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SUBJECT: HRP-CP: Conclusions Concerning the Predicted Behavior of Insoluble Corrosion and Fission Products in the HRT
TO: F. L. Culler
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ABSTRACT

Information available from HRP-CP development studies was summarized and used as a basis for predicting the probable behavior of corrosion and fission product solids in the HRT and its associated chemical processing plant. Operation of the chemical plant will probably limit the HRT circulating solids concentration to between 10 and 100 ppm; quantitative predictions are uncertain due to uncertainties concerning the characteristics of reactor solids and the effects of reactor geometries.

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1.0 SUMMARY

Data are presented to support the following conclusions which are based on observations of the behavior of simulated HRT corrosion and fission product solids and of loop corrosion products.

These conclusions for the HRT and its chemical processing plant are:

- (a) A major fraction of the solids resulting from corrosion and from fission product precipitation will appear in the circulating fuel solution.
- (b) Significant amounts of the circulating solids will be removed by "separation" due to centrifugal force or due to gravity in areas of low velocity and by "deposition" in areas which may include those of high velocity. Geometry is an important variable. The complicated HRT geometry along with uncertainty as to the corrosion product particle size precludes any exact prediction about corrosion product behavior in the HRT.
- (c) Hydroclone efficiencies are predicted for known solids and solution characteristics. The simulated corrosion products had smaller particle diameters than initially predicted for HRT corrosion products. Uncertainty concerning the HRT corrosion product particle size results in uncertainty concerning the CPP hydroclone efficiency.
- (d) Essentially continuous operation of the HRT-CPP will maintain the circulating solids concentration below 200 ppm with probably values of between 10 and 100 ppm.

- (e) The effectiveness of the chemical plant could be increased by increasing the flow through the plant and by designing the reactor system to minimize "separation".*
- (f) The fraction of solids collected by the chemical plant depends on competition of the hydroclone with deposition and/or segregation and is therefore more uncertain than the above factors individually. This factor should increase from a zero initial value to a constant average value for periods of steady state reactor operation. An average value as low as 25% of the corrosion products formed would not be surprising.
- (g) Fission products present as relatively insoluble sulfate salts will preferentially deposit on hot surfaces. Otherwise, the behavior of insoluble fission products will be obscured or controlled by the corrosion product behavior, and the same conclusions apply to both.
- (h) The quantitative behavior of solids in the reactor system cannot be predicted with confidence from any non-reactor experiments. The number of important variables and their interrelationships invalidate results from a simplified system. Until the HRT and its chemical plant operate, conclusions more definite than those presented above are not justified.

2.0 INTRODUCTION

The purpose of this memo is to summarize information presently available on and to present predictions for the probable behavior of corrosion and fission product solids in the HRT and its associated chemical processing plant. The basis is observations of the behavior of simulated HRT corrosion and fission products and of loop corrosion products in various types of laboratory equipment and

*Initial operation of the HRT-CPP is with 0.56 in. D hydroclone to give an over 50% flow increase as compared to the initially designed 0.40 in. D hydroclone.

experimental loops. Conclusions are presented with a minimum amount of supporting data since detailed data and procedures have been presented in status reports and/or will be available in topical reports. All data were evaluated, but the data presented may be biased in the sense that they were selected to support the conclusions presented. The detailed data include some confusing or conflicting material and a large amount that is indeterminate with respect to conclusions about solids behavior in the HRT.

Studies have been reported in literature for the formation and behavior of boiler scale, and for the formation of films from transported oxides in circulating water.^(1,2) None of the literature found was adequate for predicting the behavior of corrosion products in the HRT fuel system.

3.0 BEHAVIOR OF SOLIDS

The insoluble corrosion and fission products in the UO_2SO_4 solution fuel have three distinct modes of behavior depending on their type and their source. Each type was studied separately and in combination with the other types. The behavior differences were still apparent in mixtures of types although a small amount of one type will tend to follow the behavior of the predominant material. The fission products will first appear as positive ions; therefore the initial at-rest status of cations can be considered to be dissociated sulfate salts. Corrosion products form below an oxide film; circulating insoluble corrosion products are released as insoluble particles from the surface of the film. The predicted formation rates for the HRT at design power are about 50 g/week of insoluble fission product salts and 350 g/week of corrosion product oxides.⁽³⁾ Solubilities can be expressed in terms of MWD for various classes of cations (Table 1).

Table 1. Insoluble Major Fission Products⁽⁴⁾

Chemical Group or Element	Rate of Production (mg/Kg D ₂ O·MWD)	Solubility (mg/Kg D ₂ O)	Time to Saturation at 300°C MWD
Rare earths	0.67	85	127
Alkaline earths	0.15	10	67
Zirconium	0.27	<1	< 4
Ru, Rh, Pd	0.16	<5	< 31

3.1 Behavior of Insoluble Sulfates

Sulfate salts which become more insoluble without decomposing as the temperature increases were found to deposit preferentially and reversibly on "hot walls" which have surface temperatures above the solution temperature. The steady state amounts circulating in loops did not exceed the solubilities by any significant amount. The major insoluble sulfates in the UO_2SO_4 fuel solution would be the rare earth fission products since their solubility decreases as the temperature increases. The rare earths including Nd, Sm, and Ce are a major part of the fission product poisoning. Most of the studies were made with $\text{Nd}_2(\text{SO}_4)_3$ as a representative material since this salt and Nd-147 tracer were easily available. Mixed rare earths and Ce-141 tracer were used in the loop studies to compare mixtures with pure $\text{Nd}_2(\text{SO}_4)_3$. Other pure rare earths were used in laboratory studies to make more detailed comparisons.

Laboratory solubility data was obtained for a number of rare earth sulfate salts and mixtures of salts (Table 2) (Figs. 1 and 2). The solubility of a specific salt is reduced by the presence of other rare earths. The concentrations circulating in loops were found to be essentially solubility values for the loop conditions. A reversible temperature-concentration curve of this type was obtained in the absence of hot walls in loop B2 (Fig. 3). The reversibility of Nd concentration was demonstrated for twenty-five temperature cycles in loop A (Fig. 4)⁽⁵⁾. Recovery of Nd was excellent for most of these tests (85-110%).

The insoluble $\text{Nd}_2(\text{SO}_4)_3$ deposited out on loop walls very rapidly with a pronounced preference for walls with temperatures above the solution temperature. During one loop A test designed to give a high heat flux in the preheater to the pressurizer, the preheater collected 99% of the Nd added to the loop while a hydroclone after the preheater collected 1/2%⁽⁶⁾. Even when corrosion product

Table 2. Solubility of Rare Earth Sulfates in UO_2SO_4 Solution

Conditions: 0.02 M UO_2SO_4

0.005 M H_2SO_4

280°C

Salt	Solubility in mg/Kg H_2O
$\text{La}_2 (\text{SO}_4)_3$	10
$\text{Ce}_2 (\text{SO}_4)_3$	50
$\text{Nd}_2 (\text{SO}_4)_3$	110
$\text{Pr}_2 (\text{SO}_4)_3$	170
$\text{Y}_2 (\text{SO}_4)_3$	240
$\text{Sm}_2 (\text{SO}_4)_3$	420

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SOLUBILITY OF $\text{Nd}_2(\text{SO}_4)_3$ IN $0.02 \text{ M } \text{UO}_2\text{SO}_4$,
 $0.005 \text{ M } \text{H}_2\text{SO}_4$

FIGURE 1

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SOLUBILITY OF MIXED RARE EARTH SULFATES IN
0.02M UO_2SO_4 , 0.005M H_2SO_4 SOLUTION.

(48% Ce, 24% La, 19% Nd, 6% Pr, 2% Sm, 1% others)

FIGURE 2

FIGURE 3
Neodymium Sulfate Concentrations vs Fuel Temperature in Loop B₂

FIGURE 4

oxides were present, the $\text{Nd}_2(\text{SO}_4)_3$ deposited preferentially on hot surfaces (7,8) and formed separate crystals of precipitated material. The solids were redissolved completely and the effects on heat transfer were completely reversed by dropping the temperature of the circulating solution. (5)

The results with $\text{Nd}_2(\text{SO}_4)_3$ are apparently reasonably representative of the behavior of the other rare earth sulfates; this is indicated by extensive laboratory (9,10) and limited loop tests (7,11). As previously mentioned, the actual solubilities are greatly reduced by the presence of other rare earths. Various rare earths may co-precipitate with other rare earths, or, in some cases, with UO_2SO_4 . (9)

The rates of precipitation and re-solution were normally too fast for detection in dynamic loop experiments. When the $\text{Nd}_2(\text{SO}_4)_3$ concentration exceeded the solubility gradually either by slow additions or temperature increases, the amount circulating showed an excess over the solubility for as long as four hours after which the amount circulating dropped back to and stayed at the solubility value for the conditions in the loop. Efforts to obtain crystals of $\text{Nd}_2(\text{SO}_4)_3$ on filters kept at loop temperature and pressure were not successful. Any flashing reduced the temperature and dissolved the $\text{Nd}_2(\text{SO}_4)_3$ while temperatures above the loop temperature caused deposition in the filter. The deposits on the preheater hot walls were removed and examined (12) after draining the loop while maintaining the operating temperature and pressure. Some deposits completely plugged the preheater cross section for flow.

The insoluble fission product sulfates will be a major fraction of the nuclear poisoning and radioactivity in the HRT fuel solution, but will be less than 10 per cent of the total insoluble fission and corrosion products. Future reactor designs may increase the fraction of the insoluble material which is fission products, but the amount of corrosion products will probably always be at least several times the amount of fission products.

3.2 Behavior of Oxides from Hydrolyzed Sulfate Salts

When sulfate salts are hydrolyzed in UO_2SO_4 fuel solution circulating in a loop, the resulting insoluble oxides are deposited on the available surfaces and do almost no circulating. This is distinctly different from the behavior of oxides which appear in circulation as an insoluble slurry. Therefore the addition of soluble sulfate salts does not give representative results for corrosion product behavior. The sulfate salts available for hydrolyses are a fraction of the fission products; therefore the amount of this type of insoluble material is the smallest of the three types of solids studied.

Results typical of the addition of soluble Zr or Fe sulfates to precipitate oxides in the loop were 3, 4, 8, and 0.1 per cent in circulation and 110, 50, 49, and 47 per cent of the material added recovered. (13, 14, 15, 16, 17) Two loops were disassembled and inspected after the addition of large amounts of $\text{Zr}(\text{SO}_4)_2$. (15,17) Both showed layers of hard dense scale 0.002 to 0.006 in. thick over essentially all of the loop surfaces (Fig. 5).

Although the amounts of hydrolyzed oxides circulating were small percentages of the amounts of sulfate added, major amounts (16, 70, 45, and 65 per cent) were collected by the hydroclones in the loop systems. The mean apparent Stokes diameter on a weight percentage basis of ZrO_2 collected by a hydroclone was about 1 micron. The lowest percentage collection listed was during a long term continuous addition tests with low circulating solids concentrations at all times. At a 100 mg/l concentration of 1 micron diameter, the surface area of the circulating solids will approximately equal the wall area in the RRT. Since the solids are throughout the turbulently flowing fuel solution, they should effectively compete with the walls for collecting the freshly precipitated oxides from sulfate salts. This type of effect could explain most of the variations in material balances and fractions collected by the hydroclones during tests with $\text{Zr}(\text{SO}_4)_2$ addition.

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Pump Impeller
Scale hard and
dense .002 in.
thick

Thermal Barrier
Scale same as on
impeller

Scale soft
and light
colored

- 13 -

Fig. 4.1 Pump Impeller and Thermal Barrier

3.3 Behavior of Preformed Oxides

Corrosion product oxides released into the circulating fuel solution as solid particles circulate to a much greater extent than sulfate salts hydrolyzed to oxides, but the characteristics and behavior of these solids in the HRT has not been definitely established. These oxides of Zr, Fe, and Cr are expected to be over 80 per cent of the circulating solids formed in or released to the HRT fuel solution.

The initial procedure selected for studying these corrosion product oxides was to hydrolyze sulfate salts in the circulating loop solution. The oxides circulating after hydrolyses was complete were expected to behave as material released from the surface of corrosion films. The small fractions (less than 10 per cent) of the oxides which circulated after hydrolyses indicated that both the results and this method of formation were of doubtful validity. To check the above conclusion, a slurry of ZrO_2 was added to loop A for conditions otherwise comparable to those previously used for a test with $Zr (SO_4)_2$ addition (Table 3).⁽¹⁷⁾ The preformed ZrO_2 or mixed oxides were prepared by precipitating hydroxides from sulfate salt solutions with aqueous NH_4OH , washing with H_2O and autoclaving at $300^\circ C$ in $0.04 M UO_2SO_4$ with O_2 overpressure. When concentrations were to be determined from γ activity counts, Zr-95 was added before precipitation or the preformed ZrO_2 was irradiated in a nuclear reactor before use.

Higher corrosion rates for zirconium and stainless steel under radiation, as compared to non-radiative conditions, indicates changes in the oxide film on the metals and thus in the corrosion products released to the circulating solution. Settled corrosion product oxides recovered from corrosion test loops have complicated histories and their properties may not be characteristic of what is initially released from the corrosion sites. General indications are that the loop corrosion products have larger, scale like particles with higher settling rates as

Table 3. Comparison of Zr (SO₄)₂ and ZrO₂ Behavior

Results	Zr (SO ₄) ₂ Addition Test	ZrO ₂ Addition Test
Zr circulating (%)	<1%	7%*
Zr recovered (%)	47%	99%
Zr in hydroclone pot (%)	15%	87%

* This would be ~60% if the ZrO₂ removed by the hydroclone were subtracted from the amount of feed Zr.

compared to the preformed oxides used for development studies. Some properties found for preformed oxides and some other materials examined during experimental studies are tabulated for comparison (Table 4).

Results have shown that geometry including the type of surface, the surface to volume ratio, the wall to fluid temperature differences, and the existence of dead end or no flow regions is a controlling factor when comparing different systems. Because of the controlling geometry effects, variations of temperature, flow rate, and concentration did not give consistent results for the initial tests with preformed ZrO_2 .⁽¹⁸⁾ Later studies with more carefully controlled conditions showed consistent variations in the amount and rate of ZrO_2 deposition on coupons with Reynold's number changes caused by flow rate or temperature changes.⁽¹⁹⁾

Higher initial ZrO_2 concentrations gave somewhat higher, but not directly proportional concentrations throughout the test periods. Location of ZrO_2 in the circulating pump at the end of experimental studies⁽²⁰⁾ and location of corrosion products in relatively stagnant areas of the HRT Mock-up or of REED corrosion loops has confirmed the tendencies of these regions to collect solids.⁽²¹⁾

Results for successive additions of ZrO_2 to an operating system were compared to results for a single addition of the same total amount.⁽²⁰⁾ The successive additions resulted in concentration versus time curves having successively flatter slopes and higher steady state concentrations (Fig. 6). A test with continuous addition of traced ZrO_2 for three periods totaling over 250 hours and essentially continuous hydroclone operation was made to determine the steady state loop and hydroclone underflow receiver concentrations. Average steady state concentrations during the continuous ZrO_2 addition were about 380 mg/kg H_2O of solids in the loop and 2000 mg/kg H_2O in the underflow receiver.⁽¹⁹⁾ At steady state, the ZrO_2 added was collected in the underflow receiver at 50% of the addition rate; the other 50% disappeared from circulation and was not recovered by draining and flushing the loop.

Table 4. Properties of Simulated Insoluble Corrosion Products

Properties	Units	Values obtained for:							
		ZrO ₂			Iron Oxides	ThO ₂	Mixed Corrosion Product Oxides		
Method of Preparation	--	Hydrolysis in Loop	Preformed	Preformed	Hydrolysis in Loop	Oxalate Decomposition	Preformed	Preformed	Preformed
Where Used	--	Loop A	Loop A	Loop B ₂	Loop B	Loop B ₂	HRT Mock-up	HRT-CPP	Laboratory Studies
Purpose of Studies	--	Behavior of Solids	Behavior of Solids	Behavior of Solids	Hydroclone Efficiency	Hydroclone Efficiency	Behavior of Solids	Behavior of Solids	High Temperature Settling Rates
Compositions:									
ZrO ₂	Wt %	>90	100	100	--	--	47	47	46
Fe ₂ O ₃	Wt %	--	--	--	>90	--	40	40	41
Cr ₂ O ₃	Wt %	--	--	--	--	--	13	13	13
(RE) ₂ (SO ₄) ₃	Wt %	--	--	--	--	--	0	0	0
ThO ₂	Wt %	--	--	--	--	100	0	0	0
Size Distributions:									
40% smaller than	Microns	0.7	--	--	0.2	0.9	0.5	--	0.3
Mean size	Microns	1.0	--	<0.1	0.3	1.1	0.7	0.7	1
80% smaller than	Microns	4.0	0.2	0.2	0.6	1.5	2.3	2.5	5

FIGURE 6
Suspended Solids vs Time for Successive Additions of Loop to A-Loop

Tests of limited scope with preformed oxides were made with the relatively complicated geometries of the HRT-CPP and the HRT Mock-up for comparison with results from simpler loop systems.^(21,22) Results of the most typical tests in each system are most easily compared by tabulation (Table 5). The parameters for these systems have also been tabulated (Table 10). The HRT-CPP studies clearly showed sudden sharp increases in circulating solids concentrations⁽²²⁾; similar changes were observed to a lesser degree in loop A and in other studies. The reappearance of solids often followed temperature changes, but some reappearances occurred with no visible changes in experimental conditions.

The objective of the studies with preformed oxides was to determine the behavior of corrosion product oxides in the HRT. Since geometry including types of surface and temperature gradients is a controlling variable, the results are exact only for the conditions for and system in which they were obtained. The results have disproved several preconceived theories, but do not clearly support any single one of several conflicting hypothesis. No single experiment or series of experiments is likely to permit quantitative predictions as to the behavior of the corrosion product oxides in the HRT. Basic studies of the physical chemistry of these materials in a dynamic system would probably be the most effective method of obtaining additional information on corrosion product behavior.*

The results and the reasonable hypothesis permit agreement on several conclusions about behavior of the HRT corrosion product oxides. One is that corrosion product oxides released from the surfaces will circulate for average times on the order of hours before the chemical processing plant or the mechanical "separation" remove them from circulation. Another is that the chemical processing plant will collect significant amounts of the insoluble corrosion and fission products, but significant amounts will not circulate long enough to reach the chemical plant.

* Studies of this type are being carried out by the Stanford Research Institute as SRI Project No. SD-2080-2 under contract with the Oak Ridge National Laboratory.

Table 5. Summary of Typical Results with Preformed Oxides

Experimental Condition or Result	Units	Values or Conditions for Experiment Tests:						
Experimental Facility	--	HRT Mock-up	HRT-CPP	Loop A	Loop A	Loop A	Loop B ₂	Glass Loop
Run No.	--	--	--	LR-50	LR-S-15 to LR-S-24	LR-S-34 to LR-S-38	LB ₂ -S-4	GL-6 through GL-11
Material Used	--	Mixed Oxides	Mixed Oxides	ZrO ₂	ZrO ₂	ZrO ₂	ZrO ₂	ZrO ₂
Temperatures	°C	280	295	300	115-260	70	140	47-77
Amount of Solids Added	Grams	1042	1468	15.4	0.5 to 1.5	.38 - 1.3	22	0.4
Amounts Recovered:								
Underflow Receiver	%	23	53	87	--	--	23	--
Samples	%	1	--	3	--	4-25	23	--
End of Run Drain	%	1	17	10	--	15-150	24	45-80
Other	%	35	13	--	--	20-55	7	20-55
Total Recovery	%	60	83	100	--	70-190	77	85-110
Amounts of Solids Circulating after 4 hr	%	5 to 50	1 to 10	60	25-50	2-15	50	70-90
Results Reported in (See Bibliography)	--	21,22	22	17	18	20	19	19

4.0 CHEMICAL PLANT PERFORMANCE

Predictions as to the chemical plant performance can be made in terms of the performance for specified feed slurries and of what upper solids concentration limits are imposed on the reactor. The rate of solids collection and the actual circulating solids concentration are uncertain due to the lack of information on the true precipitate properties and their behavior in the reactor. Interpretation of HRT-CPP results will be based on the experience from core processing development studies.

4.1 Hydroclone Efficiency

The HRP-CPP hydroclone development studies permit predictions as to hydroclone performance as a function of the physical characteristics of the liquid and solid separated. (25) The larger hydroclones process larger volumes (Fig. 7) with less efficiency. For HRT-CPP conditions, the efficiency correlation reduces to the following expressions for the particle diameter removed with 50% efficiency, d_{50} , for various hydroclone diameters, D_c :

$$d_{50} (D_c = 0.25) = 1.9/\Delta P^{0.25}$$

$$d_{50} (D_c = 0.40) = 2.4/\Delta P^{0.25}$$

$$d_{50} (D_c = 0.56) = 2.9/\Delta P^{0.25}$$

where ΔP is the pressure drop through the hydroclone in feet of liquid. The efficiencies for constant pressure drops can be expressed as percentage efficiency versus particle diameter curves (Fig. 8) or solids removal rates versus particle diameter curves (Fig. 9).

The HRT-CPP is designed to accumulate the heavy phase in an underflow receiver; this concept of operation is called induced underflow operation. (25) If the underflow receiver slurry remains uniform, the equilibrium concentration factor,

FIGURE 7

Hydroclone Pressure Drop vs Flow Rate.

Hydroclone Efficiency vs. Particle Diameter. $\Delta P = 80$ ft; $C_f(F/B) \leq 100$ g/liter.

FIGURE 8

Fig. 11.4. Rate of Solids Removal by Hydroclone vs. Particle Diameter. $\Delta P = 80$ ft;
 $C_p(F/B) < 100$ g/liter; To convert solids removed to g per day/ppm multiply by 5.45.

FIGURE 9

CF_{∞} between the feed stream and underflow receiver is given by:

$$CF_{\infty} = \frac{F}{B} \left(\frac{E_1}{1 - E_2} \right)$$

The values of $\frac{F}{B}$, the feed volume rate to the induced underflow volume rate, are about 70 for the 0.25 and 0.40 in. D hydroclones and 50 for the 0.56 in. D hydroclone. The values of E_1 were previously presented. Values of E_2 are difficult to determine because of the elusive nature of the induced underflow stream; the following expression fits the limited amount of experimental data available. (25)

$$E_2 = 2E_1 - E_1^2$$

Based on the above expressions, values of CF_{∞} versus particle diameter can be calculated for HRT conditions* (Fig. 10). The heavy phase collected may settle out in the underflow receiver. Then the limiting concentration is the settled concentration of the heavy phase. Settled beds of solids have been collected in underflow receivers during development studies. In the HRT-CPP, the fission product heat will give thermal convection currents which will stir up the underflow receiver.

4.2 Equilibrium Solids Concentration

Values for equilibrium concentrations of solids circulating in the reactor fuel loop can be calculated after selecting values for corrosion rates, solid characteristics, and the removal of solids by other agencies than the chemical processing plant. Calculations from several logical bases are presented and give values of 10 to 125 ppm.

* Present plans are to operate the HRT-CPP with 4 P-120-130 ft instead of the 80-100 ft initially designed for.

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Fig. 10. Equilibrium HRT-CPP Concentration Factors.

Original Design Basis (All solids removed by CPP):

Solids Production = 400 g/week = 40 mg/min = 4.6 mg/l·hr

Processing rate = 2.6 l/min

Efficiency (E_1) = 40% ($\sim 0.8 \mu$ D particle)

Equilibrium solids concentration = $\frac{40 \text{ mg/min}}{(2.6 \text{ l/min})(0.40 \text{ mg/mg})} = 38 \text{ mg/l}$

Revised Design Basis (Equilibrium deposition with CPP solids removal rate equal to solids production rate):

Solids production = 400 g/week = 40 mg/min

Processing rate = 5 l/min

Efficiency = 10% ($\sim 0.5 \mu$ D particles)

Equilibrium solids concentration = 80 mg/l

Basis of Extended Operation of Loop B₂. The single test most comparable with the HRT and HRT-CPP was in loop B₂.⁽¹⁹⁾ The results and a comparison of parameters (Table 6) permit calculation of predicted HRT equilibrium solids concentrations. The HRT volume per unit surface is 60% of that for loop B₂; for equal circulating times, the removal by other agencies than the CPP should be smaller than for loop B₂; 60% of the B₂ amount would be 30% of the total present. At equilibrium, the loop B₂ hydroclone removed about 50% of the preformed ZrO₂ added. For the HRT-CPP:

Solids removal rate = (0.70)(40) = 28 mg/min
(This leaves 12 mg/min or 30% to be removed by other agencies)

Processing rate = 5 l/min

Hydroclone efficiency = 8% (The calculation will assume that this efficiency is maintained either by settling in the underflow receiver or by frequent dumping).

Equilibrium solids concentration = $\frac{28}{(5)(0.08)} = 70 \text{ mg/l}$

A second method of converting the loop B₂ results to the HRT would be:

Table 6 Parameters for LB₂-S-4 as Compared to the HRT and HRT-CPP

Quantity	Units	Loop B ₂ Values	HRT Values
Solids Addition Rate	mg/min	1.67	40
Solids Addition Rate	mg/hr-liter	10	5.
<u>Hydroclone Pot Volume</u> Loop Volume	liter/liter	1/100	3/200
<u>Hydroclone Pot Volume*</u> Induced underflow rate	min.	10	100
Processing time or loop volume/hydroclone flow	min.	15	100
Volume/area	ft ³ /ft ²	.05	.03
Hydroclone efficiency	%	8	8**
Hydroclone flow	liter/hr	40	300
Underflow pot volume	liter	0.1	8
Equilibrium Circulating Solids Concentration	mg/liter	380	50-200***
Average Solids Circulation Time	hrs.	38	10-40

*Based on a 1 1/2% induced underflow rate.

**The effects of the hydroclone diameter and the liquid viscosity changes will approximately cancel.

***This range due to effects of underflow pot dump cycle and/or amount of settling in HRT underflow pot.

$$\begin{aligned} \text{Equilibrium solids concentration} &= \frac{(380 \text{ mg/l})_{B_2} (5 \text{ mg/hr l})_{HRT} (0.64)^*}{(10 \text{ mg/hr l})_{B_2}} \\ &= 122 \text{ mg/l} \end{aligned}$$

Hydroclone Corrosion Tests. Hydroclones have collected loop corrosion products during hydroclone corrosion tests. These collection rates have been up to 0.1 g/hr averaged over 50 hour periods. Loop corrosion product concentrations were too low for accurate measurement, but were in the 1/2 to 5 ppm range for circulating solids. To collect 2.5 g/hr, the HRT solids production rate, would require a circulating solids concentration of 10 to 125 ppm.

4.3 Fraction of Solids Collected

Efforts to fix the expected value for this quantity represented a major part of the core processing development studies. However, the information obtained is not adequate for predicting a value and operation of the HRT-CPP may not be satisfactory for predicting values for other reactor systems. An expression for the fraction of solids collected would combine the characteristic behavior of the solids, the hydroclone efficiency, and the effects of system geometry and thus it would combine the uncertainties associated with these individual factors.

Several of the studies with preformed oxides^(19,22) could be considered representative of what fraction of the HRT insoluble fission and corrosion products are collected. Values of 50 to 70% from loop B₂ and HRT-CPP studies with preformed oxides seem most applicable. Other results in loop A and from settling characteristics in bombs at 300°C can be used to justify estimates down to as low as 10%.

4.4 Interpretation of HRT-CPP Results

One objective of core processing development studies was to permit interpretation of the observed HRT-CPP results; this is a less difficult objective to achieve

* Correction factor for decreased efficiency in loop B₂ due to concentration of solids in underflow receiver.

than the prediction of results before operation. The detailed development studies as a whole provide a basis of knowledge from which results will be correlated and interpreted. Several topical reports will be required to adequately present this material. The information which appears to be of greatest importance has been summarized in this memo (Section 3.0).

The performance of the HRT-CPP will have to be evaluated from analyses of highly radioactive fuel solution or slurry samples along with information deduced from the operation of reactor components. Samples during circulation of the underflow receiver before dumping and samples from the dissolver product will show what has been collected by the CPP. The solids concentrations in the CPP feed will not be directly available; the hydroclone overflow and the reactor fuel cycle samples will probably give lower and upper limits respectively. These values will establish average circulating solids concentrations and along with the total solids removed will establish average hydroclone efficiencies. The amount of solids placed in circulation and thus the fraction collected will probably not be determined with any confidence. Collection of iron oxides at a rate equivalent to the calculated corrosion rate (either from nickel concentrations or experimentally determined corrosion rates) would be excellent evidence for high collection efficiency.

From the reactor viewpoint, the duty of the CPP is to limit the nuclear poisoning due to insoluble corrosion and fission products and to limit their mechanical effects such as heat transfer surface fouling, pump bearing wear, valve failures, etc. The nuclear poisoning can be calculated for known circulating solids compositions and concentrations. Successful prevention of mechanical effects can only be demonstrated by their absence over long periods of operation. If the CPP collects insignificant amounts of insoluble materials because of low circulating solids concentrations, it may demonstrate that it is unnecessary without being a failure from the reactor viewpoint.

5.0 REVIEW OF BEHAVIOR STUDIES COMPLETED

The objectives and scope of the core processing development program have shifted even after centrifugal separation with a hydroclone was selected as the basic processing method. These shifts were due to increased knowledge about the precipitate formation mechanism and composition and about the factors controlling processing efficiency. A knowledge of the continuity of what, why, and how the studies were made is required to properly evaluate the meaning of the results.

5.1 Chronological Review

The paths followed by the solids behavior program can be outlined under three major headings. The controlling factor is the changing major objectives throughout the experimental program (Table 7). The experimental equipment used and the solid circulated for study (Table 8) were partly dictated by the objectives and partly by availability. The A loop was the first equipment available and the HRT Mock-up and Chemical Processing Plant were available for limited periods of time. The major results are tabulated on a monthly basis (Table 9). As these results and additional corrosion data became available, they changed the objectives of the solids behavior studies.

5.2 Validity of Results

Limitations on the validity of results can be conveniently grouped into: (a) those due to the experimental facilities, (b) those due to the experimental procedure, and (c) those due to the selection of synthetic materials to represent the fission and corrosion products. All three limitations must be considered when interpreting the results for preformed oxides; the first two are relatively unimportant to the results for insoluble sulfates and for sulfate salts hydrolyzed to oxides.

Table 7. Objectives of Solids Behavior Studies in Loops

Subject Studied	Material	Property (i.e., objective) of interest	1954			1955					1956					1957																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																														
			Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																
Rare earth fission product behavior	$\text{Nd}_2(\text{SO}_4)_3$	Size, shape, etc. of precipitated $\text{Nd}_2(\text{SO}_4)_3$																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																												

* Some information available from early hydroclone long term corrosion tests and from REED corrosion studies prior to August 1957.

Table 9. Solids Behavior Studies:

Chronological Tabulation of Loop Results

September 1954 } October 1954 }	A filtering sampler did not collect $\text{Nd}_2 (\text{SO}_4)_3$ crystals for examination.
October 1954	$\text{Nd}_2 (\text{SO}_4)_3$ did not remain in circulation.
November 1954	Nd collected preferentially in the loop preheater and pressurizer.
December 1954 } January 1955 }	Nd disappearance was temperature reversible almost at will, Nd could plug a preheater.
February 1955	Hydroclone efficiency of 10-12% in B loop for short periods; loop B too complicated for its purposes.
March 1955	High hydroclone efficiency for iron oxides in B which were <0.5 micron D.
April 1955	High (90% +) recovery of Nd possible, over 50% of Nd in preheater in loop A.
May 1955	Nd solubility values established in B_2 . No hot wall effects for $\pm 2^\circ \text{C}$.
June 1955	ThO_2 circulated in B_2 at below 100°C , did not circulate above 100°C . Mixed rare earths similar to Nd in loop B_2 .
July 1955	Over 50% of Nd can be collected in preheater even though this test was upset by preheater plugging. Ce concentration in B_2 were only partially reversible.
August 1955	99% of Nd in heater designed for maximum hot wall effect, about 1/2% in hydroclone pot.
October 1955	ZrO_2 precipitated by $\text{Zr} (\text{SO}_4)_2$ addition was concentrated by hydroclone, but total material balance was poor.
November 1955	Hydroclone concentration factor of 4500 for Zr in loop A. ZrO_2 was $\sim 1 \mu$ mean D after precipitation from $\text{Zr} (\text{SO}_4)_2$ addition.
December 1955	Mixed rare earths in loop A similar to $\text{Nd}_2 (\text{SO}_4)_3$. Over 50% of rare earths in preheater and filter.
January 1956	For Zr + Nd, Zr goes to hydroclone pot, Nd to preheater in loop A.

February 1956 Loop A results for Nd and Nd + Zr confirmed in loop B₂. Loop B₂ Zr recovery poor.

April 1956 Uniform scale over all of loop B₂ including pump and flow restrictor from Zr (SO₄)₂ added to precipitate ZrO₂.

May 1956 With Zr (SO₄)₂ and Nd₂ (SO₄)₃ added to loop A, Nd concentration and preheater heat transfer coefficient were completely temperature reversible while the Zr concentration showed no temperature reversibility. These results tested for 25 temperature cycles.

June 1956 Underflow pot tests results gave no conclusions on solids behavior for corrosion products as sulfate salts additions; this result due to sampling and analytical problems.

July 1956 Zr behavior from Zr (SO₄)₂ addition to blanket U concentration in titanium loop T₁ comparable to previous results except for higher Zr concentration for same temperature.

August 1956 Mock-up tests completed with no conclusive results. 1042 g of solids added.

September 1956 34 g of Zr added as Zr (SO₄)₂ to loop A all plated on walls as a dense, hard scale which was 52-68% Zr. This was a 500 hour test.

October 1956 18.4 g Zr added to loop A as ZrO₂ was recovered (98%) with 75% to the hydroclone to give a pot concentration of 100 g/l.

November 1956 Review of results indicated that hydroclone performance was well established and that behavior of corrosion products after they scaled off was most important question. Loop alterations made for these tests.

December 1956 HRT-CPP tests showed 68-73% recovery with hydroclone concentration factors of > 1700.

January 1957 }
February 1957 } Results in loop A with preformed ZrO₂ indicated that for "deposition":

- Temperature had only minor effects (115-250°C) with a slight rate increase with temperature increase.
- Flow rates had only minor effects.
- Higher initial concentration gives higher value of concentration throughout tests.

March 1957	For preformed ZrO_2 , concentrations are not proportional to initial concentrations, but are increased by larger values of initial concentrations.
April 1957 May 1957 }	Deposition of preformed ZrO_2 visible on glass loop walls, but rate of disappearance is smaller than in loop A.
June 1957	A combination of "deposition" and "mechanical segregation" required to explain all the preformed solids results.
July 1957	A hydroclone only removes small amounts of the circulating solids. "Deposition rate" is smaller after multi-additions of ZrO_2 than for initial addition.
August 1957	Loop concentrations and hydroclone pot concentrations increase together as material is added continuously to loop B ₂ .
September 1957	Review of results indicated that numbers cannot be predicted with confidence for HRT-CPP performance. Further studies with observation of solids behavior as a main objective are not justified.

None of the experimental facilities used give reactor values for all variables (Table 10). Three important variables which were only partially duplicated were the system geometry, including flow velocities and surface to volume ratios, the system temperature, and the difference between fluid and wall temperatures. The HRT Mock-up was a good approximation of the HRT temperature and geometry, but had hot walls due to the heat inputs required. Loop B₂ minimized the hot wall effects (no hot walls for up to 220°C) and approached the HRT surface to volume ratio. Loop A had the advantages of a small volume and excellent temperature control obtained by using large wall to fluid temperature differences. A glass loop provided visual observation, a small volume, easily controlled wall to fluid temperature differences, and a desirable geometry with temperatures limited to below 100°C. The HRT-CPP experiments provided results for its particular geometry. Loop B, loop T-1, loop P-1, and the hydroclone underflow pot test system were designed for special purposes and contributed little to solids behavior results.

The adequacy of operating procedures was determined by the ability to give known starting conditions, to obtain representative samples, and to obtain consistent analytical results. Circulation through bombs was the only satisfactory method of obtaining samples or of adding insoluble material to loops at high temperature and pressure. Samples taken by various procedures other than circulation through bombs were frequently found to be completely and unpredictably unrepresentative. Many conventional analytical techniques, including gravimetric, are not practical for the 1 to 1000 ppm range of concentrations of insoluble materials. Tracer and light transmission analyses were very useful, but activity and size distribution determinations respectively were required along with independent check methods of analysis for comparison purposes.

Table 10 Comparison of Experimental Facilities

Property Compared	Units	Value of Property for:							
		HRT	HRT - Mock-Up	Loop A	Loop B	Loop B-2	HRT-CPP Test	Glass Loop	Loop P-1
Liquid Charge	Kg H ₂ O	410	75	1.1	20	10	31	0.65	11
Surface	ft ²	600	52	3.3	~45	7.2	16.7	1.70	10.4
Volume/Surface	ft ³ /ft ²	.03	.07	.015	.02	.06	.08	.013	.05
Maximum Temperature	°C	300	300	320	300	300	280	90	270
Typical Flow* Velocities	ft/sec.	11-19	10-20	3-5	3-5	5-22	6-20	3-6	6
Relative Magnitude of Hot Walls	---	Negligible	Moderate	Large	Very Large	Small	Large	Small	None

*An average velocity for the major holdups in the system.

The natural fission and corrosion products in a reactor would be very complex mixtures; therefore single compounds or simple mixture were substituted to simplify the chemical analyses and the interpretation of results. The $\text{Nd}_2(\text{SO}_4)_3$ and the mixed rare earth sulfates appeared to adequately represent the sulfate salts soluble at room temperature and relatively insoluble at 300°C . Both zirconium sulfate and mixed zirconium-iron-chromium sulfate solutions were hydrolyzed in loops to precipitate oxides. This procedure is not a valid way of obtaining results representative of corrosion products. The hydrolysis gives an adherent scale on the walls which does not give circulating solids as expected from corrosion under radiation.

Most of the solids behavior studies were made with preformed oxides which may or may not have been representative substitutes for reactor corrosion products. The preformed oxides were prepared by precipitation with ammonium hydroxide from zirconium or mixed zirconium-iron-chromium sulfate solutions at atmospheric pressure. The hydrated oxides were digested, filtered, washed, slurried up in $.04 \text{ M } \text{UO}_2\text{SO}_4$, and digested in an autoclave at 300°C with O_2 overpressure. This gives particles with mean diameters (based on wt %) of 0.05 to 1 micron. Some corrosion loops have yielded scale like material with much larger particle diameters. Some laboratory bomb studies have shown marked differences in particle characteristics above 260°C depending on the composition. (23)

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