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SP-100 COATED-PARTICLE FUEL DEVELOPMENT

PHASE I, FINAL REPORT

**by
GA PROJECT STAFF**

MASTER

**Prepared under
Contract DE-AT03-82SF11690
for the San Francisco Operations Office
Department of Energy**

MARCH 1983

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CONTENTS

SP-100 COATED-PARTICLE FUEL DEVELOPMENT

	<u>Page</u>
1.0 INTRODUCTION	
1.1 SP-100 Reference and Alternative Fuel Module Concepts	1-2
1.2 Need for an Alternative Fuel	1-2
2.0 OBJECTIVES AND SCOPE OF COATED-PARTICLE FUEL PROGRAM	
2.1 Program Description	2-1
2.2 Objectives	2-1
2.3 Critical Issues	2-2
3.0 COATED-PARTICLE FUEL DESIGN	
3.1 Design of ZrC Coated Particles	3-5
3.1.1 Results of Irradiations of Experimental Coated Particles	3-5
3.1.2 Methods of Calculating Stresses Generated in ZrC Particle Coatings	3-7
3.1.2.1 Gas Pressure in the Particle	3-12
3.1.2.2 Stress in the ZrC Layer	3-14
3.1.2.3 Calculated Stresses in the Experimental Coated Particles	3-16
3.1.2.4 Design of Particles for a Space Power Reactor	3-18
3.2 Design of Tungsten-Coated Particles	3-24
3.2.1 Pressure in the Particle	3-24
3.2.2 Stress and Strain in Tungsten Coating	3-25
3.2.3 Analysis of Creep Data for CVD Tungsten	3-26
3.2.3.1 Creep Rate	3-29
3.2.3.2 Creep Rupture	3-30
3.2.4 Prediction of Particle Behavior	3-31
References of Section 3	3-37

4.0 COATED-PARTICLE FUEL FOR SPACE POWER SYSTEMS

4.1 Materials Compatibility	4-1
4.2 Fuel Properties	4-8
4.2.1 Oxide and Cermet Fuels	4-14
4.2.2 Carbide Fuels	4-16
4.2.3 Nitride Fuels	4-19
4.2.4 Comparison of Candidate Fuels	4-20
4.3 Fuel Kernel Fabrication	4-21
4.4 Fuel Compact Fabrication	4-28
References of Section 4	4-30

5.0 CORE DESIGN WITH COATED PARTICLES

5.1 Neutronic Analysis	5-1
5.2 Thermal Analysis	5-5
5.3 Core Structural Analysis	5-19
5.4 Safety Implications	5-24
5.4.1 Criticality When Immersed in Water	5-24
5.4.2 Immersion Solubility and Dispersion	5-25
5.4.3 Launch-Pad Fire and Explosion	5-25
5.4.4 Re-entry Disassembly and Burnup	5-26
References of Section 5	5-27

6.0 SUMMARY AND CONCLUSIONS

6.1 Summary of Findings	6-1
6.2 Critical Issues	6-3
6.2.1 Fabrication of UC Coated Particles and Fuel Compacts	6-4

	<u>Page</u>
6.2.2 Irradiation Testing of UC Coated Particles	6-4
6.2.3 Measurements of Fuel-Particles and Compact Properties	6-4
6.2.4 Thermal Stability of Coated Particles	6-5
6.2.5 Immersion Reactivity Control	6-5
6.2.6 Chemical and Aerodynamic Burnup	6-6

List of Figures

Page

1-1	SP-100 Heat Pipe Fuel Module Concepts: LANL Reference Design and Alternate Design	1-3
1-2	Microphotograph of a TRISO Fuel Particle	1-5
3-1	Fuel Loading Using Particles of Two Sizes	3-2
3-2	Schematic of a Proposed Fuel Particle for the Space Power Reactor	3-4
3-3	Representative (8Th, 1U)O ₂ Particles Coated with ZrC Before Irradiation	3-8
3-4	Representative (8Th, 1U)O ₂ Particles Coated With ZrC After Irradiation	3-9
3-5	HRB-15 Fuel Particle Before Irradiation	3-10
3-6	HRB-15 Fuel Particle After Irradiation	3-10
3-7	Coated LiAlO ₂ Coated Particle Before and After Irradiation . . .	3-11
3-8	Calculated Stresses in the ZrC Coatings of Irradiated Particles .	3-17
3-9	End-of-Life Stress in the ZrC Layer on the Large Particle as a Function of Coating Thickness and Kernel Porosity	3-19
3-10	End-of-Life Stress in the ZrC Layer as a Function of the Volume Fraction of Fuel	3-20
3-11	Fuel Volume in the Two-Particle Compact as a Function of the Size of the Small Particle	3-22
3-12	Comparison of the Minimum Rates for CVD and PM Tungsten	3-27
3-13	Comparison of the Creep-Rupture Properties of CVD and PM Tungsten	3-28
3-14	Larsen-Miller Plot of Tungsten Stress-Rupture Values	3-32
3-15	End-of-Life Coating Diameter Strains in Tungsten Coated Particles	3-34
3-16	End-of-Life Coating Stresses in Tungsten Coated Particles	3-35
4-1	Comparison of the Free Energy of Formation for Selected Carbides.	4-3
4-2	Dependence of Fuel Swelling on Temperature for UC, UN, UO ₂ . . .	4-10
4-3	Schematic Diagram of the Swelling-Gas Release Process to Swelling by Lenticular Cavities	4-11

List of Figures (Cont.)

		<u>Page</u>
4-4	Vapor Pressure of Various Fuels	4-13
4-5	Fuel Swelling Curves for SP-100 Core Design	4-15
4-6	CEA Data on Swelling of UO_2 with Various Initial Densities at 1500°C	4-17
4-7	Thermal Conductivity vs Temperature for Various Fuels	4-18
4-8	Uranium-Carbon-Oxygen Phase Diagram (1500°C)	4-22
4-9	Uranium Oxycarbide Kernels	4-23
4-10	GSP Process for the Fabrication of Uranium Oxycarbide Kernels . .	4-25
4-11	Internal Gelation Process for the Fabrication of Uranium Oxycarbide Kernels	4-26
4-12	Macro-photos of Various Kernels	4-27
5-1	SP-100 Reference Core Geometry	5-2
5-2	Radial Cross Section of the Homogenized SP-100 Reactor	5-3
5-3	Axial Cross Section of the Homogenized SP-100 Reactor	5-4
5-4	A One-Twelfth Symmetric Reactor Sector of SP-100 Reactor	5-7
5-5	Circular Finite-Difference Model of a One-Twelfth Sector of the SP-100 Reactor	5-9
5-6	Estimated Thermal Conductivity Distribution of Oxycarbide Fuel at 1600 K	5-13
5-7	Effective Thermal Conductivity Over the Normal to Faulted Temperature Range	5-18

List of Tables

		<u>Page</u>
3-1	Results of Irradiation of Experimental Coated Particles with Pyrolytic Carbon and Zirconium Carbide Coatings Directly on the Kernel	3-6
3-2	Zirconium Carbide Coated Particle Designs for a Space Power Reactor	3-23
4-1	Coefficients of Thermal Expansion of Candidate Materials	4-6
4-2	Selected Properties of High-Temperature Nuclear Fuels	4-12
5-1	Effective Multiplication Factor (k_{eff})	5-6
5-2	Assumed Composition of Coated-Particle Fuel Compact	5-10
5-3	Materials and Their Effective Thermal Conductivities	5-11
5-4	SP-100 Power Densities in Various Layers	5-14
5-5	Adjusted Power Densities for TAC2D Input	5-15
5-6	TAC2D Results - Maximum Fuel Temperatures	5-17
5-7	Expected Maximum Fuel Temperatures	5-20

1. INTRODUCTION

This document is the final report of Phase I of the SP-100* Coated-Particle Fuel-Development Program conducted by GA Technologies Inc. for the U. S. Department of Energy under contract DE-AT03-82SF11690. The general objective of the study conducted between September and December 1982 was to evaluate coated-particle type fuel as an alternate or backup fuel to the UO₂ tile-and-fin arrangement currently incorporated into the reference design of the SP-100 reactor core. Specifically, Phase I consisted of three tasks to meet that objective, namely, (1) to define design criteria for coated-particle fuels and an initial baseline core design, (2) to determine a preliminary design of a coated particle, and (3) to provide an initial demonstration of fabrication of coated particles suitable for a nuclear space power reactor. The SP-100 reference reactor core to which the coated-particle core is to be compared was developed by the Los Alamos National Laboratory (LANL) starting in 1976 and was defined as its current reference in June 1982**.

This report presents and discusses the following topics in the order listed:

- o The need for an alternative fuel for the SP-100 nuclear reactor
- o An abbreviated description of the reference and coated-particle fuel module concepts
- o The bases and results of the study and analysis leading to the preliminary design of a coated particle suitable for the SP-100 space power reactor
- o Incorporation of the fuel particles into compacts and heat-pipe-cooled modules

* SP-100 as used here refers to the Heat Pipe Reactor Space Power System being developed by Los Alamos National Laboratory.

** D. Buden and J. Stocky, "SP-100 Conceptual Design Description, No. 1," Jet Propulsion Laboratory, June 1982.

- o Initial efforts and plans to fabricate coated-particle fuel and fuel compacts
- o The design and performance of the proposed alternative core relative that of the reference fuel
- o A summary of critical issues and conclusions consistent with the level of effort and duration of the study

1.1 SP-100 REFERENCE AND ALTERNATIVE FUEL-MODULE CONCEPTS

A comparison of the SP-100 reference and alternative fuel module concepts is presented here to aid the reader in understanding the remainder of this report. Both the reference and alternative cores consist of 120-fuel module or "shish kebob" assemblies. The reference SP-100 reactor core is a UO_2 fueled, heat-pipe-cooled array. The heat pipes are composed of Mo-Re alloy pipes containing arterial-screen wicks and Li as the working fluid. Mo fins are located along the reactor core section of the heat pipes to provide good thermal contact between the fuel and the heat pipes. UO_2 fuel tiles are sandwiched between fins.

Both the reference core and an alternative core are formed by assembly of heat-pipe fuel modules as shown conceptually in Fig. 1-1. It can be seen that the UO_2 tile-and-fin stack of the reference core may be replaced by coated-particle fuel compacts of large and small size particles held together by a metal (Mo or W) matrix. Coated particles may contain kernels of UO_2 , UC, or mixtures of UO_2 and UC. Our assessment of the coated-particle fuel in this report is based upon the use of a metallic matrix for the fuel compacts.

1.2 NEED FOR AN ALTERNATE CORE/FUEL DESIGN

The fuel design for the LANL heat pipe reactor is unique and as yet unproven. Considerable concern has been expressed over the effects of fuel swelling on the integrity of the core. While a recent (7/82) evaluation of

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SP-100 HEAT PIPE FUEL MODULE

Fig. 1-1

available swelling data by an independent SP-100 fuel panel has supported the belief that the volumetric fuel swelling should not exceed about fifteen percent, under normal operating conditions, there still remains both some uncertainty in the data and considerable uncertainty about the effects of even limited swelling on core, and especially heat pipe, integrity.

Irradiation tests in EBR-II are planned to help define the effects of fuel swelling as well as other key issues in core performance. While these tests are well designed, and an important part of the overall SP-100 program, larger scale tests, perhaps involving several heat pipes, will eventually be required to fully understand core performance. The complexity of this design in which the fuel is closely mechanically coupled to the remainder of the core system makes it inherently difficult to test.

Any alternative core and fuel design to the LANL heat pipe reactor design should circumvent this integrity concern. The coated-particle fuel concept is uniquely suited to this role. Coated-particle fuels have been developed over the past 20 years both for the NERVA and ROVER programs and for the commercial High Temperature Gas-Cooled Reactor (HTGR) programs in the U.S. and in Europe. The Fort St. Vrain reactor, which has been operating successfully since 1977 on the grid of the Public Service Company of Colorado, contains about 15 tons of coated-particle fuels.

An example of a coated fuel particle of the type developed for the HTGR is shown in Fig. 1-2. This type of particle is generally referred to as a TRISO particle. It has four coating layers of pyrolytic carbon and silicon carbide, each designed for a specific purpose, over a fissile kernel. Many different designs using a variety of kernel and coating materials have been developed.

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THE COATED PARTICLE CONCEPT

1-5

(a) SCHEMATIC OF TRISO-COATED FUEL PARTICLE

(b) SEM MICROGRAPH OF A DELIBERATELY FRACTURED TRISO-COATED FUEL PARTICLE

Fig. 1-2

As will be shown later in this report, the different nature of the SP-100 relative to the HTGR leads to different fuel design requirements that in turn lead to different fuel materials than have conventionally been used. The key characteristics of the SP-100 that influence fuel design are that it is a high temperature, high power density, fast neutron spectrum reactor.

2. OBJECTIVES AND SCOPE OF COATED-PARTICLE FUEL PROGRAM

2.1 PROGRAM DESCRIPTION

The SP-100 coated particle fuel program was initiated to evaluate a second fuel option to the reference SP-100 core development. The decision to investigate coated-particle fuels as an alternative to the reference UO₂ tile-and-fin stack was based on GA's experience with particle fuels as a designer and supplier of such fuels for use in the NERVA, and ROVER projects and in the commercial HTGR at the Fort St. Vrain Nuclear Generating Station in Colorado. The program is divided into two phases. Phase I was a three month effort to assess the potential of particle fuels to meet the requirements of the SP-100 reactors. Phase II efforts as presently defined would establish an alternative design configuration based on particle-fuel technology for a SP-100 reactor and would include fuel development and verification. Some of the testing of coated-particle fuel modules could be integrated into the current irradiation program for the reference core fuel modules.

2.2 PROGRAM OBJECTIVES

The basic objective of the program is to design and evaluate an alternative SP-100 reactor core based on coated-particle fuel which will alleviate the concerns about the fuel-heat pipe interaction. The specific objectives of the Phase I studies are to provide a backup or alternative fuel concept for the SP-100 concept utilizing coated-particle fuel technology to:

*Dale Dutt, SP-100 Quarterly Review, DOE Headquarters, July 13, 1983.

- o Restrain fuel swelling
- o Prevent fuel migration
- o Contain fission products
- o Prevent fuel-coolant interaction

2.3 CRITICAL ISSUES

The critical issues associated with the SP-100 reference core that are related to the fuel modules (i.e. the fuel tile-and-fin stack and heat pipe) and to be addressed in Phase I of the program are the following:

- o Fuel swelling
- o Fuel migration
- o Excessive fuel temperature during faulted heat pipe operation
- o Fuel/fission products/heat pipes/coolant compatability
- o Propagation of heat pipe failure
- o Thermal resistance at the tile-fin interfaces

The statement of work to be conducted in the Phase-I study follows.

1. Design Criteria

- o Define core design criteria.
- o Develop initial baseline design.
- o Prepare a conceptual design and analysis plan.

2. Coated Particle Design

- o Define coating thickness and fuel porosity.
- o Evaluate coating and matrix material.
- o Determine composite fuel thermal conductivity.

3. Fuel Fabrication

- o Initiate fuel particle fabrication.

Coated-particle design is reported in the next section of this report. This is followed by sections reporting on fabrication of coated particles and fuel compacts and alternative core design using coated particles. The final section summarizes our findings and identifies critical issues related to the coated-particle core.

3. COATED-PARTICLE FUEL DESIGN

The relatively high fuel density and the hard neutron spectrum in space power reactors require modification in design from conventional coated particles used in HTGRs. The low fuel burnup in space reactors facilitates this modification. The TRISO particle design customarily used in land-based reactors is shown in Fig. 1-2. The coating surrounding the spherical fuel kernel consists of an inner layer of low-density pyrolytic carbon and an outer three-layer load-bearing coating consisting of a high-density isotropic carbon layer, a silicon carbide layer, and an outer high-density isotropic carbon layer. Each of these four layers performs a separate function. The low-density carbon layer provides void space for accommodation of gaseous fission products and swelling of the fuel kernel, as well as acting to absorb fission recoils. The outer three-layer coating carries the pressure generated by the retained fission gases. In addition, the inner dense pyrolytic carbon layer prevents corrosion of the fuel during silicon carbide deposition; the silicon carbide layer eliminates diffusive escape of volatile metallic fission products; and, the outer dense pyrolytic carbon provides a microscopically rough surface which allows bonding of the particles into a compact with a carbonaceous matrix.

In HTGRs where fuel density requirements are less severe, fuel compacts are made by bonding together coated particles of a single size in a close-packed arrangement. In this type of compact, the highest particle packing fraction that can be attained is about 0.64.

The coated particles described above are designed for high uranium burnups (20% to 75%). Since the uranium burnup in the space power reactor will be much lower (~3.5%), the particle design can be altered to increase the fuel loading in the particle and, in addition, particles of several sizes can be used to increase the particle packing fraction in the compact.

The particle design proposed for the space power reactor, which is shown schematically in Fig. 3-1, is simpler than the conventional TRISO design. Porosity to accommodate fission gases and swelling will be incorporated directly

SCHEMATIC DRAWING OF THE PROPOSED SPACE REACTOR COATED PARTICLE DESIGN

3-2

Fig. 3-1

into the fuel kernel, and a one-layer or two-layer load-bearing coating will be used to restrain swelling and retain the fission products.

Materials that are possible choices for the fuel kernel are UO_2 , UC, and a UO_2 -UC mixture. Because of its low metal density relative to UC and its poor compatibility with refractory metals, UC_2 is not considered to be a viable choice for the fuel kernel material. Two candidates for coating materials are tungsten and zirconium carbide. If zirconium carbide is employed an underlying carbon layer will be required to seal the fuel from corrosive gases produced during zirconium carbide deposition. Silicon carbide is not considered to be a viable coating candidate because it is subject to chemical attack by fission product and fuel materials at high fuel temperatures.

By using particles of two sizes, differing in diameter by a factor of 7 to 10, the particle packing fraction can be increased from the 0.64 value possible with single-sized particles to a value near 0.85. This is illustrated in Fig. 3-2. The mixture will consist of about 75% of the larger particles and 25% of the smaller particles. The fuel body might be either a loose or a bonded collection of these particles. Loose particles, however, have two major disadvantages. One is that they have a relatively low thermal conductivity, and the other is that they can mechanically interact with the core structure (ratchet) on thermal cycling. In the presence of gravity the bed of particles acts as a solid when forces are applied, so ratcheting may impose stresses on the core container and internal components such as heat pipes. It is not believed that this will happen in space because of the lack of gravity, but it might make ground testing difficult. In any case the desire for increased thermal conductivity probably dictates bonding of the particles into compacts. To increase thermal coupling with the heat pipes, it may be desirable to bond the fuel compacts to the heat pipes. One easy convenient way doing this would be to shrink fit the fuel compacts onto the heat pipes. Another way of thermally coupling the fuel compacts to the heat pipes would be to form the fuel compact directly on the heat pipes.

SP-100 ALTERNATE FUEL PROGRAM: FUEL LOADING CAN BE INCREASED BY USING PARTICLES OF TWO SIZES

Fig. 3-2

In this study the mechanical behavior of particles with both zirconium carbide and tungsten particle coatings will be analyzed. A kernel diameter of 1000 μm for the large particle will be used, because this is about the maximum size that can be made and coated. The dimensions of the small particle will be selected to maximize fuel loading.

3.1 DESIGN OF ZIRCONIUM CARBIDE COATED PARTICLE

Zirconium carbide coatings have been used on fuel kernels in several experimental HTGR fuel particle and target particle designs. In these particles, as in the proposed space reactor particle, there was first an initial pyrolytic carbon coating to seal the kernel from the corrosive gases produced during ZrC deposition. However, the experimental particles had outer coatings beyond the ZrC because the ZrC layer was used for purposes other than containing the fission products. (In some cases it was used to reduce oxygen activity in the particle; in other cases it was included to study its corrosion by fission products.) However, because the coating following the ZrC layer was always a low-density low-strength carbon, the outer coatings did not mechanically reinforce the inner pyrolytic carbon and ZrC layers. In all of these particle batches some fracture of the ZrC layers was observed during irradiation, but in some cases the fraction of particles with broken ZrC layer was small. Thus, the behavior of these particles can be used as guide for the design of ZrC coated particles for the space reactor.

3.1.1 Results of Irradiations of Experimental Coated Particles

The irradiation conditions, particle designs and observed fractional failure of the ZrC layer in the experimental coated particles are tabulated in Table 3-1.

The GF-1 irradiation experiments were intended as corrosion tests of ZrC. They were carried out in France in 1975. The detailed results of these experiments are described in Ref. 3-1. There was 100% failure of the ZrC layer in the UC_2 particles, but the burnup was very high (70%), and there was very

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RESULTS OF IRRADIATION OF EXPERIMENTAL COATED PARTICLES WITH PYROLYTIC CARBON AND ZIRCONIUM CARBIDE COATINGS DIRECTLY ON THE KERNEL

IRRAD. CAPSULE	TEMP (°C)	KERNEL				PYROCARBON SEAL THICKNESS (μm)	ZrC	
		TYPE	DIAMETER (μm)	FRACTION POROSITY	BURNUP		THICKNESS (μm)	FRACTION FAILED
GF1	1200	UC ₂	200	0.03	0.70	19	46	1.00
	1200	(8TH, 1U) O ₂	400	0.05	0.08	14	52	N.D.*
HRB-15A	1250	UO ₂	350	~0.05	0.25	10	14	0.22
HRB-15B	900	UO ₂	310	~0.05	0.21	9	9	~0.10
	900	UO ₂	310	~0.05	0.21	8	14	~0.10
RPR 18	530	LiAlO ₂	340	0.14	0.45	27	7	0.40
RPR 27	530	LiAlO ₂	450	0.18	0.30	31	16	0.11
RPR 33	530	LiAlO ₂	390	0.26	0.18	18	13	0.04
RPR 34	530	LiAlO ₂	400	0.26	0.18	32	16	0.11

* NOT DETERMINED.

Table 3-1

little porosity within the kernel to accommodate the fission-product gases. The (8Th,1U)O₂ particles (which are shown before and after irradiation in Fig. 3-2 and Fig. 3-3, respectively) had some failure of the ZrC layer, but the fraction of failed coatings was not quantitatively determined. When they did occur, the cracks in the coatings on these particles were very fine.

The HRB-15A and HRB-15B irradiation experiments were used to investigate the change in particle behavior caused by a reduction of CO pressure in UO₂ particles due to the presence of ZrC. They were carried out in HFIR in 1980 and are described in Ref. 3-2. Cross sections of one of these particles before and after irradiation are shown in Figs. 3-4 and 3-5. As shown in Fig. 3-5 when the ZrC layer broke there was considerable swelling of the UO₂ kernel, but as long as the coating remained intact, there was very little swelling of the kernel. This is taken as evidence that the zirconium carbide restrained expansion of the fission-product gases present in the kernel.

The Replacement Production Reactor (RPR) irradiation experiments were tests of coated LiAlO₂ particles which can be used for production of tritium in an HTGR. They were carried out in the N-reactor in 1981 and 1982. The ZrC layer in these particles is used to reduce the CO pressure in the particle and as a process seal for the kernel. Cross-sections of representative particles before and after irradiation in one of these experiments are shown in Figs. 3-6 and 3-7. The lithium-neutron reaction results in more gaseous atoms than uranium fission, and therefore, for equivalent burnups, LiAlO₂ particles contain more gas than fuel particles. However, in these particular particles this factor was offset by the larger porosity within the kernels, and thus there were similar gas pressures inside the ZrC coatings.

The results of these irradiation experiments will be used to determine a design stress where ZrC coatings will not experience failure.

3.1.2 Method of Calculating Stresses Generated in ZrC Particle Coatings

The stresses in the duplex pyrocarbon and ZrC coating will be calculated

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Fig. 3-3. Unirradiated $(8\text{Th}, 1\text{U})\text{O}_2$ coated particle with carbon and ZrC coatings on the kernel

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**(8TH, IU) O₂ COATED PARTICLE
WITH CARBON AND ZrC
COATINGS ON THE KERNEL
AFTER IRRADIATION AT
1200°C TO 8% BURNUP**

Fig. 3-4

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**UO₂ COATED PARTICLE WITH CARBON AND ZrC ON
THE KERNEL AFTER IRRADIATION AT 1250° TO 25% BURNUP
KERNEL SWELLING WAS MARKEDLY DIFFERENT ACCORDING
TO WHETHER THE CARBON-ZrC WAS INTACT (A) OR FAILED (B).**

Fig. 3-5

Fig. 3-6

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3-11

Fig. 3-7. LiAlO_2 coated particle with carbon and ZrC coatings on the kernel after irradiation at 530°C to 30% burnup. For these particles, there was no apparent change in particle appearance during irradiation.

pyrocarbon layer. Because the coatings are restrained from shrinkage by the fuel kernel, the pyrocarbon dimensional changes should only act to increase the stresses in the pyrocarbon layer, and the ZrC layer should remain unaffected.

3.1.2.1 Gas Pressure in the Particle

The gas pressure exerted on the coating will be calculated assuming that all of the gas atoms are released to the kernel porosity, that the porosity is interconnected, and that the resulting gas pressure is exerted on the coating. This calculational method overestimates the gas pressure because gas retained within the fuel as small bubbles is partially restrained by the surface tension forces of the bubble surface.

With these assumptions, the gas pressure, P , is given by

$$P = \frac{nRT}{V}$$

where n = No. of moles of gas = No. of atoms created per atom destroyed x moles of atoms destroyed

$$= n_1 \cdot n_2$$

For uranium burnup No. gas atoms created per uranium fission = $n_1 = 0.26$

For lithium burnup No. gas atoms created per lithium transmutation = $n_1 = 1.5$

The moles of atoms destroyed = $n_2 = \frac{4}{3}\pi r_k^3 \rho_k (BU) / \text{molec wt}$

where r_k = kernel radius

ρ_k = density of kernel

BU = burnup

molec wt = molecular weight

The volume available to the gases, v , is given by

$$v = \frac{4}{3}\pi r_k^3 (FP)$$

where FP = fraction porosity in kernel

The gas constant, R , is

$$R = 82 \frac{\text{atm cm}^3}{\text{mole K}}$$

Then, since $\rho_K = \rho_{TH}(1 - FP)$, where ρ_{TH} = the theoretical density of the kernel,

$$P = 82 (nl) \frac{\frac{4}{3} \pi r_K^3 \rho_{TH} (1-FP) (BU) T}{\frac{4}{3} \pi r_K^3 (FP) (\text{molec wt})} \quad (\text{atm})$$

Then,

$$P = 82 \frac{nl \rho_{TH}}{\text{molec wt}} \frac{1 - FP}{FP} (BU) T \quad (\text{atm})$$

$$\text{For } (8Th, 1U)O_2, \quad \frac{nl \rho_{TH}}{\text{molec wt}} = 0.0100$$

$$\text{For } UC_2, \quad \frac{nl \rho_{TH}}{\text{molec wt}} = 0.0113$$

$$\text{For } UO_2, \quad \frac{nl \rho_{TH}}{\text{molec wt}} = 0.0106$$

$$\text{And for, } LiAlO_2, \quad \frac{nl \rho_{TH}}{\text{molec wt}} = 0.0580$$

3.1.2.2 Stress in the ZrC Layer

The stresses produced by pressure in a two-layer hollow sphere can be determined by solving for the interface pressure, P_I , when the interfacial tangential strain is equal for each layer. Let

r_1 = inner radius of coating

r_2 = radius of interface between the layers

r_3 = outer radius of coating

For the pyrocarbon layer the tangential stress, σ_T , at the interface is given by

$$\begin{aligned}\sigma_T &= P \frac{r_1^3(2r_2^3 + r_2^3)}{2r_2^3(r_2^3 - r_1^3)} - P_I \frac{r_2^3(2r_2^3 + r_1^3)}{2r_2^3(r_2^3 - r_1^3)} \\ &= P \frac{3}{2} \frac{r_1^3}{(r_2^3 - r_1^3)} - P_I \frac{1}{2} \frac{(2r_2^3 + r_1^3)}{(r_2^3 - r_1^3)}\end{aligned}$$

The radial stress, σ_r , is

$$\sigma_r = -P_I$$

Then the tangential strain, ϵ_T , in the pyrocarbon layer at the interface is

$$\epsilon_T = \frac{(1 - \mu_1)}{E_1} \sigma_T - \frac{\mu_1}{E_1} \sigma_r$$

where μ_1 and E_1 are the Poisson's ratio and the Young's modulus of pyrolytic carbon. Then

$$\epsilon_T = \frac{(1 - \mu_1)}{E_1} \frac{3}{2} \frac{r_1^3}{(r_2^3 - r_1^3)} P - \frac{1}{2} \frac{(2r_2^3 + r_1^3)}{(r_2^3 - r_1^3)} P_I + \frac{\mu_1}{E_1} P_I$$

For the ZrC layer the tangential stress at the interface is given by

$$\begin{aligned}\sigma_T &= P_I \frac{r_2^3(2r_2^3 + r_3^3)}{2r_2^3(r_3^3 - r_2^3)} \\ &= P_I \frac{1}{2} \frac{(2r_2^3 + r_3^3)}{(r_3^3 - r_2^3)}\end{aligned}$$

The radial stress at the interface is

$$\sigma_r = -P_I$$

Then, the tangential strain at the interface is

$$\epsilon_T = \frac{(1 - \mu_2)}{E_2} \sigma_T - \frac{\mu_2}{E_2} \sigma_r$$

where μ_2 and E_2 are the Poisson's ratio and the Young's modulus of ZrC. Then,

$$\epsilon_T = \frac{(1 - \mu_2)}{E_2} \left[\frac{1}{2} \frac{(2r_2^3 + r_3^3)}{(r_3^3 - r_2^3)} P_I \right] + \frac{\mu_2}{E_2} P_I$$

Because the layers do not separate at the interface, their tangential strains must be equal. solving for P_I from this equality,

$$P_I = P \left[\frac{\frac{(1 - \mu_1)}{E_1} \frac{3}{2} \frac{r_1^3}{(r_2^3 - r_1^3)}}{\frac{(1 - \mu_1)}{E_1} \frac{1}{2} \frac{(2r_2^3 + r_1^3)}{(r_2^3 - r_1^3)} + \frac{1 - \mu_2}{E_2} \frac{1}{2} \frac{(2r_2^3 + r_3^3)}{(r_3^3 - r_2^3)} + \frac{\mu_2}{E_2} - \frac{\mu_1}{E_1}} \right]$$

Then the maximum stress in the pyrocarbon layer is

$$\sigma_T = P \frac{(2r_1^3 + r_2^3)}{2(r_2^3 - r_1^3)} - P_I \frac{3r_2^3}{2(r_2^3 - r_1^3)}$$

and the maximum stress in the ZrC layer is

$$\sigma_T = P_I \frac{(2r_2^3 + r_3^3)}{2(r_3^3 - r_2^3)}$$

In case the pyrolytic carbon layer fails, the maximum stress in the ZrC layer is given by

$$\sigma_T = P \frac{(2r_2^3 + r_3^3)}{2(r_3^3 - r_2^3)}$$

Because pyrolytic carbon has a Young's modulus more than an order of magnitude less than that of ZrC, the carbon layer deforms easily and transmits almost all of the internal pressure to the ZrC. In effect this means that there is little difference in the ZrC stress between the case where the pyrolytic carbon is intact and the case where it is broken.

3.1.2.3 Calculated Stresses in the Experimental Coated Particles. The Young's modulus of isotropic pyrolytic carbon is about 4×10^6 psi (Fig. 3-3) and the Poisson's ratio is about 0.2 (Fig. 3-4). The Young's modulus of ZrC is about 60×10^6 psi (Fig. 3-5) and its Poisson's ratio was assumed to be 0.2. Using these values the maximum stresses in the ZrC layers were calculated for the experimental fuel particles described in Table 3-1. The calculated stresses are shown as a function of the observed fraction of failed ZrC layers in Fig. 3-8. Very high stresses were calculated. This probably occurs because the calculated pressure is larger than the actual pressure because all of the gases are not released from the kernel. However, it seems acceptable to use a stress

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CALCULATIONAL STRESSES IN THE ZrC COATINGS OF IRRADIATED PARTICLES

Fig. 3-8

extrapolated to zero failure in Fig. 3-8 as a preliminary design basis. The value chosen is 100 ksi.

3.1.2.4 Design of Particles for a Space Power Reactor. Coated particles were designed for the nominal conditions expected in a space power reactor using the 100 ksi ZrC stress extrapolated in Fig. 3-8 for zero coating failure. Three kernel materials were considered in this design study. They were UO_2 , UC and a two phase mixture of 50 v/o UO_2 and 50 v/o UC. Particles have been made from UO_2 and from a UO_2 -UC mixture in the HTGR fuel development program, and these particles have performed well in irradiation. Particles have not yet been made from UC, but the sol gel kernel fabrication technology should be adaptable to make this type of fuel.

The design technique will be described in detail for particles with a UO_2 kernel. The technique is identical for the other particle types although the designs differ because of the differing uranium atom density in the fuel material.

The large particle was designed first. The large particle diameter was chosen as 1000 μm because this places the large particle and the small particle, which must be about 10 times smaller, within practical production limits. The pyrocarbon coating thickness was chosen as 5 μm because this is about the minimum thickness that can be used to seal the kernel from the corrosive gases present during ZrC deposition. With these conditions and for a temperature of 1450°C and a burnup of 3.5%, the maximum stress in the ZrC is shown as a function of the total coating thickness for three different kernel porosities in Fig. 3-9. For a maximum ZrC stress of 100 ksi at the end of life, 15.5 μm of coating is required for a porosity fraction of 0.15; 22 μm of coating is required for a porosity fraction of 0.10; and, 30 μm of coating is required for a porosity fraction of 0.07.

It is important to optimize the particle design to maximize the fuel loading in the particle. As is shown in Fig. 3-10, for particles designed for a 100 ksi ZrC coating stress, higher fuel loading is attained with a kernel

END OF LIFE STRESS IN THE ZrC LAYER ON THE LARGE PARTICLE AS A FUNCTION OF COATING THICKNESS AND KERNEL POROSITY

Fig. 3-9

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END OF LIFE STRESS IN THE ZrC LAYER AS A FUNCTION OF THE VOLUME FRACTION OF FUEL

Fig. 3-10

porosity of 0.10 than with kernel porosities of 0.15 and 0.07. Therefore, the large particle design for a 1000 μm diameter UO_2 kernel was selected to be a kernel porosity fraction of 0.10, a 5 μm pyrolytic carbon coating and a 17 μm ZrC coating.

If the small particle is scaled down by a factor of 10 from the large particle dimensions, the kernel diameter becomes 100 μm , the thickness of the pyrolytic carbon becomes 0.5 μm and the thickness of the ZrC becomes 1.7 μm . These are not practical coating thicknesses. Minimum practical coating thicknesses are about 5 μm . Thus, the fuel loading of the small particle is reduced because of the need to use 5 μm coating thicknesses for the pyrolytic carbon and for the ZrC. The fuel volume fraction in the particle can be increased using a smaller porosity in the kernel to maintain the 100 ksi design stress, but a higher overall fuel loading can be achieved by increasing the diameter of the small particle and accepting some decrease in particle packing fraction. The fractional volume of fuel in a coated particle compact is shown in Fig. 3-10 as a function of the diameter of the small fuel kernel. To calculate this curve the data of Ref. 3-6 on the random packing of spheres of different sizes has been used along with the selected 1000 μm diameter particle and a small particle with a 10 μm total coating thickness and a fractional kernel porosity selected to maintain a 100 ksi ZrC stress. For maximum fuel loading in the compact, the small particle should have a diameter of 150 μm , a total coating thickness of 10 μm , and a kernel porosity fraction of 0.05. Thus, the selected designs for the two UO_2 particles, which are summarized in Table 3-2, produce a fuel volume fraction of 0.60 in the compact. This would result in a uranium atom density of 0.0249 moles U per cc as compared with a uranium atom density of 0.0306 in the reference SP-100 core considering just the fuel and the fins. (A coated particle fuel compact would replace the fuel and the fins in the reference SP-100 core.)

If either a UO_2 -UC mixture or UC is used as the kernel material a higher uranium atom density results because the uranium density in these materials is higher than that in UO_2 . However, because of this higher uranium density the particle designs must be changed to accommodate the increased amount of

FUEL VOLUME IN THE TWO-PARTICLE COMPACT AS A FUNCTION OF THE SIZE OF THE SMALL PARTICLE

Fig. 3-11

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ZIRCONIUM CARBIDE COATED PARTICLE DESIGNS FOR A SPACE POWER REACTOR

1. UO_2 KERNEL

PARTICLE TYPE	KERNEL		COATING		COMPACT
	DIAMETER (μm)	FRACTION POROSITY	PYROCARBON THICKNESS (μm)	ZIRCONIUM CARBIDE THICKNESS (μm)	URANIUM* ATOM DENSITY (MOLE U/CC)
LARGE	1000	0.10	5	17	0.0249
SMALL	150	0.05	5	5	

2. 50 V/O UO_2 – 50 V/O UC KERNEL

PARTICLE TYPE	KERNEL		COATING		COMPACT
	DIAMETER (μm)	FRACTION POROSITY	PYROCARBON THICKNESS (μm)	ZIRCONIUM CARBIDE THICKNESS (μm)	URANIUM* ATOM DENSITY (MOLE U/CC)
LARGE	1000	0.10	5	20	0.0286
SMALL	150	0.06	5	5	

3. UC KERNEL

PARTICLE TYPE	KERNEL		COATING		COMPACT
	DIAMETER (μm)	FRACTION POROSITY	PYROCARBON THICKNESS (μm)	ZIRCONIUM CARBIDE THICKNESS (μm)	URANIUM* ATOM DENSITY (MOLE U/CC)
LARGE	1000	0.10	5	23	0.0322
SMALL	150	0.07	5	5	

* THE URANIUM ATOM DENSITY IN THE REFERENCE SP-100 CORE CONSIDERING JUST THE FUEL, FUEL POROSITY, INTRA-MODULE GAPS AND THE FINS IS 0.0306 MOLE U/CC.

Table 2

fission-product gases contained within the particle. The particle designs for these two fuel materials are described in Table 3-2. The procedure used to obtain these designs was the same as that described above for particles with UO_2 kernels. As can be seen with a UC kernel the uranium atom density in the fuel compact is slightly greater than that in the reference SP-100 fuel and fins arrangement, while the uranium atom density in a coated particle fuel compact with 50 v/o UO_2 and 50 v/o UC in the fuel kernel is equidistant between that for the UC kernels and that for UO_2 kernels.

3.2 Design of Tungsten Coated Particles

The design of tungsten-coated fuel particles needs to consider the coating from the standpoint of creep strain in the coating and from possible creep rupture of the coating. As opposed to the ZrC coated particles there are no experimental data for tungsten-coated fuel particles on which to base this calculation, but there are data on the high-temperature creep behavior of CVD tungsten that can be applied.

3.2.1 Pressure in the Particle

The expression for the pressure in the particle is somewhat different from that for the ZrC coatings because creep of the tungsten will open up a volume for accommodation of the fission product gases. The pressure in the particle is given by

$$p = \frac{nRT}{V}$$

where as with the ZrC particles

$$n = n_1 \cdot n_2$$

$$n_1 = 0.26 \text{ for uranium fission, and}$$

$$n_2 = \frac{4}{3} \pi r_K^3 \rho_K (\text{BU}) / \text{molec}$$

However in this case

$$V = \frac{4}{3} \pi r_K^3 (FP) + \frac{4}{3} \pi (r_C^3 - r_K^3)$$

where r_C = inner radius of the coating

$$r_C = r_K (1 + \epsilon_T)$$

where σ_T is the tangential strain at the inner radius of the coating.

Thus

$$V = \frac{4}{3} \pi r_K^3 [FP + (1 + \epsilon_T)^3 - 1]$$

and

$$P = 82 \frac{n_1 \rho_{TH}}{\text{molec wt}} \frac{(1 - FP)}{[FP + (1 + \epsilon_T)^3 - 1]} (BU) T \quad (\text{atm})$$

3.2.2 Stress and Strain of the Tungsten Coating

In order to simplify the calculations, the thin wall expression for the stress in a spherical vessel will be used.

$$\sigma_T = \frac{Pr}{2t}$$

where σ_T = the tangential stress

r = the radius of the coating $\approx r_C = r_K (1 + \epsilon_T)$

$\dagger$ = the thickness of the coating

Because creep occurs at constant volume, $\dagger$ will decrease with increasing creep strain according to

$$\dagger = \dagger_I \frac{r_K^2}{r_C^2} = \dagger_I \frac{1}{(1 + \epsilon_T)^2}$$

Thus

$$\sigma_T = \frac{Pr_K}{2\dagger_I} (1 + \epsilon_T)^3$$

The diametral strain of the coating is given by ϵ_T since $\epsilon_T = u/r_C$ where u is the radial displacement. Elastic strains will be neglected. Then because the stress state is biaxial

$$\epsilon_T = \int_0^t (1 - \mu_C) \dot{\epsilon} dt$$

where μ_C is Poisson's ratio in creep, t is time and $\dot{\epsilon}$ is the uniaxial creep rate.

Because creep occurs at constant volume $\mu_C = 0.5$.

3.2.3 Analysis of Creep Data for CVD Tungsten

Creep rate and creep-rupture data for CVD tungsten are reported in Ref. 3-7, the data from which are reproduced in Figs. 3-12 and 3-13.

Fig. 3-12. Comparison of the minimum creep rates of CVD and PM tungsten.

Fig. 3-13. Comparison of the creep-rupture properties of CVD and PM tungsten.

3.2.3.1 Creep Rate

From Fig. 3-12, it seems that

$$\dot{\epsilon} = B\sigma^A$$

is an appropriate creep rate law for CVD tungsten.

This expression can also be written

$$\log \dot{\epsilon} = \log B + A \log \sigma$$

Two points from the straight line drawn through the 1650°C data in Fig. 3-12 are:

σ	$\dot{\epsilon}$
4,000	0.00004
20,000	0.6

Then $A = 6$ and $\log B = -26.03$

Two points from the straight line drawn through the 2200°C data in Fig 3-12 are:

σ	$\dot{\epsilon}$
1,000	0.0003
6,000	0.7

Then $A = 4.3$ and $\log B = -16.4$

In creep it is usual that A is temperature independent and that B follows an expression of the form.

$$B = B_0 e^{-Q/RT}$$

For this analysis, $A = 6$ will be chosen.

Then at 1650°C $\log B = -26.03$. With $A = 6$, $\log B = -21.81$ at 2200°C using the mid point of the 2200°C curve.

Then solving for Q/R and B_0

$$Q/R = 84,035 \text{ and } B_0 = 8.91 \times 10^{-8}$$

Then at the fuel operating temperature of 1730°K

$$B = 7.14 \times 10^{-29}$$

Thus the uniaxial creep law becomes

$$\dot{\epsilon} = 7.14 \times 10^{-29}$$

3.2.3.2 Creep Rupture. The creep rupture data in Fig. 3-13 need to be extrapolated to obtain the rupture stresses at the 1730°K fuel temperature for one-half of fuel lifetime (30,000h) and for the full fuel life time (60,000h).

Two methods have been used. One is a linear extrapolation of the data in Fig. 3-13 in temperature and time. This method yields a full-life stress rupture value of 3.2 ksi and an one-half life value of 3.8 ksi.

The other method uses the Larsen-Miller parameter which is an empirical

parameter that commonly is used to relate stress rupture data at different temperatures. The Larsen-Miller plot of the tungsten data of Fig. 3-13 are shown in Fig. 3-14. The Larsen-Miller parameter for a 60,000 hr life at 1730°K is 7.72×10^4 . Thus, this method predicts a full life rupture stress of 4.6 ksi. The 30,000 hour Larsen-Miller parameter at 1730°K is 7.62×10^4 , and the predicted rupture stress is 5.0 ksi.

3.2.4 Prediction of Particle Behavior

Using the above expressions and assuming a UO₂ fuel kernel, the tangential stress in the particle coating is given by:

$$\sigma_T = 0.435 \frac{(1 - FP) (BU) Tr_K (1 + \epsilon_T)^3}{r_I [FP + (1 + \epsilon_T)^3 - 1]}$$

If one assumes BU increases linearly with time to a value of 0.035 then $BU = (0.035/60,000) \times t = 5.83 \times 10^{-7} t$, where t is the time in hours.

Also since $T = 1730^\circ K$,

$$\sigma_T = 4.40 \times 10^{-4} \frac{(1 - FP) (1 + \epsilon_T)^3}{[FP + (1 + \epsilon_T)^3 - 1]} \frac{r_K}{r_I} t \quad (\text{atm})$$

$$\sigma_T = 6.45 \times 10^{-3} \frac{(1 - FP) (1 + \epsilon_T)^3}{[FP + (1 + \epsilon_T)^3 - 1]} \frac{r_K}{r_I} t \quad (\text{psi})$$

The diametral strain is given by

$$\epsilon_T = (1 - \nu_C) \int_0^t B \sigma^n dt$$

GA Technologies

3-32

Fig. 3-14. Larsen-Miller plot of tungsten stress-rupture values.

Since creep occurs at constant volume, $c = 0.5$ and

$$\epsilon_T = 0.5 \int_0^t 7.14 \times 10^{-29} \left[\frac{6.45 \times 10^{-3} (1 - FP) (1 + \epsilon_T)^3}{[FP + (1 + \epsilon_T)^3 - 1]} \frac{r_K}{r_I} \right]^p dt$$

In order to evaluate particle behavior during irradiation this expression was solved using a stepped iterative numerical technique.

Calculated particle diametral strains in tungsten coated UO₂ particles are shown as a function of the coating thickness and the fraction of porosity in the kernel in Fig. 3-15. Very large swelling values are predicted. If these swelling values actually occurred, this would preclude the use of tungsten coatings. In addition, the end of life coating stresses in the particle designs, which are shown in Fig. 3-16, are all higher than the one-half life creep rupture stress extrapolated by either the linear method or the Larsen-Miller method. Since the stresses are relatively constant in the last one-half of the particle life, this implies that the coatings will break before the end of life. Thus, based on these calculations tungsten coatings on UO₂ particles will not serve to contain fuel swelling.

It should be noted, however, that based on the ZrC coated particle calculations, the pressure inside the particle is probably too high because a ZrC fracture stress of 100 ksi was calculated for the experimental particle while the actual fracture stress is probably about 50 ksi. The high pressure comes from the assumption that the gas is released from the fuel when it actually is retained in bubbles in the fuel. Because the surface tension on the surface of the gas bubbles helps to restrain the gas pressure, the pressure exerted on the coating is less than if the gas is released.

END OF LIFE COATING DIAMETER STRAINS IN TUNGSTEN COATED PARTICLES

Fig. 3-15

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END OF LIFE COATING STRESSES IN TUNGSTEN COATED PARTICLES

Fig. 3-16

If one-half of the completely released gas pressure is used in calculation of tungsten coating stresses and strains, the particle swelling is greatly reduced to acceptable values in some particle designs ($\sim 0.3\%$). However, the end-of-life coating stresses are still calculated to be larger than the two extrapolated one-half life stress rupture values for all of the coating designs considered above. Thus, it is still predicted that the coatings will fail.

- 3-1 "Irradiation Behavior of Fuel Particles Containing Chemically Vapor Deposited Zirconium Carbide Coatings," G. H. Reynolds, J. C. Jouvier, J. L. Kaae, and J. P. Movlevat, J. Nucl. Mater., 62, 9 (1976).
- 3-2 "Performance of Coated Oxide Particles Gettered with ZrC," R. E. Bullock and J. L. Kaae, GA-Al6624, June 1982 (to be published in J. Nucl. Mater.)
- 3-3 "Relations Between the Structure and the Mechanical Properties of Fluidized Bed Pyrolytic Carbons," J. L. Kaae, Carbon, 9, 291 (1971).
- 3-4 "Poisson's Rates of Pyrolytic Carbon," R. J. Price and J. L. Kaae, Carbon, 7, 706 (1969).
- 3-5 "High Temperature Materials," Peter T. B. Shaffer, Plenum Press, New York (1964).
- 3-6 "Mechanical Packing of Spherical Particles," R. K. McGeary, J. Amer. Cer. Soc., 44, 513 (1962).
- 3-7 "Mechanical Behavior of CVD Tungsten at Elevated Temperatures," H. E. McCoy and J. O. Stiegler, Proceedings of the International Conference on Chemical Vapor Deposition of Refractory Metals, Alloys and Compounds, Gatlinburg, TN, September 1967.

4. COATED PARTICLE FUEL FOR SPACE POWER SYSTEMS

4.1 MATERIALS COMPATIBILITY

There are three main areas of concern about the chemical compatibility of materials used in a space power reactor (SP-100) having fuel elements consisting of coated fuel particles embedded in a matrix material. The most important of these concerns regards compatibility of the bonding matrix and of the coating material on fuel particles with the Mo-Re heat pipes that cool the core. The second concern regards compatibility of the particle coating with the encapsulated nuclear fuel material. Finally, in the case of coating failure, the compatibility of the nuclear fuel material with the Mo-Re heat pipes and with the matrix material surrounding the particles becomes a concern.

The use of the coated particle de-emphasizes heat pipe-fuel compatibility and gives added flexibility in selecting the type of nuclear fuel to be used. In uncoated fuel, for example, UO_2 would be strongly preferred over UC or UC_2 fuels because of reactions between the uranium carbides and the refractory metals in the heat pipes (Refs. 4-1 to 4-6), but uranium carbides would likely be acceptable fuels in a coated particle system. Likewise, uncoated UN fuel would not be attractive because of its higher rate of vaporization at high temperatures (Ref. 4-7), which could be prevented with the closed system provided by coated fuel particles. Thus, the primary compatibility issue in coated particle fuel shifts from fuel-heat pipe reactions to coating/matrix-heat pipe reactions provided that high coating reliability is obtained, and this allows wider ranges of nuclear fuels to be considered for space-power applications.

The standard coated fuel particle developed for the high-temperature gas-cooled reactor (HTGR) has an outer coating of vapor-deposited pyrocarbon and is bonded into fuel compacts with a carbonaceous matrix (Ref. 4-8). Carbon would react readily with Mo in the heat pipes to form Mo_2C at temperatures as low as 1200°C , with the growth in thickness, X , of the Mo_2C layer with time at 1200°C being defined by the parabolic relationship (Ref. 4-5)

$$X(m) \approx 8 \times 10^{-8} [t(s)]^{1/2},$$

which predicts about 0.3 mm of carbide formation for 3600 h, for example. Moreover, this rate of formation goes up very sharply with temperature, such that $X \approx 3.5$ mm for the same time at 1500°C (Ref. 4-5). Therefore, carbon coatings and/or matrix materials cannot be considered for use in a space power reactor having Mo-Re heat pipes, but they could be used in an advanced design using graphite heat pipes.

For a system having heat pipes made of refractory metals, the potential coating materials for fuel particles include the refractory metals (W, Mo, Re, Ta, and Nb) and high-temperature metal carbides that have a higher affinity for carbon atoms than the heat pipe materials or any other refractory metals that might be used as a matrix material in bonding the coated particles together. An excellent carbide candidate of the latter type is ZrC, which has demonstrated outstanding resistance to chemical attack from uranium and from uranium fission products in irradiated fuel particles (Ref. 4-9). The ZrC coating has also been shown to provide good retention of gaseous (Ref. 4-10) and metallic (Ref. 4-11) fission products.

The carbide-forming reaction $Zr + C \rightarrow ZrC$ has a lower free energy of formation than does the reaction of carbon with Mo or W for temperatures up to 2000°C (Fig. 4-1); therefore, ZrC should not give up its carbon to these refractory metals or to Re, which does not form a stable carbide (Ref. 4-1), and should be compatible with the MoRe heat pipes or with Mo, W, or Re matrix materials. However, Ta and Nb form carbides (Ta_2C and Nb_2C) that have lower free energy states than ZrC (Ref. 4-2), and these metals should be expected to remove and react with the carbon in ZrC. Thus, Ta or Nb would not be suitable for use as matrix materials for ZrC-coated fuel particles. All candidate coating materials must also be compatible with the uranium fuel kernel, of course, and Ta or Nb are not compatible with UC or with UC_2 over the temperature range from 1200 to 1600°C (Ref. 4-5). Consequently, Ta and Nb can also be eliminated as possible coating materials for carbide fuel systems.

GA Technologies

4-3

Fig. 4-1. Comparison of free energy of formation for selected carbides (from Ref. 2).

Of the possible refractory metal coating materials for fuel particles, tungsten has been most thoroughly investigated and seems to have the best potential. After extensive experimental compatibility studies conducted during research on thermionic emitters (Refs. 4-1 and 4-3), tungsten was selected as the best thermionic cathode material with which to cover nuclear candidate fuels consisting of UC, UC + UC₂, and mixtures of uranium carbides and ZrC. Tungsten was found to be compatible with UC up to temperatures of 1800°C in these studies, whereas the metals M = Ta, Nb, and Mo were found to react with UC as follows:

The free uranium metal produced here will be in a molten phase at 1800°C (Ref. 4-1), and in some cases this holds down to temperatures as low as 1200°C (Ref. 4-3), and reaction rates between liquid uranium and the refractory metals are greatly increased over those between the metals and UC (Ref. 4-5), with the latter rates being determined largely by the time required for carbon from UC to diffuse across the carbide layer and reach the metal surface.

These very detrimental liquid-forming reactions in refractory metals other than W and Re at high temperatures can be suppressed by adding UC₂ to UC, with the reaction obtained then being

Solid carbide layers are still formed on the refractory metals, of course, but these can be reduced by the addition of ZrC to stabilize the uranium carbide and retard the migration of carbon out of the kernel, thereby reducing carbide-forming reactions in the metals (Ref. 4-1). The safest procedure for any fuel material, however, is to limit the metallic coating materials for fuel particles to W and Re or their combinations. Of these two candidates, tungsten has been much more widely studied and should be given priority.

Thus, because of compatibility requirements of particle coatings with both

heat pipes in the core and with the nuclear fuel, the two coating materials selected for detailed evaluation in a subsequent section on coated particle design are ZrC and W. Likewise, because of compatibility requirements of matrix materials with both particle coatings and with heat pipes, it is recalled that the possible refractory metals that might be suitable for use in bonding W-coated and ZrC-coated particles together had previously been reduced to W, Re, and Mo. As a result of subsequent considerations, ZrC can also be added to this list of candidate matrix materials. All of these candidate materials have rather well matched coefficients of thermal expansion (Table 4-1), and there should be no major thermal compatibility problems. The only remaining area of compatibility concern is that between the nuclear fuel material and the Mo-Re heat pipes, and this is of importance only if there is significant failure of coatings during reactor operation. Nevertheless, some consideration of this might serve to rank the wider variety of nuclear fuels that can be used in a coated particle system.

In ordinary carbon-coated HTGR particles, UC_2 is used as the carbide fuel form because it is the equilibrium uranium carbide phase in contact with carbon at high temperatures (Ref. 4-12). However, the UC monocarbide phase would be preferred in a coated particle without carbon, because of its 10% higher uranium density. Also, in the case of coating failure on carbide fuel particles adjacent to the Mo-Re heat pipes, the refractory metals should not react with stoichiometric UC fuel at temperatures below 1900°C according to thermodynamic predictions, because UC has a lower free energy of formation than MoC or Mo_2C over this temperature range (Fig. 4-1) and Re does not form a stable carbide (Ref. 4-1).

On the other hand, if excess carbon is present to form a UC_2 phase, which has a higher free energy of formation than molybdenum carbides above 1250°C (Fig. 4-1), then carbide formation on the heat pipes should occur until all the UC_2 phase is converted to UC. This predicted reaction behavior has been verified experimentally for temperatures between 1200 and 1600°C (Ref. 4-5). However, at 1800°C, UC apparently reacts with Mo in the absence of UC_2 (Ref. 4-1), but this temperature is well beyond that for normal space power operation.

Table 4-1. Coefficients of Thermal Expansion for Candidate Materials

<u>Materials Coating/Matrix</u>	<u>CTE (10⁶/°C)</u>
W	4.6-6.4
Mo	4.9-6.5
ZrC	6.3-8.5
Re	6.7-7.3
Ta	5.5-8.0
Nb	7.1-9.0
<u>Fuels</u>	
UO ₂	9.2-12.6
UC	9.3-13.1
UC ₂	6.3-14.6
UN	8.0- 9.7

Therefore, stoichiometric UC should be an acceptable fuel for a coated particle system, but UC₂ would not be preferred because of heat pipe compatibility concerns in the case of coating failure. Likewise, UN fuel should also be acceptable for coated particles based on compatibility with refractory metals (Ref. 4-6) and on suppression of vaporization (Ref. 4-7); however, it should be expected to react with zirconium in a ZrC coating, since ZrN has a lower free energy of formation than ZrC (Ref. 4-6), and might not be preferred because of this limitation on candidate coating materials.

The fuel material with the fewest chemical compatibility problems for a space power reactor is stoichiometric UO₂, which could probably be used even in an uncoated form. However, even here, one might get into problems if the stoichiometry is not carefully controlled. Substoichiometric UO₂ can contain low-melting uranium metal (Ref. 4-13), which would be unacceptable. At the same time, hyperstoichiometric UO₂ has a sharply increased oxygen potential, which can cause compatibility problems with materials that are subject to oxidation reactions. At sufficiently high temperatures, for example, it is known that UO_{2+x} will react with W and Re (Ref. 4-13). Nevertheless, strictly from a chemical compatibility standpoint, stoichiometric UO₂ would be the preferred fuel for the space power reactor. However, many other factors enter into the consideration of a fuel choice, such as: fuel swelling under irradiation, thermal conductivity and stability, uranium density, fuel vapor pressure as a function of temperature, and development status of the fuel. Some of these additional fuel considerations will be addressed in the next section on fuel properties.

In addition to chemical compatibility, when two different refractory metals are placed in close contact there is potential for interdiffusion of the metals at sufficiently high temperatures, which can produce Kirkendall voids near the interface of the lower-melting component (Ref. 4-14). The formation of such voids is believed to be caused by the coalescence of lattice vacancies left behind when atoms of the faster-diffusing, lower-melting components cross the interface at higher rates than those of the higher-melting component diffusing in the opposite direction. The diffusion rates are large for tempera

tures in the range of 2000 to 2200°C where they have been measured (Ref. 4-14); they fall off rapidly with decreasing temperature over the range of measurement, but are still not insignificant when projected down to normal space power temperatures. The width, w , of the interdiffusion zone for W and Mo in close contact at 1500°C for t hours, for example, is extrapolated to be

$$w(\mu\text{m}) = 2 t^{1/2} ,$$

and for 10,000 h this gives $w = 200 \text{ } \mu\text{m}$. If different refractory metals are used in SP-100, projected diffusion widths of this magnitude are probably sufficient to justify additional work on interdiffusion at normal space reactor temperatures.

4.2 FUEL PROPERTIES

Much of the discussion of this section is taken from a recent review by Gulden and Watson on high-temperature fuels for advanced nuclear systems (Ref. 4-7). The first fuel property to be discussed is that of fuel swelling caused by the precipitation of gaseous fission products into bubbles within the fuel. The pressure of gas in the bubble (P) is balanced by the surface tension (γ) plus the hydrostatic component of externally applied stress (σ) so that an equilibrium size (r) is established according to the classical expression $P = (2\gamma/r) + \sigma$. An addition of fission gas to the bubble or a change in the applied stress will cause the bubble to grow or shrink by the addition or loss of vacancies in order to maintain equilibrium.

Since the gas in the bubbles occupies a much larger volume than the equivalent amount of gas dispersed as individual atoms in the matrix, the precipitation of fission gas into bubbles results in a volume increase, or swelling of the fuel. Bubbles can migrate, grow, and coalesce both by diffusional processes and by interaction with fission tracks. Large bubbles can also grow by plastic deformation processes. It is clear from the bubble equation that as a

bubble grows, its internal pressure diminishes. Consequently, bubble growth results in an increase in swelling. Fortunately for many applications, bubble growth and swelling do not continue indefinitely. Large bubbles tend to form at the grain boundaries where they can link up and release their gas to the surrounding environment. Thus, the phenomena of swelling and fission gas release are inextricably interrelated.

A comparison of swelling rates of UC, UO_2 , and UN fuels is given in Fig. 4-2. All of the fuel types follow the same trends, as would be expected, but the nitride fuel appears to have a clear advantage in dimensional stability. These swelling data should be used with caution, however, since they are valid only over a limited range of volume change and were in some cases obtained from measurements on cladding rather than fuel.

In a practical sense swelling, fission gas release, and columnar grain growth can result in substantial shape changes and redistribution of the fuel as illustrated schematically in Fig. 4-3. In Stage 2 of this illustration, the fuel swells uniformly due to the effect of fission gas bubbles. Under conditions in which columnar grain growth occurs, as in Stage 3, the fission gases will be swept up and released, leaving redensified, but displaced, fuel as shown in Stage 4. The process can then repeat itself. This type of process can lead to substantial creep deformations of the surrounding clad or core structure that are difficult to predict or treat in design. While these processes are generally related to conventional metal clad fuel pins, they can also occur in less conventional structures such as the SP-100 plate fuel if the temperatures and temperature gradients are sufficiently high. Fortunately, in the current SP-100 design, the conditions do not appear to be sufficiently severe to result in significant fuel redistribution by this mechanism. However, the possibility should be considered in advanced designs and under faulted conditions.

Other critical properties for the various candidate fuels include: thermal conductivity, uranium density, melting temperature (all listed in Table 4-2), and fuel vapor pressure as a function of temperature (Fig. 4-4). Based on the

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Fig. 4-2 . Dependence of fuel swelling on temperature for UC, UN, and UO₂
(from Bauer, 1972)

Fig. 4-3. Schematic diagram of the swelling - gas release process due to swelling by lenticular cavities (from Olander, 1976)

Table 4-2. Selected Properties of High-Temperature Nuclear Fuels

<u>Fuel Type</u>	<u>Melting Temperature °C</u>	<u>Thermal Conductivity W/cm-K (to 1000°C)</u>	<u>Uranium Density gm/cc</u>
UO ₂	2865	0.03	9.5
UC	2525	0.2	13
UC ₂	2480	0.35	11.7
UN	~2800 (@2.5 atm.)	0.2	13.5

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Fig. 4-4. Vapor pressure of various fuels (from Buden, et al., 1979)

properties surveyed to this point, it will now be convenient to discuss the strengths and weaknesses of each of the fuel candidates separately, with emphasis on their use in a space power reactor.

4.2.1 Oxide and Cermet Fuels

UO₂ is by far the most widely used, the most studied, and the best understood of the high-temperature nuclear fuels. It is the standard fuel for the Light Water Reactor, Advanced Gas-Cooled Reactor (AGR), and Fast Breeder Reactor (FBR), as well as the Space Power Reactor (SP-100) under development at Los Alamos National Laboratory (LANL). It is generally isolated from the reactor coolant by a stainless steel or zircaloy cladding. However, in the case of SP-100, it is used as unclad wafers separated by thin Mo-Re spacers.

The high melting temperature, compatibility with metallic materials, irradiation stability, and ease of fabrication of UO₂ have made it the front runner among the high-temperature fuels. The primary disadvantages of UO₂ relative to other high-temperature fuels are its low thermal conductivity and relatively low uranium density. The thermal conductivity of UO₂ is a factor of 7 to 10 lower, and the uranium density is as much as 30% lower than the other high-temperature fuels.

Because of the low thermal conductivity of UO₂, temperature and temperature gradients in most fuel designs tend to be high. Consequently, restructuring, swelling, and fission gas release figure prominently in the design of oxide fuels. Swelling data for UO₂ fuel are shown in Fig. 4-5.

Fuel swelling is of particular concern in the design of the SP-100 reactor because of the potential for dimensional changes to disrupt the integrity of the heat pipes that cool the core. Moreover, the design of the SP-100, with unclad fuel plates, is particularly susceptible to swelling.

Some recent French [Commissariat à l'Energie Atomique (CEA)] data on swelling of UO₂ microspheres suggest a way to minimize swelling that could sig-

Fig. 4-5. Fuel swelling curves for SP-100 core design (from Buden and Stocky, 1982)

nificantly simplify the design of a reactor such as SP-100. Figure 4-6 shows the CEA swelling data at 1500°C as a function of initial density. While the high density material follows a fairly normal swelling curve the low initial density material (92% to 95%) swells only a few percent and then saturates or even redensifies somewhat. Although the work done to date is somewhat sketchy, the key to this behavior appears to be the production of a controlled uniform fine porosity. With further development this type of fuel could be very attractive for the SP-100.

Oxide fuel has also been proposed for use in the cermet form, consisting of UO_2 with additions of 20% to 40% of the refractory metals tungsten or molybdenum. The principal advantage of cermet fuel is its greatly improved thermal conductivity relative to the oxide as shown in Fig. 4-7. It may also exhibit improved swelling behavior, but this is not confirmed. The principal disadvantage of cermet fuel is the reduced fuel density which leads to increased core sizes and weights.

4.2.2 Carbide Fuels

At least three types of uranium carbide fuels have been used in nuclear systems: the monocarbide UC, the dicarbide UC_2 , and mixed carbide UCZrC . Mixed thorium-uranium carbide has been used in HTGR's such as the Fort St. Vrain Reactor. Each of these has its own unique characteristics.

Uranium monocarbide has a uranium density nearly 30% higher than UO_2 (Table 4-2) and a thermal conductivity nearly an order of magnitude higher (Fig. 4-7). A major limitation in the use of carbide fuels in high-temperature applications is chemical compatibility with refractory metals that might be used as cladding or core structural materials (Section 4.1).

Uranium dicarbide was the reference fuel for the NERVA and ROVER Projects, and is now the reference fuel for the rotating bed and fixed bed reactors. The Fort St. Vrain HTGR, now operating on the grid of Public Service Company of Colorado, is fueled with $(\text{Th,U})\text{C}_2$. All of these systems have in common the use

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Fig. 4-6. CEA data on swelling of UO_2 with various initial densities at 1500°C (from Kauffman, et al., 1980)

Fig. 4-7. Thermal conductivity vs temperature for various fuels (from Buden, et al., 1979)

of pyrolytic carbon coatings, which could not be used in most present space power designs because of their hard neutron spectrum.

The mixed-carbide fuel 90% UC_2 -10% ZrC was under development in the in-core thermionic program at General Atomic Company (Ref. 4-1). To improve the dimensional stability and the thermionic performance of carbide-fueled emitters during in-pile operation, it was necessary to exercise rigid control of the structures, compositions and stoichiometries of the carbide fuel material. To facilitate fission gas release in order to reduce fuel swelling, the carbide fuel bodies had low densities (75% to 79% theoretical value) and contained a large amount of open porosity. To prevent the dissolution of the tungsten cladding at the operating temperature of the emitter, about 4 wt% of tungsten was added to the carbide fuel material. The C/U atom ratio was set at between 1.03 and 1.05 in order to reduce the diffusion rate of uranium through the tungsten cladding and to minimize the carburization of the tungsten cladding. Such carbide fuel material consisted of a uranium-zirconium carbide matrix containing a few wt% of dissolved tungsten and dispersions of UWC_2 phase. The fuel bodies usually contained a central hole of about 10% of the fuel cavity volume in order to accommodate fuel swelling. Irradiation testing showed that this structure was effective in reducing swelling below that for oxide fuels.

4.2.3 Nitride Fuels

Uranium nitride is the least studied and the least understood of the potential high-temperature fuels. However, nitride fuels have been considered both as a fuel for space power reactors and as an advanced fast breeder reactor fuel. Uranium nitride has the highest uranium density and the highest thermal conductivity of any of the unalloyed high-temperature fuels (Table 4-2). It also appears to have the lowest irradiation induced swelling rate, as was shown previously in Fig. 4-2.

A key concern with uranium nitride fuel is its high vapor pressure at high temperature as illustrated in Fig. 4-4. The vapor pressure reaches 1 atmos-

phere at about 2200K. This is a particular concern for the SP-100 where the fuel is unclad and operates in a vacuum. Practical application of nitride fuels at high temperatures will require a nitrogen overpressure and some form of cladding.

4.2.4 Comparison of the Various Fuel Candidates

While a variety of high-temperature fuels is available for advanced nuclear systems, the choice of a fuel type is controlled by a complex interaction between systems design constraints and fuel properties and behavior. Key considerations include fuel compatibility with cladding and coolant, fuel swelling and fission gas release, and density of fissile material.

Uranium dioxide has become the standard fuel for most reactors operating today that use conventional metal clad fuel. It has excellent compatibility with cladding materials, good irradiation behavior, and perhaps most important, it has an impressive and successful record of use. Monocarbide and mononitride fuels remain of interest for advanced applications because of the substantial improvements they offer in both fissile density and thermal conductivity. However, materials compatibility issues must be carefully considered with the carbides; and vaporization is a concern with the nitrides. In both cases, but especially with nitrides, the advantages for a particular system must be substantial to outweigh the relative lack of experience and data as compared to the oxide. Cermets, especially UO_2 with refractory metals, and the mixed carbide UC-ZrC are also potential high-temperature fuel choices.

The range of available high-temperature fuels for advanced nuclear systems is broad. In general, no single fuel will be optimum for all systems requirements. The fuel selection process involves a tradeoff between systems design and materials properties. In some cases, the fuel properties can be modified to better match systems requirements as in cermets, alloyed fuels, or coated particles.

4.3 Fuel Kernel Fabrication

The fuel kernels of the coated particles used for fueling the SP-100 core have to possess higher uranium concentration than UO_2 in order to compensate for the space lost to the particle coating and the interstices among the particles. In addition, with the elimination of the heat pipe fins, the fuel kernels need higher thermal conductivity than that of UO_2 in order to avoid excessively high fuel temperature. Uranium monocarbide and uranium oxycarbide, which have both higher uranium concentration and higher thermal conductivity than that of UO_2 , are attractive fuel kernel materials for SP-100 application. To achieve high fuel loading in the core, a two-particle system is recommended, in which the larger particle has a kernel diameter of about 1000 microns, while the smaller particle which fits into the interstices of the large particles has a kernel diameter of about 150 microns. One of the primary requirements for developing the coated particle concept for the SP-100 application is the fabrication of uranium monocarbide and uranium oxycarbide kernels of such sizes.

Figure 4-8 shows the U-C-O ternary diagram between 1300 and 1750°C (Refs. 4-15 and 4-16). The compositions of interest to the present application from the point of view of uranium concentration and high temperature compatibility with molybdenum and tungsten lie in the single phase UC_xO_y region (including UC when $x=1$, $y=0$) and the two-phase $\text{UC}_x\text{O}_y + \text{UO}_2$ region. The single phase UC_xO_y has higher uranium concentration than the two-phase $\text{UC}_x\text{O}_y + \text{UO}_2$.

Under the HTGR fuel development program, GA Technologies Inc. has fabricated uranium oxycarbide kernels of 200 to 350 micron diameters by the gel-supported precipitation method (GSP) for compositions in the $\text{UC}_x\text{O}_y + \text{UO}_2$ two-phase region. Figures 4-9(a), (b), and (c) are examples of these kernels. Similar techniques can be used for preparing the 150 micron kernels.

For the 1000 micron kernels, the GSP technique which relies on the diffusion of NH_3 into liquid drops to form the gelled kernels, is not applicable, since the diffusion path is too long for the gelling process to be completed within a reasonable dropping distance. Two other techniques have been used for

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URANIUM-CARBON-OXYGEN PHASE DIAGRAM (1500°C)

Fig. 4-8. Uranium-carbon-oxygen phase diagram (1500°C).

URANIUM OXYCARBIDE KERNELS, GREY PHASE = UO_2 , WHITE PHASE = SINGLE PHASE UC_xO_y

200X

(A) $\text{UC}_{0.66}\text{O}_{0.50}$
330 MICRONS

400X

(B) $\text{UC}_{0.44}\text{O}_{0.93}$
200 MICRONS

200X

(C) $\text{UC}_{0.16}\text{O}_{1.64}$
350 MICRONS

Fig. 4-9. Macro-photos of various kernels.

preparing kernels of such large diameter. The first is the powder agglomeration method which was used for the fabrication of porous UO_2 kernels (~800 micron diameter, 80% dense) for the DRAGON Project in England (Refs. 4-17 and 4-18). The second one is the internal gelation method developed at KFA (Refs. 4-19 and 4-20) for the fabrication of UO_2 and UC_2 kernels by the hydrolysis of concentrated $\text{UO}_2(\text{NO}_3)_2$ solution containing urea and hexamethylenetetramine (HMTA) in hot paraffin oil at 95°C or the precipitation of UO_2 sol containing HMTA under similar conditions. It has also been used (Ref. 4-21) for the preparation of uranium-plutonium carbide fuel kernels. The internal gelation method which avoids the handling of powder and offers better control of the uniformity of kernel size is preferred for the present application. $\text{UO}_2(\text{NO}_3)_2$ solution is the preferred starting material since the preparation of UO_2 sol is a rather complicated operation. Figure 4-10 and Figure 4-11 show, respectively, the various steps of the GSP process and the internal gelation process for the preparation of uranium oxycarbide kernels.

The GSP equipment is available at GA Technologies Inc. Uranium oxycarbide kernels of 200 micron diameters are routinely prepared. To prepare 150 micron kernels, adjustments in feed solution concentration, size of sphere-forming nozzle and other process parameters are required. Such work is currently in progress.

An internal gelation apparatus has been set up. Preliminary trial runs were made on the preparation of 1000 micron UO_2 kernels. Optimization of the feed solution composition and process parameters for the preparation of uranium oxycarbide kernels is being carried out.

Figures 4-12(a), (b), and (c) are, respectively, macro-photos of 200 micron uranium oxycarbide kernels prepared by the GSP method, 1000 micron UO_2 kernels prepared by the internal gelation method, and the same UO_2 kernels after drying but before sintering. These photos illustrate the difference in the sizes of these kernels and the large shrinkage in kernel volume when the dried UO_2 kernels are sintered.

INTERNAL GELATION PROCESS FOR THE FABRICATION OF URANIUM OXYCARBIDE KERNELS

Fig. 4-10. GSP process for the fabrication of uranium oxycarbide kernels.

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GSP PROCESS FOR THE FABRICATION OF URANIUM OXYCARBIDE KERNELS

Fig. 4-11. Internal gelation process for the fabrication of uranium oxycarbide kernels.

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MACRO-PHOTOS OF VARIOUS KERNELS

(A) 200 MICRON URANIUM
OXYCARBIDE KERNELS (6X)

(B) 1000 MICRON UO_2 KERNELS (6X)

(C) UO_2 KERNELS OF (B) BEFORE SINTERING (6X)

Fig. 4-12. Uranium oxycarbide kernels. Grey phase = UO_2 , White phase = single phase UC_xO_y .

4.4 Coated Particle Compact Fabrication

To reduce the maximum fuel temperature in the coated-particle fueled SP-100 core, it is necessary to improve the effective thermal conductivity of the coated particle system. This can be accomplished by adding refractory metal powder (molybdenum or tungsten) to the interstices of the particles and bonding them into a compact. Besides the improvement of thermal conductivity, this will also help to prevent the movement of these fuel particles in the core in a zero-gravity environment and the ratchetting of the core components surrounding the particles in a gravitational field during ground testing.

Although the development of the compact fabrication techniques cannot be initiated until the coated particles are available, some thoughts have been given to the possible avenues of approach to compact fabrication. The choice of molybdenum and tungsten powder as matrix materials for filling the space among the particles is based on their high thermal conductivity, compatibility with ZrC coating of the particles, and the close match of their thermal expansion coefficients with that of ZrC.

Two approaches are being considered for consolidating the particle-matrix powder assembly into a compact. The first involves the cold-pressing of the vibratory compacted particle-matrix powder assembly in a steel die into a green compact containing a central hole for accommodating the heat pipe. The pressure applied has to be lower than the crushing strength of the ZrC coating of the particle. The green compact is then sintered to form the compact. A fugitive sintering aid (such as nickel) will be added to the matrix powder to lower the sintering temperature required. After completion of the sintering operation, the sintering aid is removed by heating in vacuum.

The second approach involves the infiltration of the void space of the vibratory compacted particle-matrix powder assembly with tungsten or molybdenum by chemical vapor deposition techniques. The height of the particle-matrix powder assembly and the coating parameters will be optimized to insure good accessibility of the coating gas to the space between the particles and the

matrix powder. A chemical vapor deposition apparatus is being built for this purpose.

Both approaches will yield annular shaped compacts, which can slide onto the heat pipe. The gap between the compact and the heat pipe can be filled with molybdenum or tungsten by vapor deposition. Or the gap can be filled with fine molybdenum or tungsten powder to improve the heat transfer between the compact and the heat pipe.

Efforts on compact fabrication development will be initiated as soon as the coated particles are available. If the results show promise, then compact samples will be fabricated for irradiation testing.

- 4-1. A.F. Weinberg, L. Yang, and R.G. Hudson, Carbide Research as Related to Thermionic Emission, General Atomic Report GA-4392, 1963.
- 4-2. C.A. Alexander, J.J. Ward, J.S. Ogden, and G.W. Cunningham, "Thermodynamics for Compatibility Studies of Metal Clad Uranium Carbide," Carbides in Nuclear Energy, Vol. 1 (Macmillan, London, 1963) p. 192.
- 4-3. J.D. Allinson and J.C. Riviere, J. Nucl. Mater. 17, 97 (1965).
- 4-4. W. Chubb, J. Nucl. Mater. 23, 336 (1967).
- 4-5. V. Coen, H. Hausner, H. Kolbe, and D. Quataert, "Interaction between Uranium Carbide and Refractory Metals," Ceramic Nuclear Fuels (Am. Ceram. Soc, Columbus, Ohio, 1969) p. 147.
- 4-6. S. Katz, J. Nucl. Mater. 6, 1972 (1962).
- 4-7. T.D. Gulden and J.F. Watson, High-Temperature Fuels for Advanced Nuclear Systems, General Atomic Report GA-A16908, 1982.
- 4-8. R.E. Bullock, High Temp.-High Pressures 12, 599 (1980).
- 4-9. G.H. Reynolds, J.C. Janvier, J.L. Kaae, and J.P. Morlevat, J. Nucl. Mater. 62, 9 (1976).
- 4-10. A. Auskern, J. Nucl. Mater. 22, 257 (1967).
- 4-11. W.A. Stark, Jr. J. Nucl. Mater. 73, 169 (1978).
- 4-12. E.K. Storms, The Refractory Carbides (Academic Press, New York, 1967) p. 187.
- 4-13. R.E. Latta and R.E. Fryxell, J. Nucl. Mater. 35, 195 (1970).
- 4-14. R.G. Hudson, M.H. Horner, and L. Yang, "Some Investigations of Refractory Metal Systems of Thermionic Interest," 8th Annual Conference Record on Thermionic Conversion (IEEE, New York, 1969) p. 296.
- 4-15. A. Pialoux and M. Dode, "Contribution a l'Establissement du Diagramme de Phases du Systeme U-C-O a l'Aide de la Diffractometrie de Rayons X a Haute Temperature Sous Pression Controllee," J. Nucl. Mater., 56, 2, 221-28 (1975).
- 4-16. G. R. Chilton, "The Application of Thermochemical Principles to the Production of Nuclear Fuel Materials," pp. 80-88 in High Temperature Chemistry of Inorganic and Ceramic Materials, Spec. Publ. No. 30. Edited by F. P. Glasser and P. E. Potter. The Chemical Society, London, 1976.
- 4-17. R. Forthmann, A. Naoumidis, H. Nickel, and W. Bruck, "Investigations on the Preparation of UO₂ Microspheres by Internal Gelation of a UO₂ Sol and From a Uranium (VI) Solution," Jul-655-RW, Kernforschungsanlage Julich, Julich Nuclear Research Center, Institute for Reactor Materials, Federal Republic of Germany, April 1970.

- 4-18 C. R. Puschel, R. Forthmann, A. Naoumidis, and H. Nickel, "Investigations on the Production of Spherical Carbide-Containing Uranium Dioxide Particles by the Hydrolysis Process," Jul-1606, Kernforschungsanlage Julich, Julich Nuclear Research Center, Institute of Reactor Materials, Federal Republic of Germany, July 1979.
- 4-19 T. N. Washburn and R. C. Burnett, "Production of Porous UO_2 Kernels by Heat Treatment of Powder Metallurgy U_3O_8 Microspheres," D. P. Report 551 (1968).
- 4-20 R. Baschwitz, R. C. Burnett, and H. Chantot, "The Fabrication of Porous UO_2 Microspheres," D. P. Report 674 (1969).
- 4-21 K. Bischoff, V. Scherer, and H. Schumacher, "Sol-Gel Processes for the Preparation of U-Pu-Carbide Fuels," pp. 164-177 in Symposium on Sol-Gel Processes and Reactor Fuel Cycles, Gatlinburg, TN, May 4-7, 1970.

5. CORE DESIGN WITH COATED PARTICLES

The analyses reported in this section employ simplified models due to the limited scope of this study. The results are intended to aid the early evaluation of the coated-particle fuel concept as a feasible backup fuel for the SP-100 reactor.

5.1 NEUTRONIC ANALYSIS

The analysis of the coated-particle core is based on the current SP-100 reference core geometry so that direct comparison of results may be possible. The core geometry is shown in Fig. 5-1 (based on Ref. 5-1).

The objective of the neutronic analysis was to obtain the effective multiplication factor, k_{eff} , and the required fuel loading. The Monte Carlo neutron transport code KENO-IV (Ref. 5-2) was employed. A three-dimensional homogenized model was constructed. The core midplane radial and axial cross sections of the homogenized core are depicted in Figs. 5-2 and 5-3, respectively. In essence, the fuel region and heat pipes are homogenized. The core containment vessel and foil insulators are homogenized. The B_4C control drums were assumed to have rolled the absorber sectors outward and to be represented in the model as smeared into a ring outside of the radial beryllium reflector. The homogeneous model is judged to be adequate since SP-100 reactors are very small fast reactors.

For such a small fast reactor, the dominant neutron spectrum falls within the keV to low MeV energy range. Therefore, a sixteen-group, neutron cross-section set was deemed fine enough for this analysis. The GA cross-section code MICROX-2 (Ref. 5-3) was utilized to obtain the input cross-section set. The SP-100 reference reactor has a large neutron leakage fraction and a very hard neutron spectrum. Thus, no strong anisotropic scattering is anticipated. For these reasons, only P_0 and P_1 cross sections are used in the KENO calculations. It is noted here that no detailed rhenium cross sections could be generated quickly, therefore the cross sections for tungsten were generated by

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SP-100 REFERENCE CORE GEOMETRY

Fig. 5-1

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RADIAL CROSS SECTION OF THE HOMOGENIZED SP-100 REACTOR

Fig. 5-2

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AXIAL CROSS SECTION OF THE HOMOGENIZED SP-100 REACTOR

Fig. 5-3

MICROX-2 and then scaled to the rhenium values according to the data contained in the barn book, BNL-325 (Ref. 5-4). Since the total content of rhenium is small, no significant error is expected from this approximation.

The analysis considers an early design of the particle fuel, that can load the same amount of uranium as loaded into the SP-100 reference core. The early oxycarbide fuel is composed of 55 v/o of UC and 45 v/o of UO_2 . The particle coating is ZrC and the interparticle matrix material is molybdenum.

The k_{eff} results are listed in Table 5-1. It is seen that, to achieve criticality, ~ 2% less loading of the assumed coated-particle fuel is required since k_{eff} of 1.07 is ~2% greater than the 1.05 value of the SP-100 reference core. Compared with the reference core, the additional 2% k_{eff} of the coated-particle-fuel core is attributed to the large reduction in molybdenum and the introduction of a sizable amount of carbon which enhances slowing down of neutrons.

The final coated-particle fuel design required a high fraction of UC in the fuel to achieve criticality when loaded into the reference core configuration. We can conclude that the coated-particle fuel is capable of achieving the same fuel loading provided by the reference fuel in the SP-100 core configuration and thus is comparable to it from a neutronic point of view.

5.2 THERMAL ANALYSIS

The purpose of this analysis is to obtain a first estimate of the maximum fuel temperatures and associated temperature gradient of a coated-particle fuel compact during normal and faulted operating conditions. Due to the very limited scope of this work, a two-dimensional finite-difference code TAC2D (Ref. 5-3), instead of a finite-element code, is utilized.

A one-twelfth reactor sector by symmetry is considered and is shown in Fig. 5-4. The figure illustrates individual heat pipes and their associated fuel regions. This is translated into a finite-difference model as depicted in

Table 5-1

EFFECTIVE MULTIPLICATION FACTOR, K_{EFF}
(CONTROL DRUMS ROLLED OUT AT BOL)

MONTE CARLO		COATED-PARTICLE
CALCULATIONS	REFERENCE SP-100	CORE
LANL'S MCNP		
CALCULATION	1.05	—
GA'S KENO		
CALCULATION	1.05	1.07

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A ONE-TWELVETH SYMMETRIC REACTOR SECTOR OF SP-100

Fig. 5-4

Fig. 5-5. For conservatism, the circumference of the modeled heat pipe is made identical to the reference core design value. This makes the cross-sectional area of the modeled heat pipe smaller than the reference design value, and correspondingly, it leaves a larger-than-actual area fraction of the modules for active fuel. This results in a longer average length for heat conduction from the fuel to the heat pipes and consequently leads to the calculation of somewhat higher fuel temperatures toward the edges of fuel modules than should be expected. The alternative assumption of using actual heat pipe and fuel area fractions in the model geometry results in the perimeters of the heat pipes being larger than actual and would have led to the non-conservative calculation of fuel temperatures that are lower than should be expected.

The calculations were carried out for an early design of the oxycarbide fuel having the composition and volume fractions in the particle fuel compact as shown in Table 5-2.

The effective thermal conductivity of a particle-fuel compact varies with its material composition. This analysis considers only the conductivity of the above stated fuel compact and therefore gives only one result in the spectrum of the oxycarbide fuels.

The Cheng-Vachon method (Ref. 5-5) is employed for estimating the effective thermal conductivity of the particle fuel compact. According to Ref. 5-5, the method was verified against experiments and should be reliable. The individual thermal conductivities, k , used in this computation are listed in Table 5-3.

The calculated effective thermal conductivities, k_e , at the approximate normal and faulted fuel temperatures are about the same. This results from very balanced tradeoffs in the increased and decreased individual conductivities in carbides and non-carbides respectively at the different temperatures. The balanced tradeoff is largely true over the entire temperature range between the normal and faulted fuel temperatures since thermal conductivities vary quite linearly at high temperature. What this means is that the fuel tempera-

5-9

Fig. 5-5. Circular finite difference model of a one-twelfth sector of the SP-100 reactor.

Table 5-2

ASSUMED COMPOSITION OF COATED PARTICLE FUEL COMPACT

		Volume %
Fuel	UC	33
	UO ₂	27
	(UC:UO ₂ = 55 v/o:45 v/o)	
Coatings	Pyrocarbon and ZrC	14
Pores and Voids inside Particle Coatings		10
Matrix	Mo	10
Matrix Interstices	Void	6

Table 5-3

MATERIALS AND THEIR EFFECTIVE THERMAL CONDUCTIVITIES

	kW/cm.K (BTU-hr-ft-R)	
	At 1600 K	At 2350 K
k-UC	0.2423 (14)	0.2597 (15)
k-UO ₂	0.0208 (1.2)	0.0182 (1.05)
k-Mo	0.8309 (48)	0.7443 (43)
k-ZrC	0.3803 (22)	0.4328 (25)
k-pore	0.0007 (0.04) assumed	0.0007 (0.04) assumed
Calculated k _e	0.2077 (12)	0.2095 (12.1)

ture will not rise due to the lower conductivities of non-carbides at high temperature but only due to the loss of a failed heat sink (i.e. the failed heat pipe).

The effect of the content of UC on the effective thermal conductivity of the oxycarbide fuel at 1600K is shown in Figure 5-6. The difference in conductivity between 55 v/o and 100 v/o UC is about 30%. The absolute scale of this curve is affected not just by the fuel but also by the other materials, especially the pores since they have extremely low conductivity. The conductivity of the pores can be significantly increased by adding helium. Although fission gases will be released into the pores inside the particles after start-up of the reactor, the effective thermal conductivity of the gas mixture will still be much larger than the pore conductivity employed in this study (k-He at 1600K is about 0.005 W/cm-K, and for fission gas, 0.0003 W/cm-K). It is also noted that the fuel module is assumed to be a well bonded unit and that no significant gaps exist between the kernels and their coatings, the coated particles and the matrix materials and the fuel compact and the heat pipe. It is assumed that radiation is the only mode of heat transfer between modules.

SP-100 reference core was first designed by LANL with radial power zoning by varying the fuel layer thickness (see Figs. 5-1 and 5-4) to obtain approximately constant power input per heat pipe. Current LANL thermal calculations employ a set of power densities that takes into account the core power peaking factor of 1.2 (Ref. 5-6). Our analysis employs the same power input. The power densities are listed in Table 5-4. No gamma heating of the central plug is included in either the coated-particle fuel core or the SP-100 reference core analyses.

The power densities in Table 5-4 are adjusted by the ratio of the SP-100 reference core fuel area to the TAC2D modeled fuel area for every fuel module so that the total heat generation per module is identical in coated-particle fuel and the reference SP-100 cases. The adjusted power densities are shown in Table 5-5. Although the TAC2D model (Fig. 5-5) does include the modeling of the radial reflector and control drum, the inner surface of the foil-insulators

ESTIMATED EFFECTIVE THERMAL CONDUCTIVITY OF AN OXYCARBIDE PARTICLE FUEL COMPACT AT 1600 K

Fig. 5-6

Table 5-4

SP-100 POWER DENSITIES IN VARIOUS FUEL LAYERS

Fuel Layers	Power Densities (kW/cm ³)
1 (inside)	0.1148
2	0.1083
3	0.0998
4	0.0879
5 (outside)	0.0767

Table 5-5

ADJUSTED POWER DENSITIES FOR TAC2D INPUT

Fuel Layers	Adjusted Power Densities	
	kW/cm ³	BTU/hr-ft ³
1	0.09854	9.5211x10 ⁶
2	0.09402	9.0835x10 ⁶
3	0.08833	8.5345x10 ⁶
4	0.08053	7.7810x10 ⁶
5	0.06983	6.7465x10 ⁶

are treated as adiabatic surfaces in the same manner as in the current SP-100 reference core model.

The emissivity of the surfaces of the particle-fuel module is not known at this time. The surface can be designed such that a relatively high emissivity may be attained. In the mean time, parametric variation of emissivity is carried out for the faulted heat-pipe case. Emissivity is not a significant factor in the normal operating case. The maximum fuel temperatures obtained from parametric treatment of the emissivity are summarized in Table 5-6. The case of faulted heat pipe in the outer fuel layer (Layer 5, Fig. 5-5) is not considered in this study. This decision is based on two reasons. First, the current SP-100 reference reactor thermal model does not include heat loss through the radial reflector, and too many uncertainties in input data exist. Second, the TAC2D model (Fig. 5-5) is a 30° sector of the core while the reference core study has a 60° sector. Analysis of a faulted heat pipe (e.g. by failing heat pipe No. 11 as in the SP-100 reference core study) in a 30° model may be unduly conservative when compared to a 60° model. Failure of heat pipe No. 10 will leave only one fuel module with operating heat pipes flanking it on each side in a 30° model, while in the 60° model there will be two rows of modules with operating heat pipes flanking it on each side. Heat from a fuel module with a failed heat pipe may also pass to the next modules beyond those adjacent to the failed-heat-pipe module. However, the amount of heat and its effect probably will not be pronounced on such modules.

It is recalled that the composition of the coated-particle fuel compact is based on an early fuel particle design. The final design requires almost exclusive use of UC fuel. This enhances the effective thermal conductivity of the fuel compact by about 30% (See Fig. 5-6). Better geometry modeling would also improve heat transfer calculations. When these factors are considered, the expected maximum fuel temperatures may be assessed as shown in Table 5-7.

The coated-particle fuel is quite forgiving under faulted heat-pipe conditions. The effective thermal conductivity (Fig. 5-7) is quite constant over the normal to faulted temperature range as stated above. This is the principal

Table 5-6

TAC2D RESULTS - MAXIMUM FUEL TEMPERATURES

	Maximum Fuel Temperature		Location
	K	Degree R	
Normal Operation	1763	3173	Module No. 11 (Layer 5) at outer corner next to Module No. 8
Failed Second Layer Heat Pipe (Heat Pipe No. 3)			
* $\epsilon = 0.9$	2094	3769	Module No. 3 (Layer 2)
$\epsilon = 0.6$	2286	4115	at outer corner next to
$\epsilon = 0.3$	2674	4813	Module No. 1

* ϵ = Surface emissivity of the fuel module. Note that the current SP-100 reference core results is 2269K at an unreported emissivity.

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TEMPERATURE DEPENDENCY OF EFFECTIVE THERMAL CONDUCTIVITIES

Fig. 5-7

reason why the maximum temperature of the coated-particle fuel for the faulted-heat-pipe case (Table 5-7) is significantly lower than that of the SP-100 reference core.

Finally, it is worth noting and summarizing the differences, uncertainties, and limitations of the thermal analytical models, methods, and data bases used in calculation of the temperature distributions of both the coated-particle compact and tile-and-fin stack based SP-100 cores. These are listed as follows:

- o 30° model (coated particle core) vs. 60° model (SP-100 reference core).
- o Finite difference calculations of the (coated particle core) vs. finite element calculation of the (SP-100 reference core) and the larger fuel area modeled in the coated particle core analysis.
- o Uncertainty in effective thermal conductivities in both analyses.
- o Exclusion of heat rejection through radial reflector in both models.
- o Uncertainty in the emissivity of the fuel compact surface and the potential emissivity degradation over the life of either of the SP-100 cores.
- o Exclusion of gamma heating in the central plug in both models.

It is considered critical that these difficulties be resolved to verify the advantages indicated by the use of coated-particle fuel.

5.3 SP-100 CORE STRUCTURE ANALYSIS

Scoping structural analyses of the SP-100 coated-particle core have been performed for normal operating conditions and for operation with failed heat

Table 5-7

EXPECTED MAXIMUM FUEL TEMPERATURE

Normal Operation(1)	1660 K
Failed Second Layer	
Heat Pipe (2)	
$\epsilon = 0.8-0.9$	~ 1900 K
$\epsilon = 0.6$	~ 2050 K
$\epsilon = 0.3$	~ 2285 K

Note (1): Use of nearly 100% UC fuel increases effective thermal conductivity by about 30% and drops the calculated maximum fuel temperature by ~75 K. Modeling of heat pipe area is ~20% conservative, accurate modeling reduces this temperature further by ~25 K.

Note (2): Same reasons as Note (1). Note that the effective thermal conductivity of the coated-particle fuel compact is relatively invariant with respect to temperature due to the increase in conductivity of carbides which compensates for the conductivity reduction of UO_2 and Mo at high temperature. This helps mitigate the faulted heat pipe conditions.

pipes. Although further work will be required to develop details of the design, the conclusion of these scoping studies is that the basic concept is feasible and may offer some advantages over the reference SP-100 core.

An inherent feature of the coated-particle core concept is that the fuel swelling and fission products are entirely contained within the coated particles. Thus the fuel compact is expected to be dimensionally stable for normal and faulted operating conditions. This simplifies and reduces the uncertainty in the structural analysis considerably, since one need only be concerned with predictable thermal expansion distortions, rather than having to consider the much-more-difficult-to-predict phenomena of fuel swelling and redistribution.

In the following paragraphs various thermomechanical distortions are considered. For normal operation, module distortions, differential thermal expansion between the heat pipe and the fuel compact, and differential thermal expansion within the fuel compact are discussed. For operation with failed heat pipes, module distortions are investigated.

One of the major features of the coated-particle core concept is the good thermal conductivity of the fuel compact. This minimizes the thermal gradients within the core, the potential thermal distortions, and thermal stresses. During normal operation the fuel modules are expected to experience a maximum layer-to-layer temperature difference across the module of less than 25K. The resulting free bow of the module is estimated to be a maximum of about 0.1 mm, which is easily accommodated by the 0.5 mm clearance provided around each module. Even if this movement were partially restrained, the resulting stresses would be low and no damage to the module would be expected to take place.

Although the thermal expansion coefficients of the fuel compact and the heat pipe are designed to be well matched, there will be a difference in their net thermal expansions because they operate at different temperatures. It has been estimated that a maximum temperature difference of 100 to 200K will exist

during normal operation between the fuel compact and the heat pipe. This will result in a maximum thermal expansion difference between the fuel compact and the heat pipe of about 0.28% or about 0.8 mm over the length of the core. This can be accommodated by dividing the fuel axially into segments and providing a small thermal gap between segments. If ten axial segments were used a gap of 0.1 mm between each segment would be more than adequate to accommodate differential axial expansions during normal operation.

A temperature difference will also exist between the heat-pipe interface surface and the outer surfaces of the fuel compact. It has been estimated that this temperature difference will have a maximum value of about 200K and will probably be less. This temperature difference will cause axial and circumferential stresses to be developed in the fuel compact. The stresses will be compressive in the outer portion of the compact and will be tensile in the inner regions adjacent to the heat pipe. Without detailed stress analysis it is impossible to precisely determine the magnitude of these stresses. However, an estimate of stress levels was made by representing the fuel compact by a cylinder having a temperature gradient through its wall. Away from the wall, the axial and circumferential maximum stresses will be equal and can be evaluated from:

$$\sigma = \frac{\alpha T \Delta E}{2 (1 - \nu)}$$

where

- σ is the stress,
- ΔT is the temperature difference across the wall,
- α is the coefficient of thermal expansion,
- E is the modulus of elasticity,
- ν is the Poisson's ratio.

Structural properties of the fuel compacts are not known at this time so an estimate was made using the mechanical property values of ZrC. For the maximum predicted temperature difference of 200K, the maximum predicted stress is about

375 MPa or about 54 ksi. The allowable stress for the fuel compact is not known, but it is probable that this calculated stress is excessive.

The actual stresses in the fuel compact will be reduced considerably by providing stress relief in the form of gaps where stresses are excessive. Axial segmentation of the fuel compacts will reduce axial stresses, and if needed, radial slots in the compacts can be used to reduce circumferential stresses. Further analysis and materials testing are required to develop design details here. It is of interest to note, however, that because the outer portions of the fuel compact are in compression and the inner portion is in tension, the fuel is inherently self stress relieving in a manner that is safe and non-propagating. That is, axial stresses can be relieved by cracks developing in planes perpendicular to the axis of the compact, with the zone of cracking limited to the inner regions where tensile stresses exist. Similarly, the circumferential stresses can be relieved by radial cracks, but these will also be limited to the inner region of the fuel where tensile stresses exist. Such cracks, where they do occur, would not propagate, and would not result in the formation of loose pieces.

The SP-100 core must be also designed to operate with one or more failed heat pipes. A major concern for this case is whether the temperature differences which would occur across the modules immediately adjacent the failed module would cause distortions which could lead to propagation of the initial failure.

Thermal analysis of this case has been performed and shows that due to the good thermal conductivity of the coated particle fuel compact the maximum temperature difference across the outer surfaces of the unfailed modules will be limited to about 100K. The resulting free bow of the module is estimated to be about 0.4 mm. In the current design concept about 0.5 mm of free gap is allowed on all sides of the module so that minimal mechanical interference should occur as a result of heat pipe failures.

5.4 SAFETY IMPLICATIONS

Consideration has been given to the major safety questions related to nuclear reactor space power, but no quantitative analyses have been performed in this initial period of evaluating the potential of coated particles as an alternative SP-100 core fuel. The accident conditions considered here are: (1) the potential for criticality upon immersion of the core in rocket propellant or fresh and sea water, (2) solubility of fuel and dispersion of radionuclides in the immersed condition, (3) launch pad fire and explosions, and (4) re-entry disassembly and burnup. A brief review of the safety implications of particle-type cores with respect to these accidents follows.

5.4.1 Criticality When Immersed in Water

Immersion of the core in propellant, fresh or sea water could be the result of an abortive launch. In this condition, the particle-fueled core is expected to be similar in design to the SP-100 core. Detailed calculations could not be performed in the limited time available, but are required to develop the details of the design that will be required. It is expected that for a core of the same size, such as those that have been described above, a slightly greater amount of poison will be needed to maintain subcriticality than is needed for the SP-100 core. The extra poison needed results from the substitution of UC in the kernel and ZrC in the coating of the particle fuel for some of the Mo in the fins of the tile-and-fin stack of the SP-100 fuel modules. Approximately one-half of the volume of the Mo is replaced. The carbon in the UC and ZrC is a better moderator than Mo or W and less of a neutronic poison in a thermal neutron spectrum. However, LANL analyses (Ref. 5-7) have shown that the neutron spectrum is still quite hard even when the SP-100 core was immersed in water. Thus, the small spectral shift may reduce the effects of the carbon substitution to a very small difference. The importance of the differences can only be determined by the detailed calculations.

5.4.2 Immersion Solubility and Dispersion

The environmental impact of a subcritical core immersed in water will depend upon the capability of the core to contain radionuclides of fuel. Assuming the coated particles are held in a matrix of Mo metal, the fuel would be highly resistant to dissolution and dispersion by immersion. Compromise of the Mo barrier by impact on landing and corrosion by the water are the likely mechanisms. Only a small fraction of the fuel particles are likely to be exposed when a compact is rended. Fuel within unbroken, but exposed particles, continues to be protected from the water by the ZrC coating which is neither soluble nor corrosive in fresh or sea water. Those particles along the rended surface with broken coatings will be exposed to the water. The UC will be hydrolized and dispersed, more or less, depending on the detailed and generally complex physio-chemistry of the environment.

Corrosion of Mo or W is a relatively slow process although initially it will proceed relatively rapidly until inhibiting layers of corrosion products are formed.

5.4.3 Launch Pad Fire and Explosions

Launch pad explosions and fires are difficult to analyze. Such analyses are beyond the scope of this program at this time. The melting point of Mo is about 2900K and that of ZrC is about 3800K. UC melts at ~3000K. These are very high temperatures not readily reached by chemical combustion in the short duration of most launch pad fires. The fuel particles, as individual pressure vessels, will retain the fuel until quite high temperatures are reached. As virgin or very low burnup fuel, there is no or very little fission gas to exert pressure on the coating. Only a fraction of particles are likely to reach the fracture point.

Mechanical damage is likely to be quite limited by a combination of the matrix metal of the compact and the ZrC coatings. Only the surfaces exposed by mechanical damage will release fuel directly. The bulk of the particles, con-

tained in the pieces of the compacts, will retain their fuel. Compared with other fuel forms, particle fuel will release much less fuel material to the environment.

5.4.4 Re-Entry Disassembly and Burnup

The reactor core will be designed for disassembly in the event of premature re-entry from Earth orbit. The kinetic energy of the pieces of the core will be converted to heat as they enter the Earth's atmosphere. Burnup of the fuel assemblies to sub-micron size particles to form an aerosol which would result in dispersion of the debris in the huge volume of the upper reaches of the atmosphere and in dilution to low concentrations. Significant further delay (years) will occur as the dilute dust settles to Earth.

The high temperature capability of the particle fuel materials may make burnup and dispersion more difficult. Melting of the matrix material would release the fuel particles. However the fuel particles are too large to form aerosols. It is not certain, without detailed aerodynamic heating analysis, to know whether the spherical fuel particles in combination with exothermic chemical reactions will burnup to submicron-sized particles and become aerosols.

- 5-1. Ranken, W. A. (LANL), personal communication, October 8, 1982.
- 5-2. Petrie, L. M., and N. F. Cross, "KENO IV, An Improved Monte Carlo Criticality Program," ORNL-4938, November, 1975.
- 5-3. Boonstra, R. H., "TAC2D, A General Purpose Two-Dimensional Heat Transfer Computer code," GA-A14032, July 15, 1976.
- 5-4. Garber, D. I., and R. R. Kinsey, "Neutron Cross Sections, Volume II, Curves," BNL-325, Third Edition, January, 1976.
- 5-5. Cheng, S. C., and R. I. Vachon, "The Prediction of the Thermal Conductivity of Two and Three Phase Solid Heterogeneous Mixtures," Int. J. Heat Mass Transfer, Vol. 12, 1969, pp. 249-264.
- 5-6. Girrens, S. (LANL), personal communication, December 3, 1982.
- 5-7. Project Staff, SP-100 Project Semi-Annual Technical Progress Review, (April 1 - Sept. 30, 1982), Los Alamos National Laboratory, Oct. 1982.

6. SUMMARY AND CONCLUSIONS

The overall conclusion to this Phase I study is that our preliminary analyses show coated-particle fuel to be the basis for a viable alternative SP-100 core. We have shown that fuel particles with kernels of UO_2 , UC, and combinations of the two with ZrC coatings can be designed to produce compacts that achieve critical assemblies, at least as small as the reference SP-100 core. Coated-particles of UO_2 and mixed UC- UO_2 fuel have been previously fabricated with ZrC coatings at GA. Nothing has been found to cast significant doubt on the feasibility of coated-particle fuel for SP-100. In order to verify our analytical conclusions and establish coated particles as an alternative SP-100 fuel, we will need to demonstrate the ability to fabricate the fuel compact and to bond the compacts to the heat pipes. Also the coated particles, compact, and fuel module designs must be verified by irradiation testing.

In the following subsections we present a more detailed list of our findings, and recommendations for further work.

6.1 SUMMARY OF FINDINGS

Our findings are summarized in the two lists shown below, one for coated particle fuels and the other for core design.

Coated Particle Fuels

- o All work performed to date supports the feasibility of coated-particle fuel for space power applications.
- o Coated particles with UO_2 , UC_xO_y and UC kernels and ZrC coatings have been designed to meet SP-100 design requirements.
- o A two particle size system (~1000 micron and ~150 micron) with UC kernels meets the fuel density requirements for SP-100.

- o Refractory metal coatings have been eliminated because of creep-rupture limitations.
- o Previous irradiation tests confirm the good performance of ZrC coatings and their compatibility with the fuel.
- o Sol gel processes have been demonstrated for producing 1000 micron UO_2 and 200 micron uranium oxy-carbide kernels.
- o Refractory metals appear to be the best choice for matrix materials but the processes are yet to be developed.

Core Design

- o The k_{eff} of the coated-particle core of the same configuration can be somewhat greater than that of the reference core.
- o Preliminary studies show that maximum fuel temperatures for the normal and failed heat pipe operations of the alternate core are expected to be less than those of the reference core.
- o Mechanical analysis of the module need only consider thermal distortions so uncertainties in analysis will be small. Distortions due to swelling and fuel relocation are expected to be insignificant.
- o Bowing of fuel modules during normal and failed-heat-pipe operation will be small enough to be accommodated.
- o Differential thermal expansion will require axial segmenting of fuel compacts.
- o The replacement of some Mo with ZrC in the alternate core may require additional poison during launch to assure subcriticality

during a water immersion accident.

- o Dispersion of uranium from coated-particle compacts as a consequence of immersion and explosion accidents will be relatively small.

6.2 CRITICAL ISSUES

There are a number of critical issues that have been identified for the tile-and-fin-stack design of the reference SP-100 core. The coated-particle compact compact design of the alternative core has the potential to alleviate the following critical issues of the reference SP-100 core

- o Swelling of fuel
- o Fuel migration
- o High fuel temperature under faulted heat pipe operation.

In this study there are also a number of critical issues that have been identified and which must be resolved if the coated-particle core is to be established as a credible alternative to the reference core. In this subsection the significance of these critical issues will be reviewed. In the recommendations to follow in the next subsection, the actions needed for their resolution will be indicated.

Critical Issues Identified (Coated-Particle Core)

1. Fuel particle and compact fabrication
2. Particle and compact thermal conductivity, emissivity and structural properties
3. Integrity of coated-particles at faulted design temperatures
4. Thermal bonding at particle-matrix and matrix-heat pipe interface
5. Reactivity hold-down safety feature design
6. Chemical and aerodynamic reentry burnup of fuel particles.

6.2.1 Fabrication of UC Coated Particles and Fuel Compacts

UC particles coated with layers of pyrolytic carbon and ZrC of 1000-micron and 150-micron diameters are required for space power reactor cores. Fuel particles of UC- UO_2 fuel have already been fabricated at GA on another program while fabrication of coated particles of pure UC has yet to be demonstrated. Fabrication of the small size fuel kernels requires minor modification of the gel-supported precipitation process while fabrication of the large size kernels requires some extension and development of the internal gelation process.

Compact fabrication requires the filling the inter-particle voids with Mo or W to form a matrix of metal containing the particles. Two development approaches are suggested: (1) introduce Mo or W powder to the bed of fuel particles along with a sintering aide and then sinter the powder in-situ and (2) fill the inter-particle voids of the compacted particles by chemical vapor deposition. Development of a process is required for bonding compacts to heat pipes with good thermal and structural bonds. Compaction and sintering of Mo powder in a small gap between the compact and heat pipe is one suggested approach. "Shrink fitting" to assemble the two pieces is another suggestion.

6.2.2 Irradiation Testing of UC Coated Particles

The design of the UC coated particles must be verified in the SP-100 core environment over test design life or to full burnup. Some types of irradiation testing of particles can be relatively easy and quick, and not costly. Similar irradiations have been routinely conducted in the High Flux Irradiation Reactor (HFIR).

6.2.3 Measurement of Fuel Particles and Compact Properties

The thermal conductivity of the particle-fuel compact determines the maximum temperature level in the core and the temperature differential across the fuel modules. This is especially critical under failed-heat-pipe conditions. Thermal stresses and fuel assembly bowing are directly affected. The thermal

conductivity of the compacts should be verified by measurement as soon as compacts can be made available.

Heat escapes from a fuel module with a defective heat pipe primarily by radiation across the gap between its outer surfaces and those of adjacent fuel modules. The temperatures of the radiating surfaces depends on their emissivities. High emissivities at high temperature are difficult to achieve even with the use of coatings. There is considerable uncertainty in emissivities because small surface changes or contaminants can alter their values significantly. Measurements are needed under well controlled conditions that extend over significant times relative the SP-100 mission life. Also alternatives to pure radiation heat transport, such as packing the inter-module gaps with high-conductive, resilient metal sponge, need consideration.

Mechanical properties of both the coated particles and compacts must be measured to verify their design and fabrication processes and to provide input information that can be used with confidence in further design and analyses. These properties include thermal expansion coefficients, Young's modulus, and strength all as functions of the particles and compact temperatures.

6.2.4 Thermal Stability of Coated Particles

The thermal stability of fuel particles must be measured at the normal, faulted, and over-design temperature conditions. The particles are expected to be stable at normal operating conditions based on experience with other particles. Performance over a 7 year life for space application requires verification, as does stability at faulted-heat-pipe conditions. The margins between operating conditions and stability limits are necessary design information.

6.2.5 Immersion Reactivity Control

Coated-fuel-particle cores of the same size as the SP-100 reference core requires slightly less U-235 loading for the same cold, clean reactivity. Upon immersion in water, the neutron spectrum shifts to a lower average energy, but

higher reactivity. The higher reactivity must be compensated by neutronic poison that is present. However, the Mo content of the coated-particle fuel core is only half that of the SP-100. Thus, additional temporary absorber may be required to maintain the particle-fuel core subcritical upon immersion. Additional design development will be needed to assure subcriticality of particle-fuel cores.

6.2.6 Chemical and Aerodynamic Burnup

Assuming the fuel particles separate from the matrix metal once the melting point of the metal is reached, heating of spherical particles by atmospheric friction and chemical oxidation must be analyzed. Assurance that the particles burnup so as to result in aerosol size particles that delay their return to Earth is the safety issue that requires early resolution in SP-100 development.

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