**PACIFIC NORTHWEST LABORATORY
MONTHLY REPORT ON
SPACE NUCLEAR SYSTEMS PROGRAM
FOR PERIOD ENDING JULY 1974**

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

**Prepared for the U.S. Atomic Energy
Commission under Contract AT(45-1):1830**

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy **Commission**, nor any of their employees, nor any of their contractors, subcontractors, or thier employees, makes any warranty, express or **implied**, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process-disclosed, or represents that its use would not infringe privately owned rights.

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
U.S. ATOMIC ENERGY COMMISSION
Under Contract AT(45-1)-1830

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$5.45; Microfische \$2.25

3 3679 00062 1856

BNWL-1845-2

PACIFIC NORTHWEST LABORATORY
MONTHLY REPORT
ON SPACE NUCLEAR SYSTEMS PROGRAM
FOR JULY 1974

by

H. T. Fullam

August 1974

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

TABLE OF CONTENTS

STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM	1
Strontium Fluoride Compatibility Tests	1
SrF ₂ Dissolution Rates in Aqueous Media	4
SrF ₂ Physical Property Measurements	7
WESF ⁹⁰ SrF ₂ Capsule Analysis	7
Thermodynamic Analysis of ⁹⁰ SrF ₂ Compatibility	8
SrF ₂ Compatibility Program Plan	8

PACIFIC NORTHWEST LABORATORY
MONTHLY REPORT
ON SPACE NUCLEAR SYSTEMS PROGRAM
FOR JULY 1974

STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM

At Hanford, strontium will be separated from the high-Zevel waste, then 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). This 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 $^{90}\text{SrF}_2$ heat sources and specifically the WESF $^{90}\text{SrF}_2$ 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 $^{90}\text{SrF}_2$.*
- 3. Capsule property data such as external corrosion resistance, crush strength, etc.*

The current program is designed to provide the required information.

STRONTIUM FLUORIDE COMPATIBILITY TESTS

The short-term compatibility tests with high-purity, nonradioactive SrF_2 , containing controlled levels of added impurities, are continuing as scheduled. The 1500-hour tests, conducted at 1000°C , have been completed and the couples are being evaluated; the 4400-hour 1000°C tests will be completed in October. Metallographic examination of a number of the 1500-hour couples has been completed and photomicrographs have been obtained. Preliminary estimates of the amount and types of metal attack observed, based on the photomicrographs, are presented in Table 1. A more detailed analysis of metal-fluoride interaction and correlation of impurity effects will be reported when all of the photomicrographs and electron microprobe (EMP) data are available.

TABLE 1. Preliminary Compatibility Data for Couples Containing High-Purity, Nonradioactive SrF_2 with Controlled Levels of Specific Impurities Added - After 1500 hr at 1000°C

Metal	Impurity Added	Depth of Metal** Affected, Mils	Remarks (types of attack observed)**
TZM*	Na	<< 1	Slight surface roughening-slight pitting.
TZW	Mn	<< 1	Slight surface roughening.
TZM	Pb	< 1	Pitting, intergranular attack.
TZW	Cu	1-2	Intergranular attack-subsurface void formation.
TZW	Fe	1-2	Heavy Pitting.
TZM*	Al	<< 1	Slight surface roughening.
TZM	Ca-Ba-Mg	<< 1	Slight surface roughening.
TZM	0-2	< 1	Surface roughening.
TZM*	NO ₃	<< 1	Slight surface roughening.
TZM	1-20	< 1	Surface roughening - slight pitting.
W	Na	<< 1	Slight surface roughening, metal sample badly fractured. (a)
W*	Mn	< 1	Slight layered surface attack.
W*	Pb	0	No visible attack, metal sample fractured.
W	Cu	1-2	Pitting, some surface attack.
W	Fe	<< 1	Slight surface roughening.
W*	Al	0	No visible attack, metal sample fractured.
W	Ca-Ba-Mg	<< 1	Slight surface roughening, metal sample badly fractured.
W	0-2	< 1	Layered surface attack, slight pitting, sample fractured.
W*	NO ₃	0	No visible attack, sample fractured.
W*	1-20	< 1	Slight pitting, samples fractured.
Hastelloy N*	Na	3-4	Layered surface attack, heavy subsurface void formation.
Hastelloy N	Mn	2	Layered surface attack, subsurface void formation.
Hastelloy N	Pb	2-3	Pitting, subsurface void formation.
Hastelloy N*	Cu	4-5	General surface attack, pitting, heavy subsurface void formation.
Hastelloy N*	Fe	10-12	Extremely heavy subsurface void formation.

* Indicates couples which were overheated to 1200°C for a few hours.

** Estimated from photomicrographs - more detailed data will be presented when electron microprobe results are available.

(a) Fracture of the tungsten specimens appeared to have occurred during couple fabrication rather than as the result of SrF_2 attack.

TABLE 1. (contd)

Metal	Impurity Added	Depth of Metal** Affected, MILS	Remarks (types of attack observed)**
Hastelloy N*	Al	2-3	Slight subsurface void formation.
Hastelloy N	Ca-Ba-Mg	2-3	Slight pitting, slight subsurface void formation.
Hastelloy N	O ₂	20-25	Extremely heavy intergranular attack and leaching of surface.
Hastelloy N*	NO ₂	---	Results not yet available.
Hastelloy N*	H ₂ O	1-2	Subsurface void formation.
Hast C-276*	Na	2-3	Some pitting, some subsurface void formation.
Hast C-276	Mn	2-3	Some intergranular attack and pitting.
Hast C-276*	Pb	1-2	Subsurface void formation and pitting.
Hast C-276	Cu	6-8	General surface attack, some deep subsurface void formation.
Hast C-276	Fe	---	Results not yet available.
Hast C-276*	Al	1-2	Subsurface void formation.
Hast C-276	Ca-Ba-Mg	2-3	General surface attack, some pitting and sub-surface voids.
Hast C-276	O ₂	10-12	Heavy intergranular attack and subsurface void formation.
Hast C-276*	NO ₂	3-4	General surface attack, some pitting and subsurface voids.
Hast C-276*	H ₂ O	4-6	Pitting, intergranular attack, some subsurface voids.
Inconel 600*	Na	1	General surface attack, some pitting.
Inconel 600	Mn	2-5	Some pitting, subsurface void formation.***
Inconel 600	Pb	2-3	General surface attack, subsurface void formation.***
Inconel 600*	Cu	2-3	Pitting, subsurface void formation.***
Inconel 600*	Fe	4-6	Heavy subsurface void formation.***
Inconel 600	Al	< 1	Slight pitting and surface roughening.
Inconel 600	Ca-Ba-Mg	2-3	Slight layered surface attack, some subsurface void formation.***
Inconel 600	O ₂	15-20	Heavy intergranular attack and surface leaching.
Inconel 600*	NO ₂	2-3	General surface attack, subsurface void formation.***
Inconel 600*	H ₂ O	1	Slight surface roughening, subsurface void formation.***
Haynes 188	Na	< 1	Slight surface roughening.
Haynes 188*	Mn	?	Need electron microprobe (EMP) data to evaluate attack.
Haynes 188	Pb	4-67	Some pitting, subsurface voids (need EMP data to evaluate attack).
Haynes 188*	Cu	10-12?	Subsurface void formation? (need EMP data to evaluate attack).
Haynes 188	Fe	10-12?	Subsurface void formation? (need EMP data to evaluate attack).
Haynes 188	Al	< 1	Slight surface roughening.
Haynes 188	Ca-Ba-Mg	< 1	Slight surface roughening.
Haynes 188	O ₂	10-12	Heavy intergranular attack.
Haynes 188*	NO ₂	3-41	Slight surface attack (need EMP data to evaluate subsurface voids).
Haynes 188*	H ₂ O	3-41	Slight surface attack (need EMP data to evaluate subsurface voids).

*** Indications of subsurface void formation in Inconel 600 are questionable, will be adequately determined by electron microprobe analyses.

In the 1500-hour tests, a furnace controller failure occurred the day the tests were to be stopped. As a result, one furnace (of six) overheated to about 1200°C when failure occurred and the couples in the furnace were above 1000°C for about 4 hours which could affect the results. Of a total 108-1500-hour couples, 42 were overheated. These affected couples are indicated by the footnotes in Table 1. Results of the 4400-hour tests will be needed to determine the overall effect of the overheating.

The overheating occurred due to failure of two separate systems in the temperature controller. The controller SRC failed, allowing continuous power input while, at the same time, an over-temperature control switch failed. To prevent a similar occurrence with the other furnaces, another over-temperature switch will be installed in each furnace system. These switches will operate off the thermocouples used to monitor each furnace temperature and will be set up to shut off power to the furnace if the temperature exceeds a predetermined level.

A total of 108 couples, containing high-purity $^{90}\text{SrF}_2$ or WESF grade $^{90}\text{SrF}_2$ (prepared by PNL), have been tested as part of the short-term compatibility test program and evaluation of these couples is now underway. To date, seven couples have been examined, 12 are at Postirradiation Testing (operated by HEDL) for metallographic examination, 20 are at PNL ready for examination, and 15 have been sectioned. The other couples are in the process of being sectioned.

SrF₂ DISSOLUTION RATES IN AQUEOUS MEDIA

The dissolution rates of several grades of nonradioactive SrF_2 in demineralized water were obtained using a dynamic (flow) test system. A sketch of the experimental setup is shown in Figure 1. In the tests, demineralized water was flowed at a constant rate into the reaction chamber containing the SrF_2 . The overflow from the reaction vessel was sampled periodically, filtered through an 0.05- μm Millipore filter, and analyzed for its strontium and fluoride content. In general, the strontium dissolution rate was high initially, then decreased with time until an equilibrium level was reached. The time required to reach equilibrium

FIGURE 1. Dynamic (Flow) System for SrF_2 Dissolution Rate Measurements

varied somewhat between samples, but normally the equilibrium level was attained within 24 hours. Results obtained with the various SrF_2 samples are summarized in Table 2. In two of the tests the water flow rate was varied periodically to determine the effect on the dissolution rate, but in each case it was found the dissolution rate was unaffected (within experimental error) by changes in water flow.

With SrF_2 , the water leaving the reaction vessel should contain two moles of fluoride per mole of strontium, which was the case when the system was at equilibrium. However, during the early stages of each run, when the dissolution rate was changing, the $\text{F}^-/\text{Sr}^{+2}$ was not 2. If the SrF_2 contained a substantial amount of NaF (see Run 4 in Table 2) the

TABLE 2. Dissolution Rates of Nonradioactive SrF_2 in Demineralized Water at 25°C - Obtained with a Dynamic (Flow) System

Run No,	Grade of SrF_2	Equil. Dissolution Rate ($\mu\text{g Sr/hr-g SrF}_2$)
1	High purity SrF_2 containing 0.5% impurities (including 0.4% Na) which was fired at 1100°C for 20 hours, cooled to room temperature and crushed to $-1/4''$ $+1/8''$ lumps.	435
2	Same as #1 except material was fired at 900°C for 20 hours.	640
3	Commercial SrF_2 (0.5% impurities) fired at 1100°C for 84 hours, cooled to room temperature and crushed to $-1/4''$ $+1/8''$ lumps.	160
4	High purity SrF_2 (99.9%) mixed with 10% NaF and fired at 1100°C for 20 hours, cooled and crushed to $-1/4''$ $+1/8''$ lumps.	640
5	High purity SrF_2 (99.9%) fused at 1500°C , cooled to room temperature and crushed to $-1/4''$ $+1/8''$ lumps.	120
6	High purity SrF_2 (99.9%) cold pressed into pellet (70% of TD), fired four hours at 1100°C , cooled and broken into $-1/4''$ $+1/8''$ lumps.	246
7	High purity SrF_2 (99.9%) fired at 1100°C for 20 hours, cooled and crushed to $-1/4''$ $+1/8''$ lumps.	Samples being analyzed

F^-/Sr^{+2} ratio of the solution was much greater than 2 during the initial phase of the run because of the rapid dissolution of the NaF. In addition, the initial dissolution rate of the SrF_2 was lower than usual. When the SrF_2 contained very little NaF (Runs 5 and 6 in Table 2), the F^-/Sr^{+2} ratio in the solution was less than 2 during the initial portion of the run and the initial dissolution rate was quite high. Preliminary indications are that this is due to the presence of SrO in the SrF_2 and the SrO dissolves quite rapidly.

SrF₂ PHYSICAL PROPERTY MEASUREMENTS

Efforts are underway to determine the thermal conductivity of SrF_2 by measuring the thermal diffusivity and heat capacity of SrF_2 , and calculating the thermal conductivity. (The experimental measurements will be made by J. L. Bates of the Ceramics and Graphite Section.) Three grades of SrF_2 will be studied: high purity SrF_2 (<0.1% impurities), commercial SrF_2 (0.5% impurities), and WESF grade SrF_2 (~4% impurities). We are currently in the process of preparing small cold-pressed pellets of various densities from the three grades of SrF_2 to be used in the tests. While SrF_2 is easily compacted to fairly high densities by cold pressing, it is difficult to obtain stable pellets in the dimensions needed (0.375-in. diameter x 0.05-in. thick) for the tests. Satisfactory pellets can be obtained using a die lubricant, but removal of the lubricant from the pellet is difficult. Ways to remove the lubricant are now being investigated.

WESF ⁹⁰SrF₂ CAPSULE ANALYSIS

A preliminary analysis of the WESF capsule design indicates it will probably fail to meet one or more of the IAEA Series 33 tests. This holds true whether the capsule material is Hastelloy C-276, Hastelloy X, Inconel 600 or Haynes 25. W. E. Anderson (Structures and Mechanics Section) studied the problem in detail and concluded that a more thorough study

involving fracture mechanics and experimental testing is required to adequately predict capsule integrity under IAEA Series 33 test conditions. The possibility of including such a study in the current program is being investigated.

THERMODYNAMIC ANALYSIS OF $^{90}\text{SrF}_2$ COMPATIBILITY

The computer program to be used in the thermodynamic analysis of $^{90}\text{SrF}_2$ compatibility is ready for use. The study will be made by R. P. Turcotte (and others) in the Ceramics and Graphite Section. Work on the program is underway at a limited rate for the present time, but the major effort will start in September.

SrF_2 COMPATIBILITY PROGRAM PLAN

Efforts are underway to develop the long-range program plan (through FY-78) that will be "finalized" as soon as the "Program Letter" is received from SNS. The principal effort, at the present time, is on the design of the long-term compatibility tests. The program plan should be ready for review by SNS by the latter part of August.

DISTRIBUTION

No. of
Copies

OFFSITE

1	<u>AEC Chicago Patent Attorney</u> A. A. Churm
1	<u>AEC Division of Applied Technology</u> E. E. Fowler J. E. Machurek R. W. Shivers
2	<u>AEC Division of Biomedical and Environmental Research</u> J. N. Maddox L. A. Miller
3	<u>AEC Division of Production and Materials Management</u> F. P. Baranowski R. K. Heusser J. W. Pollock
12	<u>AEC Division of Space Nuclear Systems</u> R. T. Carpenter G. P. Dix T. J. Dobry, Jr. D. S. Gabriel N. Goldenberg H. Jaffe W. K. Kern A. P. Litman (3) G. A. Newby B. J. Rock
2	<u>AEC Division of Waste Management and Transportation</u> F. K. Pittman R. W. Ramsey, Jr.
1	<u>AEC Oak Ridge Operations Office</u> D. C. Davis, Jr.
1	<u>AEC Savannah River Operations Office</u> T. B. Hindman

No. of
Copies

5	<u>AEC Technical Information Center</u>
1	<u>General Atomic Company (San Diego)</u> H. C. Carney
1	<u>General Electric Company MSVD, Valley Forge, PA</u> P. E. Brown
1	<u>General Electric Company, Vallecitos Laboratory</u> G. E. Robinson
2	<u>Los Alamos Scientific Laboratory, Los Alamos, NM</u> T. K. Keenan R. N. Mulford
1	<u>Mound Laboratory (AEC)</u> W. T. Cave
1	<u>Naval Nuclear Power Division (Ft. Belvoir, VA)</u> F. E. Roselle
3	<u>Naval Facilities Engineering Command</u> T. P. Fleming G. E. Krauter M. Starr
1	<u>Navy Office of the Chief of Naval Operations (Wash., D.C.)</u> F. S. Adair
7	<u>Oak Ridge National Laboratory (AEC)</u> R. S. Crouse J. R. DiStefano J. H. Gillette E. Lamb C. Ottinger J. E. Ratledge A. F. Rupp
1	<u>Savannah River Laboratory (AEC)</u> R. K. Huntoon

No. of
Copies

1	<u>Savannah River Plant (AEC)</u> R. H. Bass
1	<u>Teledyne Isotopes</u> P. Vogelberger
1	<u>Westinghouse Astronuclear Laboratory</u> C. C. Silverstein

ONSITE

3	<u>AEC Richland Operations Office</u> P. G. Holsted D. K. Jones B. J. Melton
8	<u>Atlantic Richfield Hanford Company</u> R. E. Isaacson L. M. Knights C. W. Malody J. D. Moore G. C. Oberg H. P. Shaw A. E. Smith M. J. Szulinski
2	<u>D. W. Douglas Laboratories</u> D. E. Knapp W. E. Matheson
24	<u>Battelle-Northwest</u> E. L. Alpen J. W. Bartlett R. E. Burns T. D. Chikalla G. J. Dau R. L. Dillon J. W. Finnigan H. T. Fullam (3) A. J. Haverfield J. H. Jarrett R. S. Kemper R. P. Marshall R. W. McKee

J. M. Nielsen
R. E. Nightingale
L. D. Perrigo
A. M. Platt
C. A. Rohrmann
H. H. Van Tuyl
Technical Information Files (3)