228 + 120 FAST REACTOR

ANL-7475

1/29/69

key

#634

ANL-7475

MASTER

Argonne National Laboratory

URANIUM NITRIDE-SODIUM PASTE FUEL FOR FAST-REACTOR BLANKETS

by

P. A. Nelson and M. G. Chasanov

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

The facilities of Argonne National Laboratory are owned by the United States Government. Under the terms of a contract (W-31-109-Eng-38) between the U. S. Atomic Energy Commission, Argonne Universities Association and The University of Chicago, the University employs the staff and operates the Laboratory in accordance with policies and programs formulated, approved and reviewed by the Association.

MEMBERS OF ARGONNE UNIVERSITIES ASSOCIATION

The University of Arizona	Kansas State University	The Ohio State University
Carnegie-Mellon University	The University of Kansas	Ohio University
Case Western Reserve University	Loyola University	The Pennsylvania State University
The University of Chicago	Marquette University	Purdue University
University of Cincinnati	Michigan State University	Saint Louis University
Illinois Institute of Technology	The University of Michigan	Southern Illinois University
University of Illinois	University of Minnesota	University of Texas
Indiana University	University of Missouri	Washington University
Iowa State University	Northwestern University	Wayne State University
The University of Iowa	University of Notre Dame	The University of Wisconsin

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in the United States of America

Available from

Clearinghouse for Federal Scientific and Technical Information

National Bureau of Standards, U. S. Department of Commerce

Springfield, Virginia 22151

Price: Printed Copy \$3.00; Microfiche \$0.65

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

URANIUM NITRIDE-SODIUM PASTE FUEL
FOR FAST-REACTOR BLANKETS

by

P. A. Nelson and M. G. Chasanov

Chemical Engineering Division

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

August 1968

**THIS PAGE
WAS INTENTIONALLY
LEFT BLANK**

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	7
I. INTRODUCTION	7
II. EVALUATION OF BLANKET-FUEL REQUIREMENTS	8
A. Solid-fuel Blankets	8
1. Nuclear Properties	8
2. Heat-transfer Considerations	10
B. Selection of a Fluid Blanket Fuel	13
1. Molten Alloys and Fused Salts	13
2. Slurries and Pastes	14
3. Advantages of Uranium Mononitride-Sodium Suspensions	15
C. Engineering and Economic Considerations for Fluid-fuel Blankets	15
1. Reactor Requirements	15
2. Static-fuel Systems	15
3. Paste Flow Systems	16
4. Heat Transfer	17
5. Economic Advantages	19
a. Removing and Shuffling the Fuel	19
b. Coolant Inefficiency	20
c. Fabrication	20
d. Reprocessing	21
e. Total Savings	22
III. EXPERIMENTAL WORK IN DEVELOPMENT OF URANIUM NITRIDE-SODIUM PASTE FUEL	22
A. Preparation of Uranium Mononitride-Sodium Suspensions	22
1. Preparation of High-density UN Particles	22
2. Wetting of Uranium Ceramics by Liquid Sodium	26
3. Mixing of Uranium Mononitride-Sodium Paste Constituents	28
B. Flow Properties of High-solids Suspensions	28

TABLE OF CONTENTS

	<u>Page</u>
C. Compatibility of Type 304 Stainless Steel with Sodium-paste Reactor Fuels.	29
1. Control Experiments without Ceramic Fuels	30
2. Sodium-Uranium Dioxide Paste	31
3. Sodium-Uranium Nitride Pastes	31
4. Sodium-Uranium Carbide Pastes	31
5. Summary of Corrosion Data	32
IV. DISCUSSION AND CONCLUSIONS	33
ACKNOWLEDGMENT	34
REFERENCES	35

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Power Density in Blanket Region Adjacent to the Core for 1000-MW(e) Ceramic-fueled Fast Reactors	11
2.	Photomicrograph of Uranium Mononitride from Run UN-4	24
3.	Photomicrograph of Uranium Mononitride from Run UN-14 . . .	25

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.	Densities of Some Uranium Compounds	9
II.	Performance of Sodium Coolant in Radial Blanket of Reactor Designed by Combustion Engineering, Inc.	13
III.	Heat-transfer Comparison for Various Blanket Fuels	18
IV.	Estimated Savings for Paste-fuel Radial Blankets	19
V.	Run Conditions and Product Analysis for Nitridation of Uranium Shot.	23
VI.	Effect of Oxidation during Grinding of Bulk Density of Uranium Mononitride	25
VII.	Contact Angles Determined by Sessile-drop Method for Liquid Sodium and Uranium Ceramics	27
VIII.	Results of Static Corrosion Tests on Type 304 Stainless Steel Coupons Immersed in Sodium-Ceramic Pastes for 1000 hr at 1400°F	30

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

URANIUM NITRIDE-SODIUM PASTE FUEL FOR FAST-REACTOR BLANKETS

by

P. A. Nelson and M. G. Chasanov

ABSTRACT

An evaluation of various fluid fuels for fast-reactor blankets indicated that a paste of UN particles in a sodium vehicle is the most attractive. This fuel concept might save 0.06-0.07 mill/kW-hr or about 20% of the total blanket costs, including the costs of removing and shuffling fuel, coolant inefficiency, fuel fabrication, and fuel reprocessing. To support the conclusions of the evaluation study, a UN-sodium paste of 49 v/o solids was prepared and shown to be compatible with a Type 304 stainless steel container. Paste-flow measurements and sodium-wetting experiments also indicated the feasibility of this fuel concept.

I. INTRODUCTION

Fluid fuels for fast reactors are attractive because they avoid some of the disadvantages of solid fuels, namely, (1) failure of the cladding as a result of fuel swelling, (2) intricate and costly fuel fabrication, and (3) difficult fuel disassembly before reprocessing. However, fluid fuels add other complexities, even when used in heterogeneous reactors. Fuel-pumping apparatus may be required for loading and unloading the reactor and for fuel circulation during irradiation. The formation and collapse of gas bubbles in the fuel may cause sudden changes in reactivity. Gas bubbles may also interfere with heat transfer. The fuel centerline temperature may be limited by the boiling point of the fuel, or that of the vehicle in a suspension fuel. The fuel flow may be blocked by bubbles, by accumulation of precipitates, or by solids agglomeration in a suspension fuel.

Some of these problems of fluid fuels, particularly those having to do with reactor reactivity and heat transfer, are less severe for blanket fuel than for core fuel. Also, there are additional advantages for the use of fluid fuel in the blanket that do not apply for its use in the core. For instance, the large change in heat-generation rate during the reactor cycle that takes place in solid-fuel blanket assemblies may be avoided in fluid-fuel blanket assemblies by circulating the fuel or withdrawing it frequently for processing. Thus, utilization of the sodium coolant would be improved.

Since a fluid fuel in the blanket appears to offer significant improvements over solid fuels and less severe problems than would its use in the core, this investigation was restricted to fluid blanket fuels. The design of paste-fuel blankets was also considered. The first part of this report evaluates blanket-fuel requirements; the second part discusses experimental work in developing UN-sodium paste fuel, the fluid fuel selected as the most attractive.

II. EVALUATION OF BLANKET-FUEL REQUIREMENTS

A. Solid-fuel Blankets

1. Nuclear Properties

The breeding ratio¹ for a reactor is given by

$$\text{Breeding ratio} = \frac{\nu - 1 - \alpha - A - L + (\nu' - 1) F}{1 + \alpha}, \quad (1)$$

where

ν = number of neutrons emitted per fission in fissile material,

α = the ratio of capture to fission,

A = parasitic absorption of neutrons in nonfissile and nonfertile materials,

L = leakage of neutrons from the blanket,

F = fissions in fertile material,

and

ν' = number of neutrons emitted per fission in fertile material.

The blanket characteristics affect the breeding ratio in that they moderately influence the average neutron energy and parasitic absorption for the entire reactor (A), and are the controlling factor in leakage of neutrons from the blanket (L). The breeding potential of the primary fissionable material is determined by $\eta - 1$, where $\eta = \nu/(1 + \alpha)$ = the number of neutrons emitted per absorption in fissile material. For ^{239}Pu , ν becomes larger and α becomes smaller as the neutron energy increases. As a result, $\eta - 1$ is largest at high neutron energies.²

Thus, breeding is enhanced by increasing the neutron energy. However, the capture cross section of ^{238}U also decreases with increasing energy; hence the L term in Eq. 1 would become larger unless the blanket were made thicker. Addition of a moderator to the blanket to degrade the

spectrum would reduce L for a given blanket thickness; however, for a thinner blanket, ν , ν' , and F would also be reduced. That this may not result in a serious net loss in breeding was indicated by calculations of Hasnain and Okrent³ for 800-liter spherical cores (50 v/o Na, 25 v/o Fe, 25 v/o $^{238}\text{U} + ^{239}\text{Pu}$) surrounded by 45-cm blankets of uranium, sodium, and iron that were either nonmoderated or graphite-moderated; the nonmoderated blankets were only slightly superior in breeding. In design of blankets for large reactors, the structural material probably has a lesser effect on breeding gain in the blanket than in the core.¹

To use a high percentage of the neutrons for breeding, the density of fuel material in the blanket should be as large as practicable. Recent proposals⁴⁻⁹ for 1000-MW(e) systems with carbide and oxide fuels use blankets with fuel volume fractions of the order of 50%. Table I gives the densities for some possible blanket materials. The peritectoid reaction at 930°C for U_3Si produces γ -uranium and U_3Si_2 ; the volume increase involved in the transformation could rule against the use of U_3Si . The order of preference, based on density, of ceramic blanket fuels is UN, UC, U_3Si_2 , UO_2 , US, and UP. The densities of UCl_3 and UF_4 are quite low in comparison to the others.

TABLE I. Densities of Some Uranium Compounds

Material	Melting Point (°C)	Density (g/cc)	Uranium Density (g/cc)	Uranium Density of Compound Relative to That of Uranium Metal
Uranium Metal	1132	19.05	19.05	1.000
U_3Si	930 ^a	15.58	14.99	0.787
UN	2850 ^b	14.32	13.52	0.710
UC	2375	13.63	12.97	0.681
U_3Si_2	1665	12.20	11.31	0.594
UO_2	2760	10.97	9.67	0.508
US	2462	10.83	9.55	0.501
UP	~2540	10.3	9.16	0.481
UF_4	960	6.70	5.25	0.276
UCl_3	835	5.51	3.81	0.200

^aPeritectoid.

^bAt 2.5 atm nitrogen.

The selection of blanket thickness should be based on an economic optimization of fabrication and reprocessing costs and plutonium credit. The concentration of plutonium bred in the outer portions of the blanket is considerably less than that in the portions next to the core. Designs proposed for the large power reactors mentioned above have blankets of the order of 30-40 cm thick selected by approximate economic optimizations.

The requirements for the radiation stability of solid blanket fuels are almost as important as for core fuels, since high burnups can be expected in parts of the blanket. The chief problems, those of dimensional stability and changes in physical, chemical, and mechanical properties, are the same as for core fuels.

2. Heat-transfer Considerations

The power density in the blankets of fast reactors varies considerably with the local plutonium density and the nearness to the core (i.e., neutron-flux level). The power density in the blanket region near the core where the average neutron energy is above 10 keV is given by

$$P_b[\text{kW(t)/liter}] = \left(\frac{\rho_{\text{Pu}}}{A_{\text{Pu}}} \bar{\sigma}_{\text{Pu}_f} + \frac{\rho_{\text{U}}}{A_{\text{U}}} \bar{\sigma}_{\text{U}_f} \right) \frac{E_f N_A \phi}{10^{24}}, \quad (2)$$

where

E_f = fission energy, 3.25×10^{-11} W-sec/fission of actinide atom;

N_A = Avogadro's number, 6.02×10^{23} atoms/g-atom;

ϕ = neutron flux, $n/(\text{cm}^2)(\text{sec})$;

ρ_{Pu} = fissile plutonium density, g/cc of blanket volume;

ρ_{U} = uranium (nonfissile) density, g/cc of blanket volume;

A_{Pu} = atomic weight of plutonium, g/g-atom;

A_{U} = atomic weight of uranium, g/g-atom;

$\bar{\sigma}_{\text{Pu}_f}$ = average microscopic cross section of fissile plutonium, b;

and

$\bar{\sigma}_{\text{U}_f}$ = average microscopic cross section of uranium, b.

For 1000-MW(e) ceramic-fueled reactors and for the blanket region immediately adjacent to the core, where heat generation is at a maximum for the blanket,

$$\phi = \sim 3 \times 10^{15} n/(\text{cm}^2)(\text{sec}) \text{ [normal range } 1.5 \times 10^{15} \text{ to } 5 \times 10^{15} n/(\text{cm}^2)(\text{sec})],$$

$$\bar{\sigma}_{\text{Pu}_f} = \sim 2 \text{ b (neutron energies of 0.01-1.0 MeV),}$$

and

$$\bar{\sigma}_{U_f} = 0.03 \text{ b (normal range 0.015-0.05 barn).}^*$$

The following relationship was derived for the power density in the blanket region adjacent to the core:

$$P_{bc} [\text{kW(t)/liter}] = 500(\rho_{Pu} + 0.015\rho_U) \approx 500\rho_a(Pu + 0.015), \quad (3)$$

where

ρ_a = density of actinide elements, g/cc of blanket volume,

and

Pu = fraction fissile plutonium in blanket actinides.

Fig. 1. Power Density in Blanket Region Adjacent to the Core for 1000-MW(e) Ceramic-fueled Fast Reactors

Thus, for this typical case, the initial ^{238}U content of the fuel is equivalent in heat-generation capacity to about 1.5 a/o plutonium in the blanket fuel.

Figure 1 is a plot of Eq. 3, showing the power buildup in the blanket region adjacent to the core as the plutonium concentration increases. In solid-fueled radial blankets, the plutonium concentration in the blanket pins adjacent to the core may reach 6 a/o even though the average plutonium concentration for the entire assembly is only 3-4 a/o.⁷ The heat-generation rate will vary considerably with the location within the blanket and with the length of time the assembly has been in the reactor.

In the current design concepts, the use of sodium coolant in

solid-fueled blankets is inefficient and reduces the overall energy-conversion efficiency of the reactor below what it would be for the core alone. The sodium flow to each assembly is controlled by an orifice at the bottom of

*The threshold energy for fission of ^{238}U is about 1.0 MeV. At 2-6 MeV, $\bar{\sigma}_{238U_f} = 0.6 \text{ b}$. The fraction of flux with neutron energy above 1.5 MeV (average energy between threshold energy and energy range of constant cross section) is about 0.05 for large fast reactors in the region near the core-blanket boundary. Hence, $\bar{\sigma}_{U_f} = 0.6(0.05) = 0.03\text{b}$.

each assembly. In reactors in which the fuel is not shuffled, the sodium flowrate to the assembly remains constant throughout the residence time of the assembly in the reactor. When the assemblies are shuffled periodically, the flowrate at each position remains constant during the entire period. The coolant flowrate must be sufficient to prevent failure of the fuel cladding for the highest heat-generation rates attained during the reactor cycle. For high plutonium buildup (2-3 a/o average), Fig. 1 shows that the disparity between the initial and final heat-generation rates is too great for efficient utilization of the sodium. Another factor contributing to poor sodium utilization is the disparity between the heat-generation rate for rods nearest the core in a fuel assembly and that for rods farthest from the core in the same assembly. The flowrate through the entire assembly must be sufficient for the hottest channel; therefore, the average sodium temperature is considerably lower than the maximum allowable temperature.

To mitigate this problem, the movement of blanket assemblies at regular intervals during the reactor cycle has been proposed. Such motion can be mere rotation of the assemblies or radial movements of the assemblies toward the core or away from it. Since production of fissile material is greatest near the core, outward movement of the rods from the positions nearest the core would result in more uniform concentration of bred material, but would result in a high plutonium inventory in the blanket. Inward movement of the blanket rods lowers plutonium inventory, but results in a wide disparity between the initial and final power density in the fuel assemblies. Many schemes and mechanics of moving blanket assemblies have been proposed that optimize breeding and power generation while reducing inventory and downtime costs. These can become quite complex. Most designs incorporate the axial blanket as part of the core fuel assemblies; thus, movement of the axial blanket is tied up with movement of the core assemblies. Inversion of the axial blanket has also been proposed, although this introduces additional fuel-handling complications.

Table II is an example of the effect of the buildup of power generation in the blanket subassemblies and of rotation of assemblies on efficiency of sodium utilization. Without rotation of assemblies, the average blanket outlet temperature is low. The relatively cool sodium from the blanket would result in a reduction in the average reactor outlet temperature and a serious loss in conversion efficiency. Even with rotation of the assemblies after half the irradiation exposure, the sodium outlet temperature is far below the maximum outlet temperature of 1260°F (682°C) that could be reached with a constant plutonium concentration.

Therefore a blanket fuel, such as a fluid fuel, that would allow continuous or frequent movement of the fuel within the blanket would result in much more efficient sodium utilization than can be attained with solid fuels.

TABLE II. Performance of Sodium Coolant in Radial Blanket of Reactor Designed by Combustion Engineering, Inc.⁷

	180° Rotation of Subassembly at Half of Total Exposure	No Rotation
Average power density, kW(t)/liter	74	(Not given)
Maximum power density, kW(t)/liter	558	(Not given)
Average fissile concentration at discharge, %	3.86	(Not given)
Total blanket flowrate, lb/hr	20×10^6	46×10^6
Final temperature of hottest tube cladding, °F	1400	1400
Sodium inlet temperature, °F	850	850
Average reactor outlet temperature, °F	1100	<1100
Average blanket outlet temperature, °F	1050	936
Final blanket outlet temperature of hottest channel, °F	1260	(Not given)

B. Selection of a Fluid Blanket Fuel

Fluid-fuel blankets have been proposed for use with both breeder and converter reactors. Slurry-type blankets have had several proponents. The Liquid Metal Fuel Reactor¹⁰ was to use a slurry of thorium bismuthide in molten bismuth. The finite solubility of this intermetallic compound in bismuth resulted in interparticle mass transfer during thermal cycling of loops with frequent blockage of the pumped circuit.¹¹⁻¹³ Frost¹⁴ stated that such a fuel is probably not feasible because a very thick slurry would be required and sintering could occur at elevated temperatures. Use of a dispersed phase, which is insoluble in the vehicle, can eliminate the mass-transfer problem, and use of a slowly moving or settled paste can reduce the erosion and comminution problems inherent in the fast-flowing slurry systems. Paste blankets of these types have been proposed for a Fused Salt Fast Breeder¹⁵ and a Mobile Fuel Plutonium Breeder.¹⁶

The economics of paste blankets for fast reactors was considered by Hasnain and Okrent³ and more recently in an analytical study by Klickman et al.¹⁷ of Atomic Power Development Associates (APDA). The APDA report concludes that a paste blanket offers more economic advantage for fast ceramic-fueled reactors than either a fixed blanket or a settled bed of clad spheres. Their choice for a paste was uranium monocarbide dispersed in molten sodium; this choice was based on the high thermal conductivity of UC in comparison with UO₂.

1. Molten Alloys and Fused Salts

Few metallic solvents will dissolve appreciable quantities of uranium. The better solvents are aluminum (~32 w/o uranium at 900°C),

bismuth (~16 w/o uranium at 900°C), and alloys of magnesium and zinc (37 w/o uranium at 900°C). However, the uranium densities obtainable are relatively low, of the order of 1-3 g/cc. Uranium-based eutectic alloys can have greater densities, but their melting points are usually quite high (~700-1100°C). The major difficulty with uranium-based molten-alloy systems is corrosion; these systems are quite corrosive to all metals except a few of the refractory metals. Plug formation can also result because of the temperature coefficient of uranium solubility.

Fused-salt systems containing uranium halides are characterized by lower melting points than the above molten-alloy systems. However, the low uranium density and corrosive nature of fused salts detract from their utility in a fluid blanket.

2. Slurries and Pastes

For long-term applications, the components of a slurry or paste must be chemically stable with respect to one another and to the container material. In addition, to avoid particle consolidation, the dispersed phase must be insoluble in the vehicle. Suspensions that might meet these criteria are dispersions of uranium compounds in liquid metals.

Liquid sodium appears to be a logical initial choice for the vehicle because it does not dissolve uranium compounds, it has a low melting point (97.9°C) and a high thermal conductivity, and it can be contained by stainless steel. The main disadvantage of sodium is its low boiling point (882.9°C). Although lead-bismuth alloys have low melting points and much higher boiling points, they have the disadvantage of low thermal conductivities.

On the basis of uranium density alone, UN is the most attractive solid material for such suspensions, followed by UC, UO_2 , and US (as shown in Table I).

Compatibility with the cladding is another important criterion. Since it is difficult to prepare these uranium compounds with precise stoichiometry, slight deviations from stoichiometry must be considered in evaluating compatibility with the cladding. Markedly hypostoichiometric UO_2 , UC, UN, and other such uranium compounds contain free uranium that may react with stainless steel cladding to form low-melting uranium-iron-chromium eutectics, which would cause rapid destruction of the cladding. Hyperstoichiometric UO_2 would be expected to react with sodium to form Na_2O , which is extremely corrosive to stainless steel in a liquid-sodium medium. Hyperstoichiometric UC would be expected to cause carburization and embrittlement of stainless steel. However, release of excess nitrogen from hyperstoichiometric UN is less likely to be deleterious to stainless steel. These reactions were studied in a preliminary series of

experiments (discussed in Section III.C of this report) to estimate the degree of corrosion that might occur. From thermodynamic considerations alone, however, UN in a sodium medium is more likely to be compatible with stainless steel than UO_2 or UC.

3. Advantages of Uranium Mononitride-Sodium Suspensions

On the basis of the physical, chemical, and corrosion properties discussed above, the most promising blanket paste fuel for initial study appears to be UN dispersed in molten sodium; other systems that might also be considered are UC-sodium and UO_2 -sodium. Uranium nitride appears the most attractive of the fuels because of its high uranium density, good thermal conductivity, and apparent compatibility with sodium and with stainless steels at the temperatures of interest. Furthermore, UN is relatively easy to prepare, can be stored without difficulty in moist air atmospheres, and is unlikely to sinter at reactor temperatures.

C. Engineering and Economic Considerations for Fluid-fuel Blankets

1. Reactor Requirements

If reactors with fluid-fuel blankets are to have an economic advantage over reactors with solid-fuel blankets, this advantage must be gained without great changes in the core design. The nuclear characteristics of the core, the ease of removal of the core fuel, and the core fuel-cycle costs have much greater effect on the overall economics of the reactor than do blanket economics. Accordingly, a fluid fuel would probably not be suitable for axial blankets, particularly the top blanket.

Fluid blanket fuel appears to be best suited for reactors requiring large radial blankets and producing a high fraction of the breeding in the radial blanket. Reactors having pancake-shaped cores and, thus, low-volume radial blankets,⁸ would not be suitable for fluid-fuel blankets.

2. Static-fuel Systems

It may be feasible to operate a paste-fuel blanket with stationary fuel. To do so economically and safely, the following criteria must be met:

- a. The reprocessing rate must be high enough to limit the plutonium buildup to less than 1% to avoid wide variation in the heat-generation rate with exposure time and the resulting inefficiency in coolant utilization.
- b. Fuel assemblies must be removed from the reactor to process the fuel.
- c. The total volume of fission gas released from the fertile particles must be a small percentage of the volume of the fuel.

Because of probable savings in fabrication costs for paste fuels as compared to solid fuels, it may be economically feasible to maintain a low plutonium concentration in paste fuels by frequent processing and thus meet the first criterion. Also, removal of the blanket assemblies from the reactor during shutdown periods (second criterion) might be relatively inexpensive if this procedure did not lengthen the shutdown period. However, fission-gas buildup in the blanket fuel (third criterion) may be a problem. Gas bubbles would have a deleterious effect on heat transfer,¹⁸ resulting in hot spots that might cause local boiling of sodium.

The volume of fission gas released from the fertile solids in a paste fuel is a function of burnup, temperature, and the particle composition, size, and structure. In the early stages of irradiation of blanket fuel, the power density is low, as indicated in Fig. 1, and therefore, fuel burnup proceeds slowly. If fuel is removed for processing when the plutonium concentration reaches 1%, the burnup may be limited to about 0.5%. The exact burnup for a given plutonium concentration would depend on the flux spectrum. In experiments at BMI,¹⁹ at temperatures below 800°C, required for sodium-vehicle pastes because of the low boiling point of sodium, the fission-gas release rate for sintered UN was temperature-independent. The instantaneous release rates under these conditions for ^{85m}Kr, ¹³⁵Xe, and ¹³³Xe were 0.08, 0.1, and 0.2% of the instantaneous production rates, respectively. With fine particles, the release rates would be higher. At 1400°C, the amount of xenon released in 3 hr from UO₂ passing through a 325-mesh screen was 3.5% compared with 0.75% for material passing through a 100-mesh screen; the release rates at 800°C should be markedly less than at 1400°C. If 1% of the gas produced is released from the particles, the volume of gas at 0.5% burnup and 800°C will be about 3% of the paste volume for 50 v/o UN in sodium. This amount of gas release is not likely to have a significant effect on heat-transfer rates.

It is concluded that blanket paste fuel contained in a static condition in a fuel assembly may be an improvement over solid fuel if the fission-gas release rate is sufficiently low.

3. Paste Flow Systems

Systems in which the paste is circulated throughout the blanket would have uniform plutonium concentration and, thus, high sodium-utilization efficiencies. Fission gas could be removed by a device in the flow system. The fuel could be replaced by fresh fuel either continuously or intermittently, thus maintaining a desired plutonium concentration. The required flow velocity would be only a few feet per hour. The disadvantages of flow systems are the additional costs of building and operating the pumping and piping, the risk of a flow blockage, and particle comminution.

One method for moving the paste is by an eductor.^{17,20,21} Sodium pumped at high velocity through the eductor would fluidize the paste entering the eductor at a point of low pressure. The slurry would flow from the reactor, a side stream would be withdrawn for processing, and the paste would be reconstituted in a cyclone separator and re-enter the reactor. The sodium from the cyclone separator would flow to a pump, the only motive force in the system, and back to the eductor. Another method of moving the paste, that of direct pumping, should also be considered. However, erosion in the pump may be a serious problem.

To minimize the risk of flow blockage in the paste system, the flow passages should be as large as possible and free of constrictions.

The flow characteristics of pastes are a function of size and shape of the particle and viscosity of the vehicle. Experimental studies of paste flow characteristics were undertaken as a part of this investigation; the results are discussed in Section III.B below.

4. Heat Transfer

At the low flow velocity required for the paste fuel, the flow would be laminar and the rate of heat transfer within the paste would be the same as for a solid of the same conductivity as the paste. The thermal conductivity of a paste having 50-100 v/o liquid phase may be determined from the relationship^{22,23}

$$k = k_v \frac{1 - (1 - ak_p/k_v) b}{1 + (a - 1) b}, \quad (4)$$

where k_v = thermal conductivity of the vehicle, k_p = thermal conductivity of the solid particles, $a = 3k_v/(2k_v + k_p)$, and b = volume fraction of solids.

The thermal conductivities at 1500°F for UO_2 -50 v/o Na, UN-50 v/o Na, and UN-50 v/o Pb-Bi eutectic (44.5 w/o lead) were calculated from Eq. 4; these are presented in Table III with other physical-property data. The ratio of the thermal conductivity of the liquid-metal vehicle to that of UN at 1500°C is 2.48 for a sodium vehicle and 0.635 for a lead-bismuth eutectic vehicle. However, the favorable thermal conductivities of sodium-vehicle pastes are offset by the low maximum centerline temperatures as restricted by the volatility of the sodium.

Approximate relative fuel diameters were calculated for solid and paste blanket fuels from the relationship for steady-state heat transfer from a rod with uniform internal heat generation and uniform thermal conductivity,

$$\Delta T = \frac{qr^2}{4k}, \quad (5)$$

where

ΔT = temperature differential between the centerline and the surface of the fuel rod,

q = heat-generation rate per unit volume,

r = radius of fuel rod,

and

k = thermal conductivity of fuel.

TABLE III. Heat-transfer Comparison for Various Blanket Fuels

Fuel	Density at 1500°F (g/cc)	Uranium Density (ρ_U) at 1500°F (g/cc)	Thermal Conductivity (k) at 1500°F [Btu/(hr)(ft)(°F)]	Maximum Centerline Temperature (°F)	Fuel Diameter (Arbitrary scale ^b)
Uranium Metal	18.2	18.2	3.0	1800 ^a	0.45
UO ₂	10.9	9.5	1.5	4700 ^c	0.34
UC	13.5	12.9	12	2500 ^a	0.50
UN	14.2	13.5	12	2500 ^a	0.49
UO ₂ -50 v/o Na	5.8	5.1	13	1500 ^d	0.40
UN-50 v/o Na	7.5	6.7	21	1500 ^d	0.44
UN-50 v/o Pb-Bi Eutectic ^e	11.9	6.7	10	2400 ^d	0.61

^aLimited by irradiation damage.

^bFuel diameter, d , calculated by assuming the neutron flux and the plutonium-to-uranium ratio to be the same for all fuels and a temperature differential, ΔT , obtained from the maximum centerline temperature and an external fuel temperature of 1200°F; $d = c/\sqrt{k\Delta T/\rho_U}$, where c was chosen to yield a diameter of 0.5 for UC.

^cLimited by melting.

^dLimited by the boiling point of the vehicle, which is 1621°F for sodium and about 2800°F for lead-bismuth eutectic.

^e44.5 w/o lead, 54.5 w/o bismuth.

The neutron flux in the blanket region immediately adjacent to the core is a function mainly of the reactor-core characteristics and is not greatly affected by the blanket-fuel composition. Therefore, the heat-generation rate per unit volume in the blanket fuel near the core is chiefly a function of the uranium and plutonium density in the blanket fuel. For a stated plutonium-to-uranium ratio and a stated neutron flux, Eq. 5 shows the allowable diameter of the fuel rod for a given temperature differential to be proportional to $\sqrt{k\Delta T/\rho_U}$, where ρ_U is the uranium density in g/cc of fuel.

Based on this relationship, relative diameters of paste and solid blanket fuel elements are presented in Table III for a ΔT calculated from the maximum centerline temperatures given in Table III and an assumed external fuel temperature of 1200°F. The allowable fuel diameter for UN-50 v/o paste would be slightly less than that for solid UC or UN. The tubes containing UN-50 v/o Pb-Bi blanket paste could be about 40% larger than the tubes containing UN-50 v/o Na paste.

If paste fuel is circulated to distribute bred plutonium throughout the blanket, the heat-generation rate per unit weight of fertile uranium

could be maintained at one-third the peak values (which limit the fuel diameter) reached in solid blanket fuels. This would allow fuel tube diameters for UN-50 v/o Na paste that are 50% larger than the 1/2-in.-diam solid ceramic blanket pins of current design concepts. Thus, a fuel tube diameter of about 3/4 in. would be acceptable for UN-50 v/o Na blanket fuel.

5. Economic Advantages

There are four possible economic advantages for paste-fuel blankets as compared with solid-fuel blanket materials: (a) reduced downtime for fuel shuffling, (b) improved sodium coolant utilization, and, thus, higher conversion efficiency, (c) decreased fuel-processing cost, and (d) decreased fuel-refabrication costs. Table IV presents typical costs of these items for solid-fuel blankets for 1000-MW(e) reactors; these costs are based primarily on Refs. 4-9. Table IV also includes our estimates of what could be saved by the use of static and circulated paste blanket fuels.

TABLE IV. Estimated Savings for Paste-fuel Radial Blankets

Cost Item	Cost for Solid-fuel Blanket (mill/kW-hr)	Estimated Savings for Paste-fuel Blanket (mill/kW-hr)	
		Static Fuel	Circulating Fuel
Removing and shuffling fuel ^a	0.04	0	0.01
Coolant inefficiency	0.05	0.02	0.04
Fabrication ^b	0.17	0.04	0
Reprocessing ^b	0.10	0	0.02
Total cost or savings	0.36	0.06	0.07

^aIncludes fuel circulation for circulating paste blanket fuel.

^bSolid and circulated paste fuel removed for processing on attaining about 2% average plutonium concentration; static paste fuel removed at about 1% plutonium concentration.

a. Removing and Shuffling the Fuel. The time required for removing and shuffling solid blanket fuel for a 1000-MW(e) reactor was estimated⁵ to be 6.1 days per year or 1.67%. Since other operations, such as maintenance of the turbine-generator, could be carried out during this period, the time would not be entirely lost. Therefore, the economic incentive for eliminating downtime for blanket reshuffling was assumed to be 1% (rather than 1.67%) of the fixed power costs. For fixed power costs (which are 60 to 80% of the total power cost) of 3 mills/kW-hr, the cost of removing and shuffling solid blanket fuel would be about 0.03 mill/kW-hr. An additional 0.01 mill/kW-hr was allowed for equipment and labor, yielding a total cost for removing and shuffling solid blanket fuel of 0.04 mill/kW-hr.

For static paste blanket fuel, shuffling would probably be unnecessary because of the low plutonium concentrations attained. However, removal of assemblies would be more frequent, and no savings in this category are foreseen for static-paste-fuel systems. A circulating paste fuel would require no reactor downtime for removing or shuffling of fuel. However, the permanent circulating equipment would add approximately \$1,000,000 to the cost of capital equipment²⁰ or, at an annual capital cost rate of 15%, \$150,000 per year. This cost, together with an operating cost of \$50,000 per year for withdrawal and circulation of blanket fuel (at 80% plant load factor), results in a total cost of paste-fuel movement of 0.03 mill/kW-hr. Thus, there is a net saving for the circulating paste fuel of 0.01 mill/kW-hr. This small indicated saving is probably less than the accuracy of the estimate.

b. Coolant Inefficiency. The cost of poor sodium utilization in solid-fuel blankets was estimated from the data in Table II. For rotated assemblies, 20×10^6 lb/hr of blanket sodium at a mean temperature of 1050°F is mixed with 86×10^6 lb/hr of core sodium, resulting in an average temperature of 1100°F. In theory, if the plutonium concentration did not change with time and the blanket assemblies were properly orificed, the sodium outlet temperature could be maintained at 1260°F, the final temperature of the hottest channel in the solid-fuel blanket (as shown in Table II). The blanket heat would then be removed by 10×10^6 lb/hr of sodium, and the resulting reactor sodium temperature would be 1125°F. (This higher exit sodium temperature is obtained with no increase in the maximum cladding temperature.) If the discharge temperature from the steam turbine is assumed to be 150°F, then the increase in the sodium temperature from 1100 to 1125°F would result in an increase in the overall energy-conversion efficiency from 42.6 to 43.1% (70% Carnot-cycle efficiency for both). At a power cost of 4 mills/kW-hr, the higher sodium temperature would save 0.05 mill/kW-hr.

The more uniform heat generation in the static-paste system that is obtained by limiting the plutonium buildup would result in a saving estimated to be about 40% of the total potential saving or about 0.02 mill/kW-hr. The circulating-paste system could probably save 80% of the coolant inefficiency cost or about 0.04 mill/kW-hr. This allowance for less than 100% efficient coolant utilization is necessary for power excursions and for variation in flux intensity across the fuel assemblies.

c. Fabrication. Most fuel-cycle cost studies include the cost of powder preparation, fuel pelletization, encapsulation, and metal hardware in the cost of fabrication. In recent studies on 1000-MW(e) reactors,⁵⁻⁸ the total cost of these items was estimated to be about \$50 to \$78/kg for blanket fuel. However, the evaluation report⁴ for these studies estimated the fabrication costs in the original studies to be too low by 20-45%. Consequently,

in this study, the cost was assumed to be \$80/kg of blanket fuel. The amount of radial blanket fuel to be processed varies considerably: from 3860 kg/yr (metal basis) for the General Electric pancake-shaped reactor,⁸ to 21,694 kg/yr for the annular-shaped Allis-Chalmers reactor.⁵ Since a paste fuel would not be suitable for a pancake-shaped reactor, a value near the higher figure would be more appropriate for this study. A blanket-fuel processing rate of 15,000 kg/yr was assumed, which yields a fabrication cost of \$1,200,000/yr or 0.17 mill/kW-hr based on 80% plant load factor.

The main cost saving for static paste fuel would be the elimination of pelletization and part of the cost of encapsulation. The stainless steel subassembly structure should last about 6-10 yr, about twice as long as solid-fuel subassemblies, and would then have to be replaced because of irradiation-induced embrittlement. The estimated savings are about one-fourth of the total solid-fuel refabrication cost or about 0.04 mill/kW-hr.

Fabrication costs for the circulating-fuel blanket are expected to be about the same as for a solid-fuel blanket. Part of the circulating equipment, such as eductors and cyclone separators, will be permanent parts of the fuel assembly and will be discarded with the fuel assembly every 6-10 yr. Consequently, although the cost of fuel pelletization and loading of the fuel into the cladding is eliminated (or covered in the capital and operating costs of fuel circulation included in Table IV under "Removing and shuffling fuel"), the extra cost of stainless steel hardware should result in no net savings for fabrication of circulating-blanket-fuel assemblies as compared to solid-blanket-fuel assemblies.

d. Reprocessing. Reprocessing of solid blanket fuel was estimated to cost 0.03 mill/kW-hr for the General Electric reactor⁸ and 0.10 mill/kW-hr for the Allis-Chalmers reactor.⁵ Since the blanket reprocessing rate for the Allis-Chalmers reactor is about what would be expected for a reactor suitable for a paste-fuel blanket, the higher cost of 0.10 mill/kW-hr was chosen as representative.

The cost of reprocessing paste blanket fuel is difficult to estimate even though all that is necessary for this study is the cost relative to solid-fuel processing. If possible, the plutonium and fission products should be removed from the blanket fuel without converting to oxides. This operation might be possible by pyrometallurgical techniques. It would not be necessary to completely separate plutonium and fission products from each other since a product containing both could be introduced to the core fuel process.

If it is assumed that such a process can be devised, then the reprocessing cost for static paste fuel might be about equal to the cost

for solid fuel; the higher processing rate for the paste would be offset by a lower cost per kilogram processed. For the circulating fuel, the reprocessing cost was estimated to be about 0.02 mill/kW-hr less than for solid blanket fuel.

e. Total Savings. The estimated potential total saving for paste blanket fuels relative to the cost of solid blanket fuels (see Table IV) is approximately 20% of the total cost of solid blanket fuels, including the cost of fuel handling and coolant inefficiency. However, because of the uncertainty of the estimates, it can only be stated that a significant saving is likely.

III. EXPERIMENTAL WORK IN DEVELOPMENT OF URANIUM NITRIDE-SODIUM PASTE FUEL

After concluding the above evaluation of fluid fuels for breeder reactor blankets and selecting UN-sodium pastes as the most attractive fluid fuel, we thought it desirable to obtain preliminary experimental data to strengthen the conclusions. Information was particularly desirable on (1) means of preparing high-solids suspensions of UN in sodium, (2) flow properties of high-solids pastes, and (3) compatibility of sodium-vehicle pastes containing UN, UC, or UO_2 with typical containment materials. Some of these projects were also of interest to development of solid fast-reactor fuels. The work that has been done on these projects is summarized below.

A. Preparation of Uranium Mononitride-Sodium Suspensions

1. Preparation of High-density UN Particles

It is important to have a high uranium density in the blanket fuel to minimize the ratio of capture in sodium and structure to capture in the uranium. However, the classical method for preparing UN produces a powder product of low bulk density. In the classical method,²⁴ bulk uranium in the form of turnings is nitrided at 800-900°C to produce $\text{UN}_{1.75}$. The higher nitrides are decomposed by heating under vacuum at 1100-1400°C. The formation and decomposition of the higher nitrides result in low-density powder of only 4-5 g/cc, about 30-35% of theoretical density. If the entire reaction were carried out at high temperatures, no higher nitrides would form; however, the impervious layer of UN formed on the surface would prevent further reaction.

A technique for reacting uranium and nitrogen was needed that would result in UN of high bulk density suitable for preparation of a blanket paste fuel. In the procedure developed, the furnace for the nitridation

reaction consisted of a 6-in.-diam Mullite combustion tube fitted with a vacuum-tight flange and heated by silicon carbide heating elements. The uranium charge (usually 300-1000 g) was contained in a tungsten crucible. Uranium shot of about -14 +25 mesh (U.S. Standard) size and 20-mil-thick uranium turnings were both satisfactory feed material in these experiments. However, most of the experiments were done with uranium shot, and only such experiments are reported here. The nitridation reaction was carried out at 800-1350°C. The main concern was to optimize the time-temperature relationship within this temperature range to yield the maximum bulk density in a reasonable processing time. To avoid sintering, the charge was heated under nitrogen, rather than an inert atmosphere, from the beginning of the run. The extent of reaction was followed semi-quantitatively by recording the decrease in nitrogen pressure. Additional nitrogen was added intermittently to maintain a pressure of 200-500 Torr during the reaction.

The bulk density of the final product is a function of the time-temperature cycle used during nitridation, as is shown in Table V. In Runs UN-6, -7, and -9, in which the bulk of the nitridation took place below 1100°C, considerable time was required to evacuate the furnace tube at the end of nitridation, indicating evolution of nitrogen from the decomposition of higher nitrides. The bulk density of the products of these runs was low, although somewhat higher than is obtained in the standard preparation procedure for UN.

TABLE V. Run Conditions and Product Analysis for Nitridation of Uranium Shot
(Uranium charge consisted of 300-1000 g of -14 +25 mesh shot;
200-500 Torr nitrogen pressure)

Run No.	Time (hr) at Indicated Temperature (°C)				Time to Evacuate ^b to 0.05 Torr (hr)	Chemical Analysis (w/o)		Bulk Density (g/cc)	Product Agglomeration (%)
	700-1100 ^a	1100-1150	1150-1250	1250-1350		Nitrogen ^c	Oxygen		
UN-4 ^a	0.5	0.1	0.6	7.0	0.3	5.50	0.10	7.1	70
UN-6	5.0	0.1	0.4	1.8	1.5	5.51	0.15	-	20
UN-7	9.0	0.1	2.0	0.3	2.7	5.35 ^d	0.048	5.8	<5
UN-9	3.1	0.3	2.7	1.3	1.3	4.73 ^e	0.04	6.5	30
UN-10	2.1	0.3	2.4	1.6	0.3	2.8 ^f	0.6	7.3 ^f	40
UN-11	1.3	0.7	1.0	2.5	0.3	-	-	-	0
UN-13	1.8	0.7	1.0	7.2	0.3	-	0.44	7.9	0
UN-14	1.3	0.6	1.8	1.3	0.3	5.31 ^e	0.06	8.3	0

^aNitrogen added at 1080°C in UN-4; in other runs, the nitrogen was added before heatup.

^bThe evacuation was done at 1350°C with the pressure eventually reaching about 10⁻⁴ Torr.

^cStoichiometric UN contains 5.55 w/o nitrogen.

^dNitrogen analysis believed to be erroneously low.

^eNitrogen analysis of sample of ground material.

^fThe very low nitrogen analysis indicates that the product contains free uranium; if true, this would affect the bulk density of the powder.

In Runs UN-4, -10, -11, -13, and -14 (see Table V), in which the bulk of the nitridation took place above 1150°C, the time required to evacuate the furnace tube at the end of a run was only 15 or 20 min, about the same as without a charge. This indicates that very little of the higher nitrides formed. The density of the products of these runs was comparatively

100 μ

108-8901

Fig. 2. Photomicrograph of Uranium Mononitride from Run UN-4. See Table V for conditions.

tends to inhibit further reaction of the uranium metal with nitrogen. This is shown in Fig. 3, a photomicrograph of the product of Run UN-14, which was not completely converted to UN. It is estimated that 10-15 hr of nitridation would be required for the complete conversion to UN of the uranium shot used in these experiments (-14 +25 mesh).

In many of the earlier runs, a sizable fraction of the product was sintered together into a single agglomerate at the bottom of the crucible. Photomicrographs of the particles indicated that the sintering was caused by molten uranium leaking through the nitride skin formed on the particles. Subsequently, raising the temperature slowly ($\sim 70^\circ\text{C/hr}$) from 1100 to 1150°C, through the melting point of uranium, avoided agglomeration of the product. This method was successfully applied in Runs UN-11, -13, and -14. In Run UN-7, nitridation was far advanced before the melting point of uranium was reached.

high, as indicated in the table. (Figure 2 is a photomicrograph of the product from Run UN-4.) As was typical of all these runs, the spheres that had been charged were still intact, although somewhat distorted.

In the procedure used in these experiments, dense UN rather than higher nitrides is probably produced initially because of the presence of metallic uranium. If the reaction continues at less than 1200°C, the formation of higher nitrides causes disruption of the face-centered cubic lattice structure of UN, thereby creating voids. On the other hand, if the temperature is raised to about 1200°C when the nitrogen-to-uranium ratio in the product is about 0.5-0.7, no higher nitrides are formed.

A problem arising in this procedure is that the dense UN layer around the particles

200 μ

108-8898

Fig. 3. Photomicrograph of Uranium Mononitride from Run UN-14. See Table V for conditions. Outer layer is UN; inner area is unreacted uranium.

The nitrided product, which retained the form of the original shot, was ground in a ball mill, usually for 24 hr. The material was charged into a steel milling jar, which was sealed with a rubber gasket. This loading was done in a helium glovebox, so that the jar was filled with helium at the beginning of grinding.

Partial oxidation of UN during grinding had a marked effect on the bulk density of the ground product, as shown in Table VI. A high oxygen content in itself did not result in a low-density product, as evidenced by Runs UN-10 and -13. The extent of oxidation during grinding (or, at least, oxidation taking place after nitridation) appears to be the important factor. In Run UN-9A, only a small amount of oxidation (0.04-0.23 w/o oxygen) occurred during grinding. The bulk density of the ground product, 8.2 g/cc, was the highest ob-

tained, although the unground material (the nitrided product of Run UN-9) had a rather low bulk density. In Run UN-9B, the second half of the nitrided

TABLE VI. Effect of Oxidation during Grinding of Bulk Density of Uranium Mononitride

UN formed by nitriding uranium shot was ground in a ball mill for 24 hr to 50-100 μ median size

Oxygen Content					Bulk Density				
(w/o)					(g/cc)				
Not					Not				
Run No.	Ground	Ground	Ground	Ground	Run No.	Ground	Ground	Ground	Ground
UN-7	0.05	0.39	5.8	7.4	UN-10	0.6	1.9 ^a	7.3	6.6
UN-9A ^b	0.04	0.23	6.5	8.2	UN-13 ^c	0.44	0.6 ^a	7.9	7.9
UN-9B ^b	0.04	0.8 ^a	6.5	3.0					

^aThe oxygen content varied considerably between different samples of the same batch of material.

^bThe product of UN-9 was ground in two separate batches. The second batch (UN-9B) was ground in a mill that was not gas-tight.

^cThe product of UN-13 was ground for a total of 72 hr in three 24-hr periods. The extended grinding time was necessary to reduce the product to 50-100 μ size.

product of Run UN-9 was ground in a different milling jar, which apparently was not gas-tight. The oxidation to 0.8 w/o oxygen, which occurred during grinding, resulted in a bulk density for the ground product of only 3.0 g/cc or 37% of the bulk density obtained in Run UN-9A, made with the same batch of feed material. The deleterious effect of oxidation during grinding on the bulk density of UN powder, which was noted in Run UN-9B, is corroborated by the results for Runs UN-10 and -13, although the effect was not as obvious.

Grinding of dense UN, such as the product of UN-13, in a gas-tight ball mill probably would result in a product of higher bulk density than has been obtained thus far.

2. Wetting of Uranium Ceramics by Liquid Sodium

The wetting properties of the vehicle for the suspended solids is important in the sustained operation of a blanket paste fuel. Without wetting of the solids by the vehicle, a paste cannot be prepared; the liquid is excluded from the interstices and the solids either sink to the bottom or float on the top of the liquid. Dewetting of the blanket solids during operation in the reactor would be disastrous.

The wetting properties of liquid sodium for uranium ceramics were studied in a helium glovebox using an apparatus for determination of contact angles by the sessile-drop method.²⁵ The equipment used was quite simple. A plaque of the ceramic material was heated on a hot plate in the glovebox; a drop of distilled sodium was placed on the plaque and then photographed by means of a camera and a low-power telescope. The image was captured on Type 55 Polaroid film. Sample temperature was measured using a Chromel-Alumel thermocouple in contact with the plaque or on the hot-plate surface next to the plaque; temperature control was about $\pm 10^\circ\text{C}$. Although the atmosphere in the glovebox was good (oxygen, <10 ppm; nitrogen, <20 ppm; moisture, ~ 1 ppm or less), a drop of molten sodium would nevertheless become heavily coated with oxide in 6 or 7 min.

Experiments were carried out with plaques of UO_2 , UP, US, UC, and UN, which were prepared by hot-pressing or arc-melting; the surfaces were hand-polished on 600-grit paper. The contact angles were determined by visual examination of the photographs and, for reasonably flat ceramic surfaces, the contact angle was calculated from the drop parameters using a method developed for surfaces of revolution by Mack and Lee.²⁶ Table VII gives the results.

More refined apparatus probably would have resulted in better values; however, the large contact angles indicated that the wetting by molten sodium under these conditions was extremely poor. Indeed, the results obtained all fell within the limits found by Bradhurst and Buchanan²⁷

TABLE VII. Contact Angles Determined by Sessile-drop Method for Liquid Sodium and Uranium Ceramics

Ceramic	Temperature (°C)	Contact Angle (°)	
		θ_{visual}	$\theta_{\text{calc}}^{26}$
UO ₂	210	130 ± 2	130.6
UP ^a	260	130 ± 2	129.9
US	240	150 ± 4	-
UC	250	144 ± 1	143.6
UN	255	157 ± 8	-

^aContained some U₃P₄.

ceramic is first completely immersed in liquid sodium, and then the force required to raise the sample from the sodium bath is measured; from the known dimensions of the sample and the surface tension of sodium, the contact angle can then be calculated. For a slurry or paste system, in which the ceramic particles are completely immersed in molten liquid metal, this contact angle probably would be more meaningful than the sessile-drop values, which represent the advancement of a molten metal over a porous surface covered with a foreign gas, as in our experiments.

The ease of formation of a stable paste was also used as a practical indication of wetting of the solids by the vehicle. A 50 v/o UO₂-sodium paste was readily prepared at 160°C, thus indicating wetting at this temperature. Uranium mononitride, which had the largest contact angle with sodium (as shown in Table VII), was suspended by sodium vehicle at 250°C. However, the paste did not immediately form with UN powder (44 to 74 μ particle size); about an hour of stirring was required. At 300°C, a half-hour of intimate contact and stirring was sufficient to form a paste. The constituents of the pastes did not separate out on cooling; the solidified pastes appeared quite grainy, but the particles all were coated with sodium. On treatment of the paste with ethyl alcohol, black particles of UN settled out as the sodium dissolved away. This powder was found by X-ray diffraction analysis to be identical to the initially charged UN. There appeared to be no difference in wetting of UN prepared by the usual hydriding route³⁰ or that prepared by direct nitridation of uranium turnings.

These observations indicate that a surface reaction that promotes wetting takes place between the sodium and the UN. It is not clear whether this process is the removal of some interfering substance from the interface by sodium or the formation of some compound, which in turn is readily wetted by molten sodium. However, such mechanisms could explain the disparity between contact angles measured by the sessile-drop and vertical-plate methods.

for UO₂ in contact with sodium of varying oxygen content at 250°C (130° for oxygen-saturated sodium, and 160° for oxygen-free sodium). Bradhurst and Buchanan also used the sessile-drop method.

Another technique of determining contact angles, the vertical-plate method,²⁸ gives different results for UO₂ and sodium; good wetting is indicated.²⁹ In this approach, the

3. Mixing of Uranium Mononitride-Sodium Paste Constituents

Following the preliminary wetting experiments described above, a UN-sodium paste of high uranium content was prepared. The UN powder of Run UN-9A (see Table VI for product characteristics), which had a bulk density of 8.2 g/cc and a median diameter of about 100 μ , was mixed with sodium at 300°C to obtain 346 g of paste containing 49 v/o solids (6.6 g/cc of uranium). The paste contained 8 v/o gas, as determined by difference from the amounts of sodium and UN added.

At 40 v/o solids, an intermediate stage in the preparation, the paste was judged to have good flow properties. However, at its final composition (49 v/o solids) the paste was quite viscous and probably not directly suitable for a flowing system. Pastes with higher uranium densities or better flow characteristics could be prepared from denser UN powder.

B. Flow Properties of High-solids Suspensions

In this investigation, equations were developed from theoretical considerations and experimental data that relate paste viscosity to physical properties of the liquid and solid.

Accurate knowledge of paste flow properties would be essential for the detailed design of a paste-fuel system. Flow blockage must be avoided. Other complications that may result from the movement of high-solids suspensions are non-Newtonian behavior, particle agglomeration, and sintering. The difference in flow between the solids and the liquid can vary from the condition of a packed bed, in which the solids are static while the liquid flows, to the condition of a homogeneous fluid. For sodium-vehicle pastes containing 100-200 μ particles, the flow condition is between these extremes. For that case, the excess liquid flowrate (difference in flowrate between the liquid and the particles) is a complex function of the flow-channel characteristics including length-to-diameter ratio, the radius of the pipe-line bends, and the presence of restrictions such as orifices. Flow of this type paste has been studied at Atomic Power Development Associates, Inc.^{20,21,31,32}

Pastes containing fine particles of less than 50 μ have such low excess liquid flowrates that they behave essentially as homogeneous liquids of non-Newtonian viscosity. This type suspension was chosen for study in this investigation. The investigation was further restricted to suspensions of high-solids concentrations such that liquid and solids would not separate during long periods of settling. The fine-particle suspensions of this investigation were stable over a solids concentration ranging from the maximum concentration attainable by centrifugation to several percent less than that concentration.

Flow was measured in pipe-line viscometers of a unique design that minimized entrance and exit effects. Experimental flow data were obtained for suspensions consisting of nickel, alumina, copper, or glass solids in sodium, xylene, or glycerine vehicles with solids concentrations of 28-55 v/o. The basis for the correlation of the data was on analytical investigation of the flow behavior that considered the particle-particle interactions that take place in a settled suspension. Equations were developed for evaluating the flow behavior of high-solids suspensions from the physical properties of the liquid and solid components. The correlation equations fit all the systems investigated. These equations take into account the effects of liquid viscosity, liquid and solid densities, particle size, size distribution, particle surface areas, volume fraction of solids in the suspension, and volume fraction of solids at maximum settled conditions. A technique that involves the use of the correlation equations was developed for calculating suspension flowrates in pipe lines as a function of pressure drop.

The experimental work is detailed in Gay's doctoral thesis³³ and also in the open literature;³⁴ the latter contains the most recent correlation of the data.

C. Compatibility of Type 304 Stainless Steel with Sodium-paste Reactor Fuels

Section II.B discussed the problems of compatibility of the paste with the cladding. The likelihood of compatibility of UN-sodium pastes with common container materials, such as stainless steel, was one of the reasons for selecting UN as the fertile material. A brief experimental study was undertaken to confirm this choice and to test other uranium ceramic and sodium pastes.

Static corrosion tests were performed to determine the compatibility of Type 304 stainless steel with sodium pastes prepared from each of the leading ceramic fuel materials-- UO_2 , UC, and UN. The pastes were prepared by heating ACS reagent-grade sodium in a Type 304 stainless steel corrosion capsule together with weighed amounts of the ceramic materials; all manipulations were performed in a helium glovebox. A degreased coupon of Type 304 stainless steel was then inserted into each capsule, and the corrosion-test capsules were welded closed in the helium glovebox. All the corrosion-test capsules were held at 1400°F (760°C) for 1000 hr in a resistance-heated furnace and allowed to cool in the furnace. The capsules were then opened in the helium glovebox; the corrosion coupons were removed, washed with butanol, and examined by X-ray diffraction, electron microprobe, and metallographic techniques. Portions of the test coupons were submitted for chemical analyses. Table VIII presents corrosion-test data obtained in these experiments.

TABLE VIII. Results of Static Corrosion Tests on Type 304 Stainless Steel Coupons Immersed in Sodium-Ceramic Pastes for 1000 hr at 1400°F

Contents of Capsule	Fraction of Ceramic in Paste (w/o)	Coupon Weight Change (mg/cm ²)	Coupon Thickness Increase (%)
Na ^a	0	0.03	0
Na	0	0.06	3
Na + Oxygen (4000 ppm) ^a	0	0.4	10
Na + UO ₂ ^a	90	0.2	0
Na + UN ^a	92	-0.3	0
Na + UN + Oxygen (4000 ppm) ^a	92	-0.3	0
Na + UC-I ^{a,b}	88	0.9	20
Na + UC-II ^c	93	<1.0	3
Na + UC-III ^d	92	0.2	2
Na + UC-III ^d	92	0.3	3

^a5-mil-thick coupons used; other samples, 10-mil coupons.

^bHyperstoichiometric UC: 5.24 w/o C, 0.18 w/o O, 0.019 w/o N; prepared by liquid-metal process;⁹ 45 μ median particle diameter.

^cStoichiometric UC: 4.8 w/o C, 0.26 w/o O, 0.008 w/o N; prepared by carbothermic reduction; ~500 μ median particle diameter.

^dHypostoichiometric UC: 4.6 w/o C, 0.14 w/o O, 0.003 w/o N, prepared by high-intensity arc;¹⁰ 130 μ median particle diameter.

1. Control Experiments without Ceramic Fuels

Visual examination of the stainless steel coupons in contact with sodium alone showed no apparent etching or discoloration of the metal. The carbon content of the 5-mil coupon in contact with sodium was 820 ppm, whereas that of the 10-mil coupon was 1000 ppm; the as-received carbon contents of the coupons were 560 and 750 ppm, respectively.

Visual examination of the coupon in contact with the sodium that contained oxygen (added as Na₂O) indicated a greenish-gray deposit on the surface. X-ray diffraction examination showed this deposit was probably α -NaFeO₂. Corrosion of iron by oxygen-contaminated sodium has been reported by Horsley³⁵ to proceed by formation of a double oxide 2Na₂O·FeO₂. Metallographic examination of this coupon showed considerable intergranular corrosion to a depth of about 0.6 mil; carbides were observed to be precipitated at the grain boundaries (sensitized structure). The coupons in contact with only sodium had the characteristic sensitized structure resulting from heating to the temperature of these experiments.

The lack of reaction of the Type 304 stainless steel with the sodium was expected, as was the intergranular corrosion observed in the added oxide experiments. These tests served as a control for the experiments described below.

2. Sodium-Uranium Dioxide Paste

The UO_2 used in this experiment was hydrogen-reduced at 800°C prior to use; the final O/U ratio was 2.007. Visual examination of the coupon from this experiment showed no etching or discoloration of the metal. Metallographic examination revealed a sensitized structure, such as that observed for the coupon heated in sodium alone, and gave no indications of corrosive attack. A slight increase in hardness was found for the sample compared with the value for the coupon in sodium alone.

3. Sodium-Uranium Nitride Pastes

The uranium nitride used in these pastes was prepared by the method described in Section III.A.1 above. Chemical analysis of this ceramic gave 4.73 w/o nitrogen and 0.23 w/o oxygen. X-ray examination of this material showed the major phase to be UN with minor amounts of $\beta\text{-U}_2\text{N}_3$.

To the unaided eye, the stainless steel coupons heated in these pastes showed some apparent etching of the metal. Metallographically, the coupon heated in the paste, with oxygen added as Na_2O , showed no sensitization, merely the characteristic twinned structure of the as-received austenitic steel. The sample from the paste without added oxygen had the typical sensitized structure. There was no apparent corrosive attack of either coupon, although there were small pits distributed in the coupon heated in the paste with added oxide. These pits may have resulted from tearing out of brittle inclusions during polishing or possibly from etching. Chemical analysis revealed a slight decrease in average chromium content for the coupon immersed in the paste with added Na_2O : 15.8 w/o chromium was obtained for the coupon from the UN-Na + oxide paste, whereas 16.7 w/o was obtained for the coupon heated in the UN-sodium paste without added oxide, 16.6 w/o for the coupon immersed in sodium alone, and 16.8 w/o for the as-received material.

Corrosion of the coupon by the oxygen in the Na-UN paste should be reduced because of interaction of oxygen with UN to form UO_2 . Formation of a layer of UO_2 on the ceramic particle surface could hinder release of nitrogen from U_2N_3 and thus might explain the lower nitrogen content of this coupon (280 ppm) compared to the coupon in the paste without the added oxygen (1100 ppm). No reaction layers were observed metallographically on the coupons, and microprobe analysis showed no significant variation of iron, chromium, or nickel content at the surface. The reason for the lack of sensitization in the one coupon is not clear.

4. Sodium-Uranium Carbide Pastes

Visual examination of the coupons after completion of the corrosion tests revealed grayish, etched surfaces on all the coupons; the etching

appeared most severe for the coupon in the UC-I paste. All the coupons had become magnetic except for those in contact with the UC-III paste. X-ray examination showed the presence of the α -iron structure in the magnetic coupons; the nonmagnetic coupons still had the γ -iron structure and also had some sigma phase present as a minor constituent.

Metallographic examination of the cross sections of these coupons was in essential agreement with the X-ray results. The coupon from the UC-I paste showed massive precipitation of carbides throughout the material, and a reaction layer was observed at the surface. The coupons from the UC-II and UC-III pastes were similar to each other in appearance: a reaction layer at the surface, precipitates in the grain boundaries, and Widmanstätten structures within the grains.

Microhardness tests of the metallographically mounted coupons showed a variation in agreement with the apparent carbon content: a large increase in hardness for the UC-I sample, a lesser increase for the UC-II sample, and little change for the UC-III sample.

Electron-microprobe analyses of the coupons revealed that the grain structure of the coupon in contact with UC-I paste was rich in chromium, probably as chromium carbides, with a decrease in iron and nickel. The outer portions of the coupon's cross section contained an iron-rich area (over 90% iron, ~2-3% nickel, no detectable chromium), which probably gave the coupon its magnetic character. The surface reaction layer that was observed metallographically apparently corresponds to very chromium-rich particles, most likely carbides. The coupon in contact with the UC-II paste showed areas with carbon and chromium associated probably as carbides, and an enrichment of chromium at the surface. The other coupons showed no significant variations in composition.

The carbon increase (in mg/cm²) for these coupons after the corrosion tests were completed was: UC-I, 0.77; UC-II, 0.61; UC-III, 0.40. The data showed essentially a linear increase in carbon content of the coupons as the "equivalent carbon"³⁶ content of the UC increased. The changes in carbon content found by chemical analyses are consistent with the other observations reported above.

5. Summary of Corrosion Data

In the static corrosion tests described above, sodium pastes of UO₂ and UN were compatible with Type 304 stainless steel at 1400°F for the test period of 1000 hr. With the carbide pastes, although significant carburization of the steel did occur in the paste of a very hyperstoichiometric carbide, carburization by the carbide pastes was no more severe than reported in the literature for sodium-bonded carbide pellets.³⁷ The use of hypostoichiometric carbides in paste fuels could reduce the carburization of Type 304 stainless steel to quite acceptable levels.

IV. DISCUSSION AND CONCLUSIONS

The evaluation of various fluid blanket fuels indicates that a paste of UN particles in a sodium vehicle is the most attractive. This evaluation is based on uranium density, heat transfer, and compatibility with common container materials. The use of UC particles would be as attractive as that of UN particles, except for the greater likelihood of reaction of UC particles with the container.

To demonstrate the feasibility of this concept, a UN-sodium paste of 49 v/o solid was prepared. It was shown that a high-density UN powder for the paste can be prepared by direct reaction of nitrogen with uranium turnings or shot followed by crushing of the reaction product. A stable paste is formed simply by heating the UN in contact with sodium for 30 min at 300°C. The bulk density of the powder and, thus, the solids volume fraction of the paste might be improved by the use of selected size fractions as in fabrication of solid vibratory-compacted elements. By this technique, a smooth paste may be possible with solids fraction of 60-70 v/o.

Corrosion experiments corroborated the evaluation-study conclusion that UN-sodium pastes are compatible with common container materials, specifically Type 304 stainless steel. Although hyperstoichiometric UC in sodium carburized Type 304 stainless steel, the use of stoichiometric or slightly hypostoichiometric UC might circumvent this problem.

Experimental measurement of flow properties of high-solids pastes further supports the fuel concept selected in the evaluation study. To avoid significant difference in the flow velocity of the vehicle and the solids, the particles should be no more than about 50 μ in diameter. The use of fine particles and smooth flow passages would be beneficial in avoiding flow blockage. Heat-transfer calculations indicate that the fuel tubes could be as large as 3/4-in. in diameter, provided the plutonium buildup is limited to about 1 w/o of the fissile content.

The use of static fuel assemblies appears to be about as attractive as circulating the fuel. Fission-gas release and bubble formation are not likely to be problems at low burnups. Static fuel would have to be reprocessed more frequently to prevent high plutonium buildup; however, this factor is offset by equipment simplicity. Since the fuel containers can be reused, frequent reprocessing is not as expensive as for solid fuels.

Concerning the economic potential of this fuel concept, the total cost of removing and shuffling fuel assemblies, coolant inefficiency, fuel fabrication, and fuel reprocessing is estimated to be about 0.06 to 0.07 mill/kW-hr less for paste blanket fuels than for solid fuels. This is about 20% of the total of these costs for solid fuels. Static paste fuel and circulated

paste fuel appear to offer about the same savings relative to solid fuel. Though significant error may exist in these estimates, there appears to be ample evidence that a real economic advantage will accrue from the use of paste blanket fuels.

Further work on paste blanket fuel probably should be postponed until more is known about the core fuel, the fuel cycle, and the reactor configuration. For maximum economy, the fabrication and reprocessing of paste blanket fuel should be integrated with the core fuel fabrication and reprocessing. The selection of the solid constituent of the paste, such as UN or UC, may be affected by this consideration and the type of core fuel that is finally selected for fast reactors. The configuration of fast reactors of the future also cannot be predicted. The reactor configuration is important in determining the usefulness of paste blanket fuel, since this type of fuel would be most advantageous for reactors with large radial blankets. (Paste fuel is not practical for the axial blankets.) In the future, the savings of a few hundredths of a mill per kilowatt-hour by the use of paste blanket fuel may be important for a large fast-reactor economy. At present, other work on core fuel fabrication and reprocessing appears to be more important.

ACKNOWLEDGMENT

We wish to thank Dr. A. D. Tevebaugh for consultation on all aspects of this program and Mr. C. F. Lehmann for technical assistance.

REFERENCES

1. D. Okrent, *Nuclear Considerations in the Selection of Materials for Fast Power Reactors*, Nuclear Metallurgy, No. 12 Special Report Series, Vol. IX, L. R. Kelman, Ed., AIME, p. 1 (1963).
2. S. Glasstone and A. Sesonske, *Nuclear Reactor Engineering*, Van Nostrand, p. 705 (1963).
3. S. A. Hasnain and D. Okrent, *On Design and Management of Fast Reactor Blankets*, Nucl. Sci. Eng. 9, 314 (1961).
4. *An Evaluation of Four Design Studies of a 1000-MWe Ceramic-fueled Fast Breeder Reactor*, COO-279, Chicago Operations Office, USAEC (1964).
5. *Large Fast Reactor Design Study*, ACNP-64503, Allis Chalmers, Atomic Power Development Associates, Inc., Babcock & Wilcox Company (1964).
6. *Liquid Metal Fast Breeder Reactor Design Study*, WCAP-3251-1, Westinghouse Electric Corporation (1964).
7. *Liquid Metal Fast Breeder Reactor Design Study*, CEND-200, Combustion Engineering, Inc. (1964).
8. *Liquid Metal Fast Breeder Reactor Design Study (1000-MWe UO₂-PuO₂ Fueled Plant)*, GEAP-4418, General Electric Corporation (1964).
9. L. E. Link et al., *1000-MW(e) Metal-fueled Fast Breeder Reactor*, ANL-7001 (1966).
10. C. Williams and F. T. Miles, *Liquid Metal Fuel Reactor Systems for Power*, Nucleonics 12, No. 7, 11 (1954).
11. N. F. Eaton and G. W. Greenwood, *Mass Transfer and Plug Formation during the Circulation of Thorium Bismuthide Slurry by Convection*, AERE-M/R-2144, Atomic Research Establishment (1957).
12. R. I. Hawes, G. W. Horsley, and R. Sheldon, *Behavior of Soluble Slurries*, AERE-M/R-2529, Atomic Energy Research Establishment (1958).
13. G. H. Broomfield, *Mass Transfer and Plug Formation during the Circulation of Thorium Bismuthide by Convection. Pt II. Uninhibited System*, AERE-M/R-2144A, Atomic Energy Research Establishment (1958).
14. B. R. T. Frost, *Problems of Liquid-metal-fueled Reactors*, Nucl. Eng. 1, 334 (1956).
15. J. J. Bulmer et al., *Fused Salt Fast Breeder*, CF-56-8-204 (Del.), Oak Ridge National Laboratory (1956).
16. R. P. Hammond, R. E. T. Stanford, and J. R. Humphreys, Jr., *Mobile Fuel Plutonium Breeders*, LA-2644, Los Alamos Scientific Laboratory (1961).
17. A. E. Klickman et al., *Design and Economic Evaluation of Mobile Blankets for Fast Reactors*, APDA-160, Atomic Power Development Associates, Inc. (1964).
18. F. G. Hammitt, E. C. Kovacic, and F. J. Leitz, *The Fission Gas Problem for Mobile Fuel Fast Reactors*, Nucl. Sci. Eng. 7, 327 (1960).
19. J. B. Melehan and J. E. Gates, *In-pile Fission-gas Release from Uranium Carbide and Uranium Nitride*, BMI-1701, Battelle Memorial Institute, Columbus, Ohio (1964).

20. E. C. Kovacic *et al.*, *A Paste Concept for Fast Reactor Blanket Applications*, APDA-172, Atomic Power Development Associates, Inc. (1965).
21. H. V. Chamberlain *et al.*, *Summary Report on Piped Particulate Fuel Concept of Fast Reactor Core and Blanket Applications*, APDA-192, Atomic Power Development Associates, Inc. (1967).
22. M. Jakob, *Heat Transfer*, Vol. 1, p. 85, John Wiley and Sons, Inc., New York (1949).
23. Ref. 8, pp. 8-9.
24. E. O. Speidel and D. L. Kelber, *Fabrication and Properties of Hot-pressed Uranium Mononitride*, BMI-1633, Battelle Memorial Institute, Columbus, Ohio (May 1963).
25. W. D. Kingery and M. Humenik, Jr., *Surface Tension at Elevated Temperatures. I*, J. Phys. Chem. 57, 359 (1953).
26. G. L. Mack and D. A. Lee, *The Sessile Drop Method for Obtuse Angles*, Ibid. 40, 169 (1936).
27. D. H. Bradhurst and A. S. Buchanan, *Surface Properties of Liquid Sodium and Sodium-Potassium Alloys in Contact with Metal Oxide Surfaces*, Australian J. Chem. 14, 397 (1961).
28. C. C. Addison, D. H. Kerridge, and J. Lewis, *The Surface Tension of Liquid Sodium: Vertical-plate Technique*, J. Chem. Soc., 2861 (1954).
29. B. R. T. Frost, *Advances in Liquid Metal Technology*, J. Nucl. Mater. 7, 109 (1962).
30. P. E. Evans and L. J. Davies, *Uranium Nitrides*, Ibid. 10, 43 (1963).
31. W. C. Chon, E. C. Kovacic, and F. G. Hammitt, *Particle-liquid Cocurrent Downward Flow through a Tube with a Restricting Orifice End for Application to a Mobile Fuel Nuclear Reactor System*, Nucl. Sci. Eng. 13, 65 (1962).
32. W. G. Blessing *et al.*, *Conceptual Design of a 300-MWe Paste-fueled Fast Breeder Power Reactor*, APDA-146, Atomic Power Development Associates, Inc. (1961).
33. E. C. Gay, *Flow Properties of Liquid Sodium Suspensions with High Solid Concentrations*, Ph.D. thesis, Washington University (1967).
34. E. C. Gay and P. A. Nelson, *Flow Properties of Suspensions with High Solids Concentrations*, Paper 22b presented at the American Institute of Chemical Engineers National Meeting, St. Louis, Missouri, February 18-21, 1968 (also to be published in the AIChE Journal).
35. G. W. Horsley, *Corrosion of Iron by Oxygen-contaminated Sodium*, J. Iron and Steel Inst. 182, 43 (1956).
36. W. R. Kerr, *Comparison of Microstructures and Chemical Carbon Content for Hypostoichiometric Uranium Monocarbide*, NAA-SR-TDR-8943 (1965).
37. P. E. Elkins, *Compatibility of Uranium Carbide Fuels with Cladding Materials*, NAA-SR-7502 (1964).