system has been suspended.

CONCLUSIONS

1. No thorium sol which has been prepared in this program is stable for as long as sixteen hours at 300°C. The presence of considerable nitrate ion in these sols may have masked beneficial effects of additives.

2. The stability of an oxide sol or slurry, as evidenced by the growth of oxide crystallites on hydrothermal treatment, is inversely proportional to the concentration of solubilizing ions in the system.

3. Thorium oxide micels in nitrate containing sols are positively charged.

TABLE I

PREPARATION OF THORIA SOLS BY DIALYSIS

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IN ALL EXPERIMENTS LISTED BELOW, UNLESS OTHERWISE NOTED, THE CHARGE SOL FOR PURIFICATION BY DIALYSIS WAS PREPARED AS FOLLOWS:

THE APPROPRIATE AMOUNT OF APPROXIMATELY 1 M ADDITIVE SOLUTION (NITRATE OR ACID ANHYDRIDE) TO GIVE THE INDICATED CONCENTRATION OF ADDITIVE ON A THORIA BASIS WAS MIXED WITH 150 ML OF 1 M THORIUM NITRATE SOLUTION. CONCENTRATED AQUEOUS AMMONIA WAS THEN ADDED WITH CONSTANT STIRRING UNTIL THE PRECIPITATE BECAME VERY SLOW IN REDISPERSING. THE FINAL PH, RECORDED FOR EACH EXPERIMENT, WAS GENERALLY 3.6 TO 3.7. DIALYSIS WAS CARRIED OUT IN THE SMALL CELL DESCRIBED IN HODURY PROCESS CORPORATION FIRST QUARTERLY REPORT, 1956.

ADDITIVE MOL % ON ThO_2	INITIAL PH	EXPT. NO.	TIMES TAKEN TO GLASS	CUMULATIVE DAYS OF DIALYSIS	WT. RATIO $\text{ThO}_2/\text{H}_2\text{O}$	REMARKS
Cr_2O_3 1%	~3.6	1139SR-1	NONE	43	4.1	
Cr_2O_3 2%	~3.6	1139SR-2	NONE	43	4.5	
Cr_2O_3 4%	~3.6	1139SR-3	NONE	30	4.8	
Fe_2O_3 1%	~3.6	1142SR	NONE	28	4.0	
CuO 1%	~3.6	1144SR	NONE	31	3.5	
NONE	-	1150SR	NONE	3	3.3	ELECTRODIALYSIS OF 1 M $\text{Th}(\text{NO}_3)_4$ HEAVY COATING OF $\text{ThO}_2 \cdot x\text{H}_2\text{O}$ ON NEGATIVE ELECTRODE, RATIO OF OXIDE/NITRATE GIVEN FOR CLEAN SOL.
NONE	3.6	1151SR-1	NONE	4	3.1	LARGE (2 LITER) BATCH IN LARGE DIALYZER.
		2	NONE	6.5	-	
		3	NONE	9.5	4.0	
		4	NONE	11.5	4.0	
	3.5	5	NONE	13.5	4.1	
	3.5	6	NONE	20.5	4.5	
	4.0	7	NONE	25.5	4.4	
CuO	3.6	1152SR	NONE	12	3.3	
NONE	-	1153SR	NONE	-	4.1	EQUIL. SAMPLE EX SINGLE PASS OF 1 M $\text{Th}(\text{NO}_3)_4$ @ 3 ML/MIN AGAINST 210 ML/HR 0.1 M NaOH USING ROMY & HAAS ANION MEMBRANE.
NONE	-	1156SR	NONE	-	4.05 (no change)	EQUIL. SAMPLE EX SINGLE PASS OF 1 M $\text{Th}(\text{NO}_3)_4$ @ 0.4 ML/MIN VS 1 M NaOH AT 4 ML/MIN USING ROMY & HAAS ANION MEMBRANE.
Cr_2O_3 4%	3.6	1158SR-1	NONE	1	2.1	
		2	NONE	-	-	
		3	NONE	-	2.6	
		4	NONE	-	2.9	
	3.5	5	NONE	-	3.1	
	3.5	6	NONE	-	3.1	
	3.5	7	NONE	-	3.0	
HfO_2 4%	3.7	1166SR-1	NONE	1	2.0	
	3.7	2	NONE	2	2.9	
	3.3	3	NONE	4	4.3	
	3.2	4	NONE	10	6.1	
	3.2	5	NONE	12	7.2	
	4.7	6	NONE	13	-	SOL CONCENTRATED TO DRYNESS, WOULD NOT REDISPERSE.
La_2O_3 4%	3.7	1167SR-1	NONE	4	2.7	
	3.8	2	NONE	8	3.7	
	3.6	3	NONE	15	3.2	
	3.6	4	1	17	4.5	
	3.8	5	1	23	5.3	
	3.8	6	1	36	13.7	FINAL CONCENTRATION.

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TABLE 1 (CONTINUED)

ADDITIVE MOLE % OF TiO_2	INITIAL PH	EXPT. NO.	TIMES TAKEN TO GLASS	CUMULATIVE DAYS OF DIALYSIS	WT. RATIO TiO_2/H_2O_3	REMARKS
SiO_2 1%	3.7 3.7	1168SR-1A -2B	5 5	14 15	4.7	THE PH ADJUSTED CHARGE CONTAINED A SILVERY SLURRY, A BOTTLE TYPE DIALYSIS CELL WAS USED WITH CONSTANT STIRRING. PRODUCT OBTAINED WAS CLEAR SOL, STILL GAVE QUALITATIVE TEST FOR HS^{3+} .
Fe_2O_3 1%	3.6 3.6 3.5 3.5	1169SR-1 -2 -3 -4	NONE NONE NONE NONE	5 6 15 22	1.35 3.0 4.3 6.0	
		3.5 3.6 3.7	1 0 1	30 31 38	11.5 - 17.6	
P_2O_5 1%	3.7 3.5 3.5 3.5 3.8 3.7 3.9	1170SR-1 -2 -3 -4 -5 -6 -7	- - - 1 - 1 -	1 7 13 20 26 30 41	2.0 4.0 5.9 4.1 - 6.3 14.8	
Y_2O_3 1%	3.6 3.6 3.7 3.6 3.8 3.9	1171SR-1 -2 -3 -4 -5 -6	0 0 0 1 1 0	1 6 13 20 29 42	1.7 2.7 2.0 4.2 5.9 10.0	
CrO_3 16%		1176SR-1				FAILED, SOLY. OF TiO_2 X CrO_3 TOO LOW FOR THIS RATIO.
NONE	3.8	1180SR -1 -2 -3 -4	1 2 2 2	16 23 - 33	4.6 10. - 16.	STARTED WITH 100 G OF PRODUCT FROM 16 DAYS DIALYSIS IN LARGE CELL - SEE FIRST QUARTERLY P. 4.
NONE	3.5 3.1	1187SR-1 -2 -3 -4 -5	0 1 1 1 1	0 11 - - 35	- 4.1 - - 44.	

15 FIGURE 1

NR FROM FILTER AND REGULATOR

JET MIXER

SCALE: 1/4" = 1" 6-14-56

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Laboratory Records Division
Oak Ridge National Laboratory
P. O. Box P
Oak Ridge, Tennessee

July 6, 1956

Attention: Mr. F. R. Bruce

Reference - Subcontract 904 under W-7405 eng-26
Second Quarterly Report, 1956

Dear Mr. Bruce:

The second Quarterly Report is submitted herewith.

Thoria sols were prepared by dialysis of thorium nitrate solutions by a new, rapid technique. The ability of these sols to remain in suspension in unstirred bombs at 100°-300°C was measured by x-ray examination of the bombs. The sols were found to be stable at 100°C but unstable at 300°C. Addition of a series of inorganic agents did not achieve stabilization.

Thoria sols of higher purity are being prepared by reduction or hydrolytic decomposition of thorium nitrate and peptization of pure hydroxide or phosphate gels.

Thorium phosphate beads of 2 mm diameter have been prepared, looking toward the possibility of their use as a controlled non-caking "slurry".

The problem of instrumentation for measuring sol and slurry properties is being given greater attention. Electrophoresis has been used to determine the sign of the charge on thoria micels. The development of ultrasonic settling rate measuring apparatus is being explored with Sperry Products, Inc.

Very truly yours,

G. A. Mills.

Do not
Photostat

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