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PACIFIC NORTHWEST LABORATORY QUARTERLY REPORT ON SPACE NUCLEAR SYSTEMS PROGRAM FOR JULY-SEPTEMBER 1974

Battelle
Pacific Northwest Laboratories
Richland, Washington 99352

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NOTICE

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QUARTERLY REPORT
ON SPACE NUCLEAR SYSTEMS PROGRAM
FOR JULY-SEPTEMBER 1974

by

H. T. Fullam

October 1974

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

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QUARTERLY REPORT
ON SPACE NUCLEAR SYSTEMS PROGRAM
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STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM

At Hanford, strontium will be separated from the high-level waste, converted to the fluoride, and doubly encapsulated in small, high-integrity containers for subsequent long-term storage. The fluoride conversion, encapsulation and storage will take place in the Waste Encapsulation and Storage Facilities (WESF). The encapsulated strontium fluoride represents an economical source of ^{90}Sr *if the WESF capsule can be licensed for heat source applications under anticipated use conditions. The objectives of this program are to obtain the data needed to license* $^{90}SrF_2$ *heat sources and specifically the WESF* $^{90}SrF_2$ *capsules. The information needed for licensing can be divided into three general areas:*

1. *Long-term* SrF_2 *compatibility data.*
2. *Chemical and physical property data on* $^{90}SrF_2$.
3. *Capsule property data such as external corrosion resistance, crush strength, etc.*

The current program is designed to provide the required information.

SHORT-TERM SrF_2 COMPATIBILITY TESTS

The short-term compatibility tests consist of four series of tests using nine different containment materials and four fluoride fuel compositions:

1. High purity SrF_2 (>99.9%),
2. High purity $^{90}SrF_2$,
3. WESF grade $^{90}SrF_2$, and
4. High purity SrF_2 (>99.9%) containing controlled levels of added impurities.

The first three fuel compositions were tested at 800, 1000 and 1100°C while the last composition was tested at 1000°C. The tests lasted 1500 and 4400 hours.

The tests with the high purity SrF_2 have been completed and the couples analyzed; the results were presented in an earlier report. The high purity $^{90}SrF_2$ and WESF grade $^{90}SrF_2$ tests have been completed and the couples are now being analyzed.

Analysis of the $^{90}SrF_2$ couples is being conducted at the Postirradiation Testing Facility operated by HEDL. They have encountered severe problems in processing the couple test specimens because of ^{90}Sr contamination of the work area. These problems are much more severe than were encountered in processing the $^{90}SrF_2$ couples from the WESF compatibility program. As a result of the contamination problems, processing of the couples is slow and only two or three can be processed per week. To date, a total of 20 $^{90}SrF_2$ couples have been analyzed. Accordingly, arrangements have been made to ship a large fraction of the test specimens offsite for analysis early in October. Approximately 50 specimens will be shipped to ORNL and six to Battelle-Columbus for analysis. By shipping some specimens offsite, analysis of the $^{90}SrF_2$ couples should be completed before January.

The 1500 hour - 1000°C tests with high purity SrF_2 containing controlled levels of added impurities have been completed and the 4400-hour tests will be completed in October. The fuel compositions used in the tests are given in Table 1. Microscopic examination of the 1500-hour couples has been completed and photomicrographs have been obtained. The couples are now being analyzed by electron microprobe and scanning electron microscope. Preliminary estimates of metal attack observed in the 1500-hour tests, based on the photomicrographs, are given in Table 2. A complete evaluation of metal attack will be given when analysis of the couples is completed.

On the day the 1500-hour tests were scheduled to be stopped, a furnace controller failure allowed a fraction of the couples to overheat to about 1200°C for a short period of time (less than 4 hours); the overheated couples are indicated by an asterisk in Table 2, none of the 4400-hr test couples were affected. Results of these tests will be needed to determine the overall effect of this overheating.

TABLE 1. Compositions of SrF_2-Impurities Mixtures Used in Short-Term Compatibility Tests

Mixture	wt%
1. Sr^{+2} (as SrF_2)	95 wt%
Na^{+} (as NaF)	5 wt%
2. Sr^{+2} (as SrF_2)	95 wt%
Ca^{+2} (as CaF_2)	1.67 wt%
Mg^{+2} (as MgF_2)	1.67 wt%
Ba^{+2} (as BaF_2)	1.66 wt%
3. Sr^{+2} (as SrF_2)	98 wt%
Mn^{+2} (as MnF_2)	2 wt%
4. Sr^{+2} (as SrF_2)	98 wt%
Pb^{+2} (as PbF_2)	2 wt%
5. Sr^{+2} (as SrF_2)	98 wt%
Cu^{+2} (as CuF_2)	2 wt%
6. Sr^{+2} (as SrF_2)	98 wt%
Fe^{+3} (as FeF_3)	2 wt%
7. Sr^{+2} (as SrF_2)	98 wt%
Al^{+3} (as AlF_3)	2 wt%
8. Sr^{+2} (as SrF_2)	99 wt%
O^{-2} (as SrO)	1 wt%
9. Sr^{+2} (as SrF_2)	99.95 wt%
No_3^- (as $Sr(NO_3)_2$)	0.05 wt%
10. SrF_2	99.95 wt%
H_2O	0.05 wt%

TABLE 2. Preliminary Estimate of Metal Attack Observed in Couples Containing High-Purity SrF_2 with Controlled Levels of Added Impurities (After 1500 Hours at 1000°C)

Metal	Impurity Added	Depth of Metal Affected** (mils)
TZM*	NaF	<< 1
TZM*	MnF_2	<< 1
TZM	PbF_2	< 1
TZM*	CuF_2	1-2
TZM*	FeF_3	1-2
TZM*	AlF_3	<< 1
TZM	(Ca-Ba-Mg) F_2	<< 1
TZM	SrO	< 1
TZM*	$Sr(NO_3)_2$	<< 1
TZM	H_2O	< 1
W	NaF	<< 1
W*	MnF_2	< 1
W*	PbF_2	0
W	CuF_2	1-2
W	FeF_3	<< 1
W*	AlF_3	0
W	(Ca-Ba-Mg) F_2	<< 1
W	SrO	< 1
W*	$Sr(NO_3)_2$	0
W*	H_2O	< 1
Hastelloy C-276*	NaF	2-3
Hastelloy C-276	MnF_2	2-3
Hastelloy C-276*	PbF_2	1-2
Hastelloy C-276	CuF_2	6-8
Hastelloy C-276	FeF_3	2-3
Hastelloy C-276*	AlF_3	1-2
Hastelloy C-276	(Ca-Ba-Mg) F_2	2-3
Hastelloy C-276	SrO	10-12

TABLE 2. (contd)

Metal	Impurity Added	Depth of Metal Affected** (mils)
Hastelloy C-276*	$Sr(NO_3)_2$	3-4
Hastelloy C-276*	H_2O	4-6
Hastelloy N	NaF	3-4
Hastelloy N	MnF_2	2
Hastelloy N	PbF_2	2-3
Hastelloy N	CuF_2	4-5
Hastelloy N	FeF_3	10-12
Hastelloy N	AlF_3	2-3
Hastelloy N	$(Ca\text{-}Ba\text{-}Mg)\ F_2$	2-3
Hastelloy N	SrO	20-25
Hastelloy N*	$Sr(NO_3)_2$	2-3
Hastelloy N*	H_2O	1-2
Hastelloy X	NaF	2-3
Hastelloy X	MnF_2	3-4
Hastelloy X*	PbF_2	2-3
Hastelloy X*	CuF_2	20-30
Hastelloy X	FeF_3	5-6
Hastelloy X	AlF_3	3-4
Hastelloy X*	$(Ca\text{-}Ba\text{-}Mg)\ F_2$	2-3
Hastelloy X*	SrO	12-15
Hastelloy X*	$Sr(NO_3)_2$	4-5
Hastelloy X*	H_2O	3-4
Haynes 25	NaF	2-3
Haynes 25*	MnF_2	2-3
Haynes 25	PbF_2	3-4
Haynes 25*	CuF_2	10-12
Haynes 25*	FeF_3	8-10
Haynes 25	AlF_3	2-3
Haynes 25	$(Ca\text{-}Ba\text{-}Mg)\ F_2$	1-2
Haynes 25	SrO	4-5
Haynes 25*	$Sr(NO_3)_2$	4-5
Haynes 25	H_2O	< 1

TABLE 2. (contd)

Metal	Impurity Added	Depth of Metal Affected** (mils)
Haynes 188	NaF	< 1
Haynes 188*	MnF_2	4-6
Haynes 188	PbF_2	4-6
Haynes 188*	CuF_2	10-12
Haynes 188	FeF_3	10-12
Haynes 188	AlF_3	< 1
Haynes 188	(Ca-Ba-Mg) F_2	< 1
Haynes 188	SrO	10-12
Haynes 188*	$Sr(NO_3)_2$	3-4
Haynes 188*	H_2O	3-4
Inconel 600*	NaF	1
Inconel 600	MnF_2	2-5
Inconel 600	PbF_2	2-3
Inconel 600	CuF_2	2-3
Inconel 600*	FeF_3	4-6
Inconel 600	AlF_3	< 1
Inconel 600	(Ca-Ba-Mg) F_2	2-3
Inconel 600	SrO	15-20
Inconel 600*	$Sr(NO_3)_2$	2-3
Inconel 600*	H_2O	1
Inconel 625	NaF	2-3
Inconel 625	MnF_2	6-8
Inconel 625*	PbF_2	3-4
Inconel 625*	CuF_2	NA***
Inconel 625*	FeF_3	NA
Inconel 625	AlF_3	2-3
Inconel 625*	(Ca-Ba-Mg) F_2	NA
Inconel 625*	SrO	8-10
Inconel 625*	$Sr(NO_3)_2$	NA
Inconel 625	H_2O	2-3

*Indicates couples which were overheated to 1200°C for a few hours.
**Based on photomicrographs.
***NA-not available.

The furnace excursion occurred due to failure of two separate systems in the furnace controller. The controller SCR power module failed, allowing continuous power input and at the same time, the overtemperature control switch failed. To prevent similar incidents with the other furnaces being used in the compatibility testing program, a second overtemperature switch is being installed in each furnace system. The switches will operate off the temperature recorder used to monitor each furnace temperature and will be separate from the furnace controller. The switches will be set up to shut off power to the furnace if the temperature exceeds a predetermined level.

Current planning calls for the analyses of all the short-term compatibility test couples to be completed before January 1975. This will allow selection of the three containment materials to be used in the long-term compatibility tests by January and the start of long-term testing in March.

DISSOLUTION RATE OF $^{90}SrF_2$

A principal factor to be considered in the safety analysis of $^{90}SrF_2$ heat sources is the solubility and dissolution rate of $^{90}SrF_2$ in aqueous media. A major activity of the Strontium Heat Source Development Program in FY-1975 will be to measure the dissolution rate and solubility of WESF $^{90}SrF_2$ in aqueous media.

The objectives of the $^{90}SrF_2$ dissolution rates studies will be to obtain sufficient dissolution data that the regulatory agencies can accurately access the hazards involved in using a $^{90}SrF_2$ source. A secondary objective will be to provide sufficient data to permit a valid comparison between $^{90}SrF_2$, $^{90}SrTiO_3$ and $^{90}Sr_2TiO_4$ dissolution rates and solubilities.

The problem of ^{90}Sr compound dissolution in aqueous solutions is extremely complex. A number of variables can affect the dissolution rates including:

1. Compound surface area,
2. Compound bulk geometry,

3. Types and amounts of impurities present in the compound,
4. Thermal history of the compound, and
5. Temperature of dissolution media.

Because of the numerous variables involved, it is difficult to compare rate data for the different ^{90}Sr compounds and to compare rate data obtained by different investigators. When comparisons are made, care must be taken to see that the compounds being compared are adequately described with regard to composition, surface area, etc.

To demonstrate the effects of major variables on the SrF_2 dissolution rates, a number of SrF_2 compositions, prepared by various methods, will be tested.

To demonstrate the experimental techniques to be used, the dissolution rate of nonradioactive SrF_2 is being determined before work on WESF $^{90}SrF_2$ is started. Two different procedures have been considered for making the required measurements.

1. A static (nonflow) system in which SrF_2 is contacted with a known volume of solution and the increase in strontium concentration is followed as a function of time.
2. A dynamic (flow) system in which solution is pumped at a known rate into a reaction vessel containing SrF_2, and the overflow from the vessel is analyzed for its dissolved strontium content.

The nonradioactive strontium in solution is determined by flame photometry. The analytical procedure has a precision of approximately ±5% of the strontium present. An attempt was made to use ^{85}Sr tracer and gamma energy analysis to determine dissolved strontium, but the method was dropped because the counting rate was too low to provide a reliable measure of the strontium concentration. The ^{90}Sr in solution will be determined by beta counting (after aging to allow the ^{90}Y to reach equilibrium) and possibly by atomic adsorption using a flameless atomic adsorption unit.

It was found that the static system could not be used to obtain a reliable measure of the dissolution rate of SrF_2. As the strontium concentration in solution increased, the dissolution rate decreased. At the high strontium concentrations the variance of the analytical procedure exceeded the periodic increase in the strontium concentration. This made the calculation of a differential dissolution rate unreliable.

Work with the static system was dropped in favor of the dynamic (flow) system. There are several advantages to using a flow system for measuring dissolution rates as compared to a static system. The dynamic system more closely simulates an accident condition. If a $^{90}SrF_2$ source is breached in a large body of water, such as the ocean, the water currents would disperse the dissolved strontium and prevent it from approaching the equilibrium concentration (which occurs with the static system). The dynamic system measures the maximum dissolution rate. In the dynamic system the driving force for strontium dissolution is greater because the average concentration of strontium in solution is lower than is the case with the static system. With the static system the dissolution rate continually decreases with time as the strontium concentration of the solution increases. With the dynamic system, the dissolution rate is relatively constant (after the first few hours). A sketch of the equipment used in the dynamic dissolution rate measurements is shown in Figure 1. The SrF_2 to be tested is placed in a glass tube and held in place with glass wool plugs. The test solution is flowed into the tube at a controlled rate using a syringe pump or gravity flow. The temperature of the solution in the tube is measured with a thermometer. The solution overflowing the tube is sampled periodically, filtered through a 0.05-micron Millepore filter and analyzed for strontium and fluorine. In the tests with nonradioactive SrF_2, the strontium is determined by flame photometry and the fluorine by a fluoride specific ion electrode.

Several different grades of strontium fluoride, prepared by different methods, have been tested. The types of SrF_2 used, how the samples were prepared, and their surface areas are described in Table 3. For comparison purposes samples of $SrTiO_3$ and Sr_2TiO_4 were also tested. These materials are also described in Table 3.

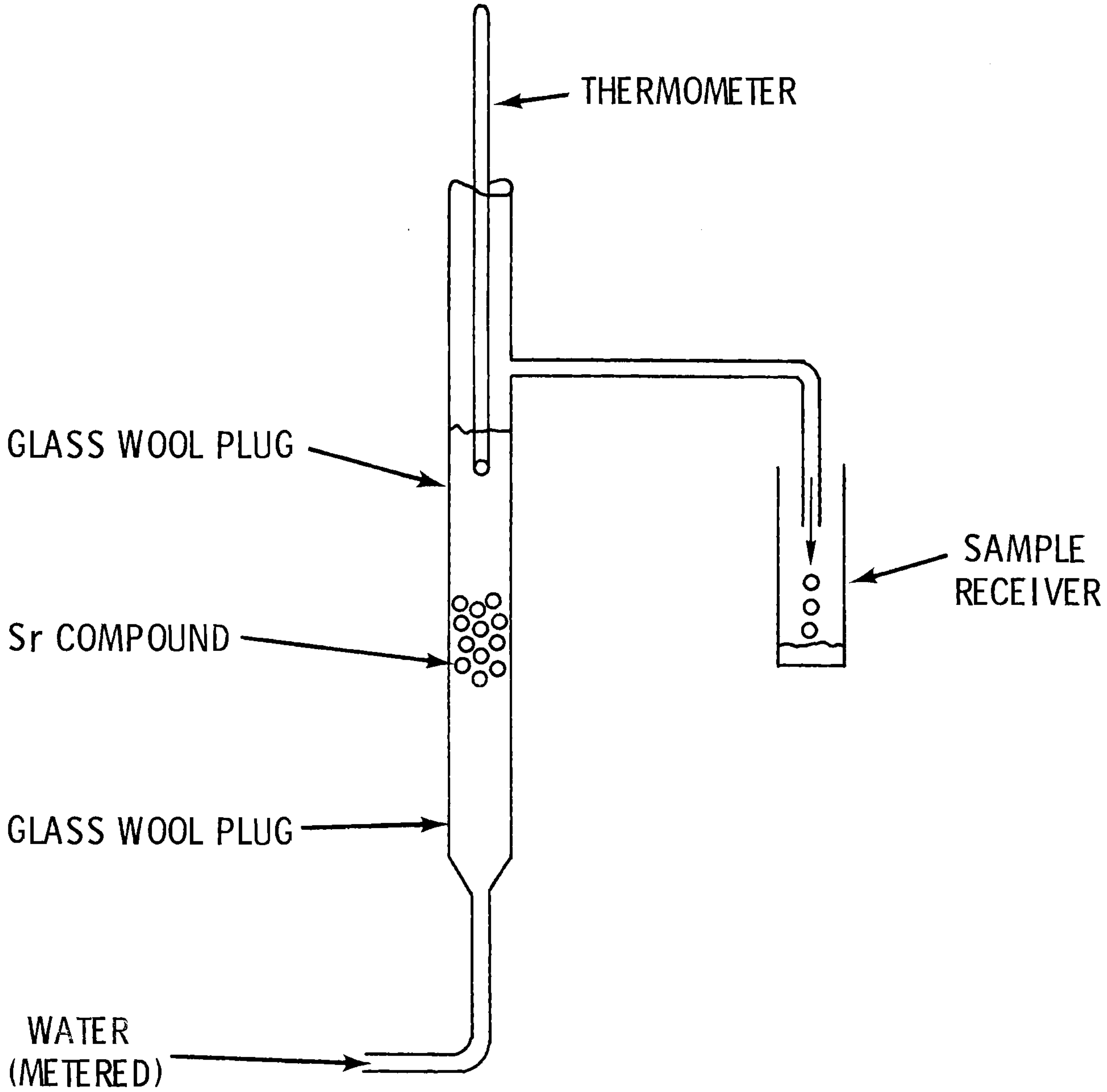

FIGURE 1. Dynamic System Used to Measure Dissolution Rate of Strontium Compounds

TABLE 3. Compounds Evaluated in Dissolution Tests

Identification Number	Description of Material	Surface Area (cm^2/gram of compound)
SR-1A	SrF_2 powder containing 0.5% impurities; fired at 1100°C for 20 hours; crushed to - 1/4 in. + 1/8 in. lumps.	370
SR-1B	SrF_2 powder containing 0.5% impurities; fired at 900°C for 20 hours; crushed to - 1/4 in. + 1/8 in. lumps.	580
SR-2	Commercial SrF_2 powder (99.5%); fired at 1100°C for 48 hours; crushed to - 1/4 in. + 1/8 in. lumps.	250
SR-3A	High purity SrF_2 powder (>99.9%); fired at 1100°C for 20 hours; crushed to - 1/4 in. + 1/8 in. lumps.	510
SR-3B	High purity SrF_2 powder (>99.9%); fused at 1500°C; crushed to - 1/4 in. + 1/8 in. lumps.	620
SR-3C	High purity SrF_2 powder (>99.9%); pressed into pellets at 70% TD; fired at 1100°C for 4 hours; crushed to - 1/4 in. + 1/8 in. lumps.	910
SR-3D	High purity SrF_2 powder (>99.9%); pressed to pellets at 70% TD; fired at 1100°C for 20 hours; crushed to - 1/4 in. + 1/8 in. lumps.	390
SR-3E	High purity SrF_2 powder (>99.9%) + 10 wt% NaF; fired at 1100°C for 20 hours; crushed to - 1/4 in. + 1/8 in. lumps.	520
SR-4	WESF grade SrF_2 powder; fired at 1100°C for 20 hours; crushed to - 1/4 in + 1/8 in. lumps.	660
SR-5	$SrTiO_3$ powder fired at 1450°C for 4 hours; crushed to - 1/4 in. + 1/8 in. lumps.	15,000
SR-6	Sr_2TiO_4 powder fired at 1450°C for 4 hours; crushed to - 1/4 in. + 1/8 in. lumps.	12,000

Four different solutions are being used in the dissolution tests: demineralized water having a resistivity of 2 megohms, plant tap water, synthetic seawater(a) containing 13.9 mg/ℓ strontium, and natural seawater taken from the Strait of Juan de Fuca at Sequim, Washington. All grades of SrF_2 and the two titanates will be tested with each solution.

With SrF_2 the dissolution rate is high initially, but decreases with time until an equilibrium level is reached. The time required to reach equilibrium varied somewhat between samples, but the equilibrium level was normally attained within 24 hours. Figure 2 shows data obtained with commercial SrF_2, which had been fired at 1100°C for 48 hours and crushed to 1/4 in. + 1/8 in. lumps (see Table 3 - SR-2), and demineralized water. In the case of the titanates the dissolution rate decreased continually with time, and did not reach an equilibrium level even though the runs continued for several days.

Dissolution rate data obtained with nonradioactive SrF_2, to date, are presented in Table 4 where the values, which are given, represent the equilibrium dissolution rates. Dissolution rate data for the titanates are summarized in Figures 3 and 4.

The rate data is presented in two forms:

1. as micrograms of strontium dissolved per hour per gram of strontium being tested, and
2. as micrograms of strontium dissolved per hour per square centimeter of compound surface.

The two sets of units were used to allow a more valid comparison of the dissolution rates of the SrF_2 and titanates.

Strontium fluoride has a relatively low melting point (∿1450°C) compared to the strontium titanates. Sintering of the SrF_2 normally occurs in the temperature regime where a $^{90}SrF_2$ source is expected to operate (800 to 1000°C). As a result the surface area of the $^{90}SrF_2$ in a source

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(a) Composition taken from T. P. May and A. T. Alexander, Proc. ASTM, vol. 50, pp. 1131-41, 1950.

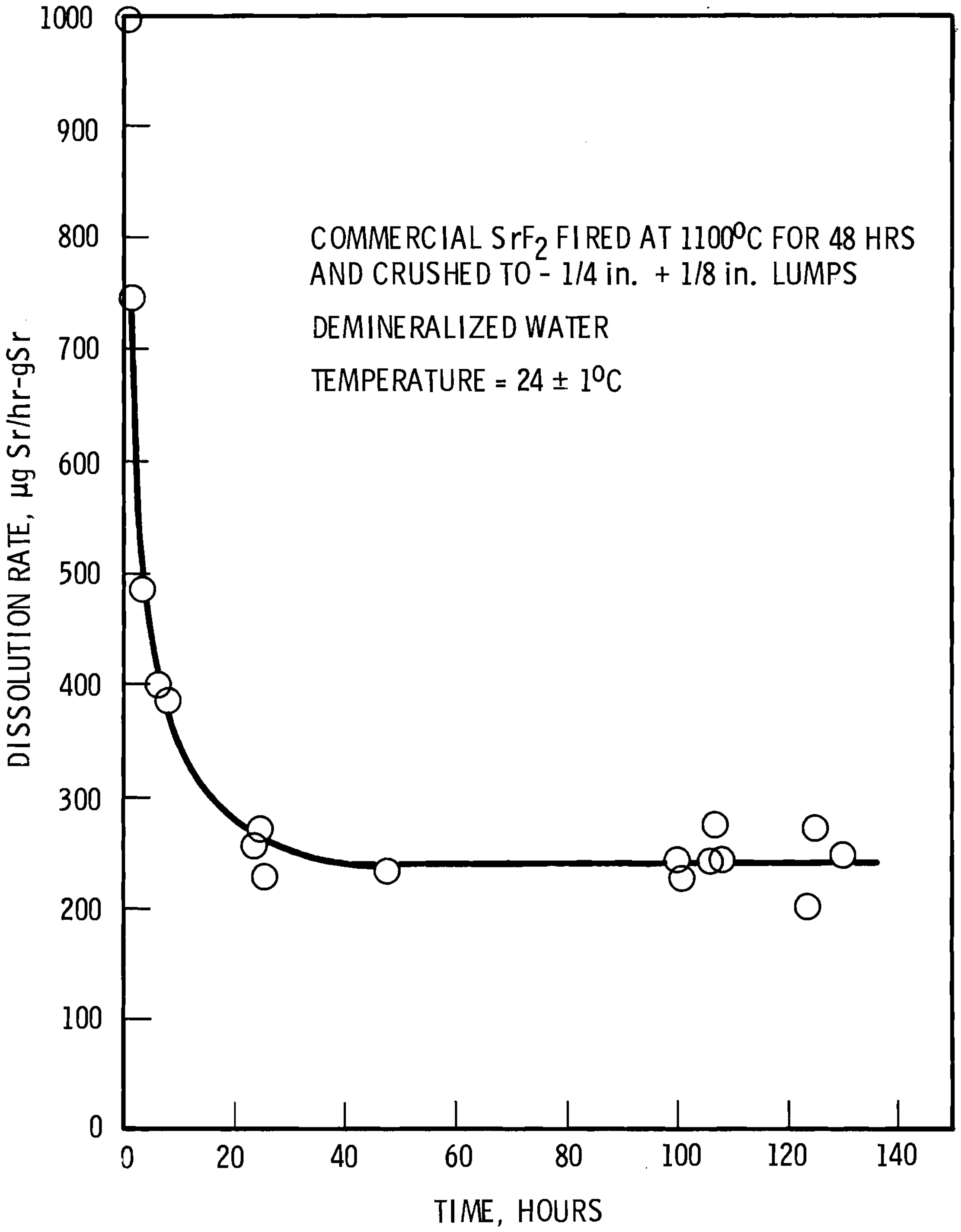

FIGURE 2. Dissolution Rate of Commercial SrF_2 in a Dynamic System

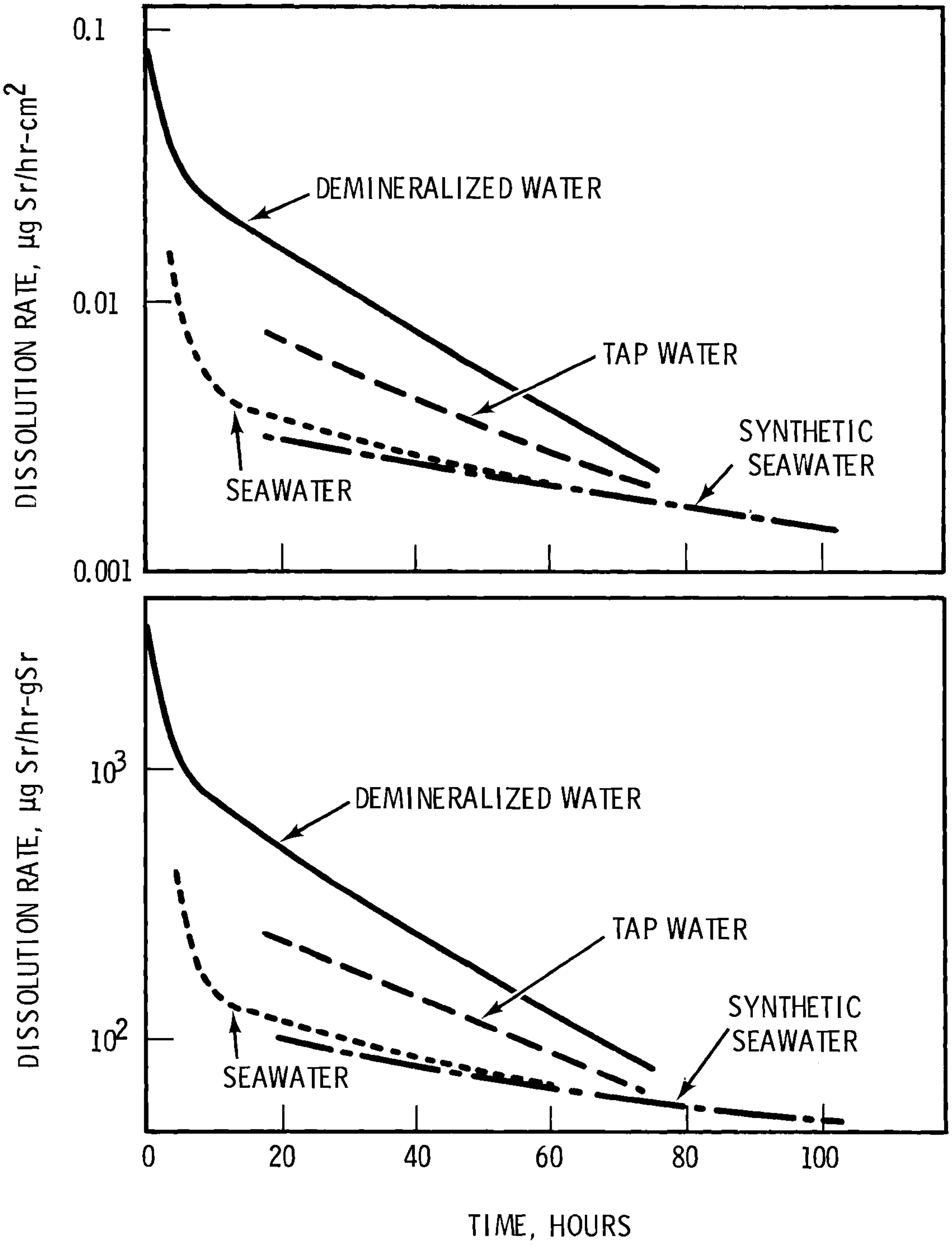

FIGURE 3. Dissolution Rate of $SrTiO_3$ at 24°C in a Dynamic System

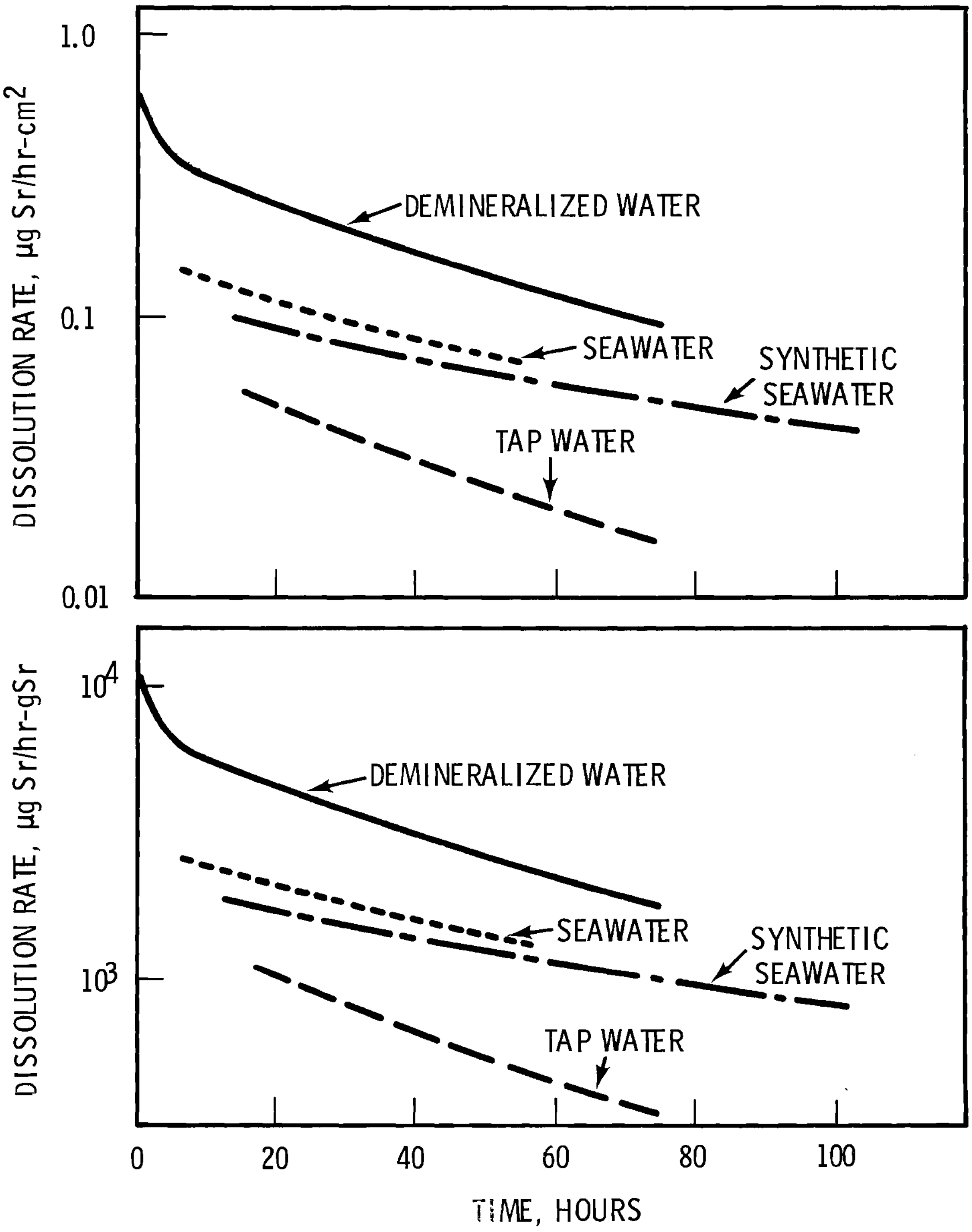

FIGURE 4. Dissolution Rate of Sr_2TiO_4 at 24°C in a Dynamic System

TABLE 4. Equilibrium Dissolution Rate[(a)] of SrF_2 in Aqueous Media at 24°C

SrF_2 Tested	Dissolution Rate: µg Sr Dissolved/hr - g Sr				Dissolution Rate: µg Sr Dissolved/hr - cm^2			
	Demineral Water	Tap Water	Synthetic Seawater	Natural Seawater	Demineral Water	Tap Water	Synthetic Seawater	Natural Seawater
SR-1A	620 ± 40	680 ± 80		110 ± 16	1.2 ±0.07	1.3 ± 0.15		0.21 ± 0.03
SR-1B	980 ±120	1160 ±240		350 ±240	1.2 ±0.15	1.4 ± 0.28		0.42 ± 0.02
SR-2	235 ± 17	330 ±200	220 ± 27 200 ± 42	150 ± 47	0.64±0.05	0.92± 0.56	0.61 ±0.08 0.56 ±0.12	0.48 ± 0.13
SR-3A	880 ±190				1.20±0.26			
SR-3B	170 ± 30	220 ± 30	90 ± 22	83 ± 4	0.19±0.03	0.24 ±0.03	0.10 ±0.02	0.094 ±0.005
SR-3C	350 ± 40	240 ± 26	130 ± 40	120 ± 17 100 ± 15	0.27±0.03	0.18 ±0.02	0.080±0.03	0.092 ±0.01 0.080 ±0.01
SR-3D				52 ± 30				0.092 ±0.05
SR-3E	1200 ±140				1.5 ±0.19			
SR-4	140 ± 24	170 ± 29	76 ± 27	50 ± 14	0.15±0.03	0.18 ±0.03	0.080±0.03	0.053 ±0.01

(a) Obtained using a dynamic (flow) system.

will be reduced significantly with time and temperature as sintering occurs. Also, impurities in the $^{90}SrF_2$ (such as NaF and decay products) will affect its sintering characteristics and surface area. These factors make it difficult, if not impossible, to accurately define the surface area of $^{90}SrF_2$ in an operating surface. The initial surface areas can be approximated but the change in surface with time and temperature is difficult to predict. This is not the case with titanate sources because no appreciable sintering or change in surface area occurs below 1100°C.

Comparison of the dissolution rates of SrF_2 and the titanates is complicated by the fact that the surface areas of the two titanates are much greater than those of the fluoride samples tested. If the comparisons are based on compound surface areas, the dissolution rates are generally higher for the fluorides than for either titanates. However, if the comparison is based on the amount of contained strontium the dissolution rates for the fluorides are lower than that of Sr_2TiO_4 and about the same as that of the $SrTiO_3$. This can be seen by referring to Figure 5 where the seawater dissolution rates of the two titanates and commercial SrF_2 are compared. Similar comparisons can be made with the other fluoride samples.

In an operating ^{90}Sr source of a given heat output, the number of grams of ^{90}Sr required will be the same regardless of the strontium compound used. Therefore, in considering source safety, it appears reasonable to base the comparison of fluoride - titanate dissolution rates on the contained strontium. This being the case, based on PNL measurements, the fluoride is less hazardous than the Sr_2TiO_4 and about equivalent to the $SrTiO_3$.

The Martin Company(1) has made some dissolution rate measurements on nonradioactive SrF_2, $SrTiO_3$ and Sr_2TiO_4 using both dynamic and static systems. A comparison of the PNL and Martin data obtained with a dynamic system is given in Table 5. The SrF_2 dissolution rates given are the equilibrium values. The titanate dissolution rates given are the nonequilibrium rates after 20 hours exposure.

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(1) J. H. Gray, "Solubility Study of Strontium Fuel Compounds," MND-3062-29, Martin Company, Baltimore, MD, 1966.

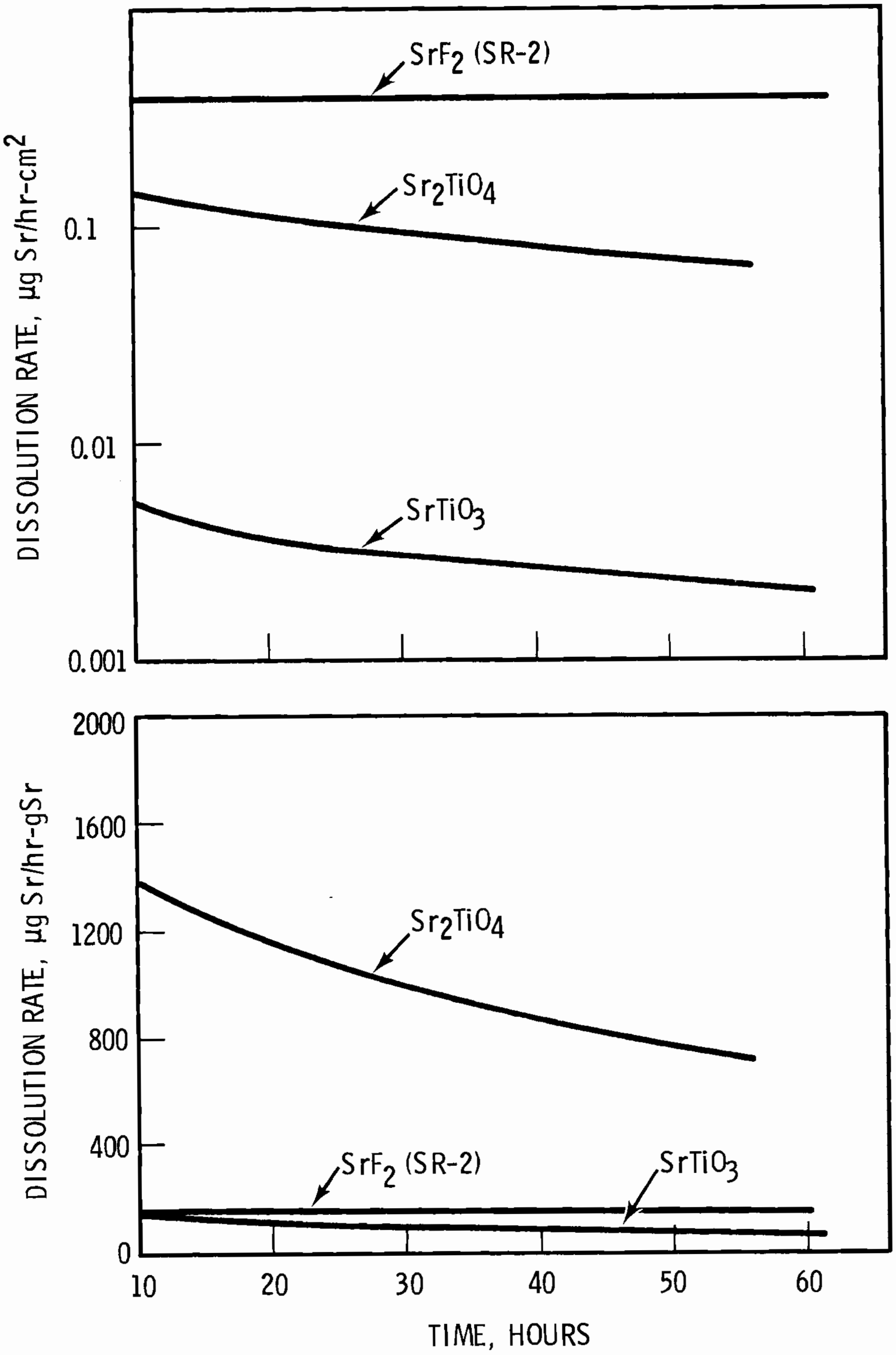

FIGURE 5. Comparison of the Dissolution Rates for Sr_2TiO_3, Sr_2TiO_4 and Commercial SrF_2 in Seawater

TABLE 5. Comparison of PNL and Martin Company Dissolution Rate Data(a)

	Dissolution Rate(e)							
	µg Sr/hr - g Sr				µg Sr/hr - cm^2			
	Demineralized Water		Seawater		Demineralized Water		Seawater	
Material	PNL	Martin	PNL	Martin	PNL	Martin	PNL	Martin
SrF_2(b)	120	340	50	560	0.15	3.7	0.05	6
$SrTiO_3$(c)	500	19	120	9	0.015	0.3	0.0037	0.15
Sr_2TiO_4(d)	4900	1170	2300	500	0.25	18	0.12	7.8

(a) At ∿24°C.

(b) The PNL SrF_2 was WESF grade (SR-4, see Table 3). The Martin SrF_2 was prepared by a flowsheet similar to the WESF flowsheet. The SrF_2 was fired at 1200°C for 7 hours and ground. The -60 + 80-mesh fraction was used in the tests. The material had a surface area of 93 cm^2/g.

(c) The Martin $SrTiO_3$ had a surface area 62 cm^2/g. The material was fired at 1400°C for 12 hours and ground. The -60 + 80-mesh fraction was used in the tests.

(d) The Martin Sr_2TiO_4 had a surface area of 65 cm^2/g. The material was fired at 1400°C for 12 hours and ground. The -60 + 80-mesh fraction was used in the tests.

(e) The SrF_2 dissolution rates are the equilibrium rates, and the titanates dissolution rates are the differential rates after 20-hours exposure. All rates were obtained with a dynamic (flow) system.

A direct comparison of the PNL and Martin data is difficult to make because of the differences in surface area of the various materials. The PNL prepared titanates have a much greater surface area than the Martin prepared compounds. On a per gram of strontium basis, the PNL titanates have a much greater dissolution rate than the Martin compounds, but much lower dissolution rates on a surface area basis. The PNL SrF_2 has a lower dissolution rate than the Martin fluoride on a per gram of strontium basis and a much lower dissolution rate on a surface area basis. From the above comparisons, it can be seen that the compounds being compared must be adequately described in order to make the comparison have meaning.

STRONTIUM FLUORIDE PROPERTY MEASUREMENTS

Work is underway to determine the thermal diffusivity, heat capacity and thermal expansion of SrF_2. The thermal conductivity of the fluoride will be calculated from the thermal diffusivity and heat capacity data. Since the radioactivity associated with the $^{90}SrF_2$ has no significant effect on these properties, the measurements are being made using non-radioactive SrF_2 by J. L. Bates of the Ceramic and Graphite Section.

Three grades of SrF_2 are being evaluated:

1. High purity SrF_2 containing less than 0.1 wt% total impurities.
2. Commercial SrF_2 containing 0.5 wt% total impurities.
3. WESF grade SrF_2 containing about 4 wt% impurities. The composition used was based on the analysis of $^{90}SrF_2$ prepared by PNL using ^{90}Sr solution obtained from ARHCO and the WESF flowsheet.

Each grade of fluoride was cold pressed into pellets and fired at 800°C in air. The diffusivity measurements require pellets approximately 3/8 in. in diameter by 0.05 in. thick. It was difficult to prepare stable pellets of the required dimensions without the use of a die lubricant. Satisfactory pellets were obtained using a stearic acid lubricant, which was removed from the pellet during the firing operation. Pellets of various densities were prepared by pressing the fluoride at different pressures. The thermal diffusivity measurements on the various pellets are now underway.

Thermal expansion measurements on the three fluorides are also underway using pellets 3/8 in. in diameter by about 3/4 in. long. The original plan was to determine the thermal expansion up to 1100°C. However, two problems were encountered which made it necessary to limit the measurements to about 900°C.

1. At temperatures above about 950°C a reaction occurs between the fluoride and the quartz portion of the dilatometer.
2. At about 900°C a softening of the fluoride pellet occurs which results in pellet shrinkage rather than expansion. This probably is due to a eutectic formation between the SrF_2 and any NaF present in the fluoride. Such a reaction is to be expected with the WESF grade SrF_2, but is surprising with the high purity SrF_2 which contains less than 1000 ppm total impurities.

The results of thermal expansion and thermal diffusivity studies will be reported when all the data are available.

Work has started on defining the SrF_2 - ZrF_4 phase diagrams. Samples of SrF_2 containing 1, 3, 5, 10, 15, 20, 30, 40 and 50 mol% ZrF_4 were sealed in individual quartz ampoules under vacuum. The samples were then heated to 800°C for 2 hours. Each sample is now being analyzed by X-ray diffraction. Initial differential thermal analysis studies in argon with the samples containing 5 mol% and 10 mol% ZrF_4 indicate that there are no molten phases present below 1150°C. The DTA also failed to show any volatilization of ZrF_4 which is surprising because ZrF_4 is reported to sublime above 600°C. This could indicate the formation of a high-melting - nonvolatile compound between the SrF_2 and ZrF_4.

PROGRAM WORK PLAN

The work plan for Strontium Heat Source Development Program has been completed. The work plan was submitted to SNS in September for review.

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