2048

Gulf General Atomic
Incorporated

P.O. Box 608, San Diego, California 92112

unclass
4/10/70
1/12/70

INFORMAL AEC RESEARCH
AND DEVELOPMENT REPORT

GAMD-9260
UC-10, Chemical Separations
Processes for Plutonium
and Uranium

HTGR FUEL REPROCESSING - HEAD-END TREATMENT OF
EXPERIMENTAL SMALL SCALE HTGR FUEL ELEMENTS

MASTER

Work done by:

L. H. Brooks
M. B. Feeney
C. L. Gray, Jr.

Report written by:

L. H. Brooks

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission
A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights, or
B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.
As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

This document, which was prepared primarily for internal use at Gulf General Atomic, may contain preliminary or incomplete data. It is informal and is subject to revision or correction; it does not, therefore, represent a final report.

P3763

DISTRIBUTION OF THIS DOCUMENT IS UNCLASSIFIED

Gulf General Atomic Project 317.
U. S. Atomic Energy Commission
Contract AT(04-3)-167, Project Agreement 17

September 9, 1969

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

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

CONTENTS

SUMMARY.	iii
INTRODUCTION	1
REVIEW OF BLOCK/PARTICLE SEPARATION PROCESSES.	1
Bonded Fuel Particles TRISO/TRISO	1
Blended Fuel Particles BISO/BISO or TRISO/BISO.	9
LABORATORY SCALE RESULTS AND PROGRAM	10
Whole-Block Crushing.	10
Screening	11
Tumbling or Crushing.	11
Burning	16
Alumina/Product Separation.	19
Grinding.	25
Leaching.	25
Filtration.	26
DISCUSSION	27
REFERENCES	31

SUMMARY

Methods available for debonding TRISO fuel particles from the graphite blocks have been reviewed and it is concluded that crushing, sawing and crushing, and whole-block burning are worth further development. In the case of blended BISO fuel particles, the simplest methods are either to cut the block in half and shake the fuel particles out of the holes, or to crush the block and screen the crushed material. GGA is concentrating on crushing as a first step followed by continuous screening.

It is planned to burn particle-free graphite in a fluidized bed or shaft kiln and the debonded particles (plus a small quantity of graphite) in a fluidized bed. In order to collect design data, a 2-in. fluidized bed has been constructed and will soon be operated.

Screening the fluid bed solids discharge may be difficult due to wear of the screen caused by sharp alumina particles. Alternatives are alumina spheres or separation by elutriation. Separation of fuel particles by screening is rapid and efficient providing the size gap is greater than 25 μm .

SiC coating cracking by grinding could be a problem if TRISO fuel particles are used in fuel elements. No work will be done on this problem until it is known definitely that the results will be useful.

Laboratory work has still to be done on leaching and filtration and the results used for scaled-up equipment construction.

The program for the next phase of work will be mainly concerned with equipment testing of each unit process on as large a scale as economically possible.

INTRODUCTION

The economics of a 15,000 MW(e) High-Temperature Gas-Cooled Reactor fuel cycle demand that a reprocessing plant be constructed for handling HTGR fuels only.^(1,2,3) The dissimilarities between this plant and one for reprocessing metallic-sheathed fuel elements from water reactors lie almost exclusively in the area of preparing the fuels for chemical separation, i.e., the head-end treatment. The most favored head-end treatment scheme is the grind-burn-leach process described in numerous reports^(4,5,6,7,8) and is the basis of the development work programmed in Section 5100 of the National HTGR Recycle Development Program Plan.⁽⁹⁾

This GGA document describes the progress made in batch laboratory-scale work (100 g) on the grind-burn-leach process and the plans for batch-continuous operation and equipment testing on the 1000 g scale.

Figures 1, 2, and 3 show the conceptual flow sheets for reprocessing TRISO/TRISO,^(a) BISO/BISO,^(b) and TRISO/BISO^(c) fuel particles, respectively. TRISO/BISO fuel, if it becomes a candidate, can be separated either by screening before burning if the particles are not agglomerated, or by screening after burning if the particles are agglomerated.

REVIEW OF BLOCK/PARTICLE SEPARATION PROCESSES

Bonded Fuel Particles TRISO/TRISO

A thorough survey was made of the many techniques for separating bonded fuel particles from graphite blocks that have been considered and/or tested. This review was conducted in order to eliminate from further consideration those techniques that have been found to be

-
- (a) TRISO fissile, TRISO mixed, recycle and fertile; bonded.
 - (b) BISO fissile, BISO mixed, recycle and fertile, blended.
 - (c) TRISO fissile, BISO mixed, recycle and fertile; blended.

Fig. 1. Head end block flow diagram
all TRISO feed bonded

Fig. 2. Head end block flow diagram all BISO or TRISO fissile/BISO others

impractical in a commercial application. Debonding processes that were reviewed include:

Debonding with

1. an O_2 , air, or steam lance
2. shock depressurization
3. a high-velocity water or steam jet
4. an abrasive jet

Mechanically removing the fuel rod by

5. wire brushing
6. trepanning
7. spade or crush drilling
8. punching

Destroying the block to free the particles by

9. whole-block burning
10. crushing
11. broaching and splitting

Gaseous Debonding

The gas-lance method has been tested using both O_2 and steam and results reported in an earlier quarterly report.⁽¹⁰⁾ The infeed was found to be 0.16 in./min under favorable conditions. Calculation shows that it would take 3 hr to debond a 30-in. block when a battery of 210 lances is used. Since the throughput in a 15,000 MW program is one block in 30 min, six batteries of 210 lances would be necessary. If one lance were to block or cease progressing for any reason, the fuel block would have to be removed, the cause of the blockage found and repaired, and production started again. Facilities must also be incorporated into the equipment to heat the fuel block to operating temperature, 750°C. This heating apparatus would also have to be easily serviced. The complexity of all this equipment operating remotely has rendered the gaseous debonding method unacceptable.

Shock Depressurization

This debonding method has been tried at GGA and results reported.⁽¹¹⁾ The graphite block and matrix are so porous that when the pressure of water, water and CO₂ mixture, or helium is released, the fluid merely flows out without dislodging the particles. This method, therefore, has been considered no further.

High-Velocity Water or Steam Jet

This has been tried at GGA and ORNL and results reported.^(4,11) When a jet of water at 5000 to 6000 psi is used, the infeed rate is quite good, but particle breakage is too high. Other disadvantages are that the water will become radioactive and will have to be recycled; this would then entail criticality limitations, filtering difficulties, and pump maintenance. As in the gaseous debonding method, a battery of jets would be required, and if one jet were to block, infeed would cease and the block would have to be removed to find the cause. This method, therefore, is being considered no further at GGA.

An Abrasive Jet

This debonding method suffers more disadvantages than the water jet in that abrasive particles would score the nozzle. Before the fluid could be recycled through the pump, it would need to be very carefully filtered. After the pump, abrasive particles would have to be reintroduced into the high-pressure fluid.⁽¹²⁾ This method is being considered no further.

Wire Brushing

This has been tried at GGA by spinning a cylindrical brush in an electric pistol drill. Results were reported.⁽¹¹⁾ The difficulties envisioned with this method are that the brush would eventually become clogged with matrix flour and the wire filaments would be wound in the form

of a helix, thus losing their capacity to dislodge the particles. If the brush were to eventually screw up into a ball, there would be no gap for the particles to drop past the brush. This method is being considered no further.

Trepanning

This technique entails operating a battery of cylindrical tubes each equipped with a cutter or saw at the top end. The block would be lowered onto the saws, and each fuel rod would be trepanned out, the cutting tubes being of larger diameter than the fuel rods. Difficulties envisioned are the drift of the tubes, the possibility of the blocks becoming bowed under irradiation, and the short life of the cutters.⁽¹²⁾ The resulting rod of fuel and graphite would still have to be broken down, with minimum particle breakage, for feed to the next stage. This method is being considered no further.

Spade or Crush Drilling

This method requires the use of a battery of drills which are designed to dislodge particles from the block wall and the matrix. The block end has to be either sawed off or drilled out first. There is a high probability that particles will be damaged during removal. Other disadvantages are the possible short tool life and the complexity of equipment.⁽¹²⁾ This method is being considered no further.

Punching

This method requires that the ends of the block be sawed off or the fuel rod plugs be drilled out first. A battery of punches would be required. The disadvantages are that if the fuel rod adheres even weakly to the graphite wall, then the pressure applied at one end would cause the particles to compress in the matrix and expand diametrically. It would then be difficult to press the fuel rod out without damaging it severely.⁽¹²⁾ This method is being considered no further.

Whole-Block Burning

Apparatus has been constructed at ORNL to investigate the possibilities of this method.⁽⁵⁾ The advantages are that it will receive any design of block and leaves only ash and particles as feed to the next stage. Temperature was controlled by mixing a variable fraction of CO₂ with the O₂ feed. The graphite burned away uniformly at all surfaces exposed to the O₂. Work is continuing at ORNL. This method could not be used with a BISO/BISO fuel in the block.

Sawing and Crushing

ORNL personnel have studied this method and find it relatively successful. The method is to saw the block down the coolant holes so as to divide into six pie-shaped segments. These are then fed to a hammer-mill to detach the fuel from the block prior to fluidized bed burning. One disadvantage is that this method depends on the coolant holes lying in a straight line across the block end without interposing fuel channels. If the block were to be redesigned such that this were not the case, sawing could not be used. Further work is being done to assess particle damage.

Whole Block Crushing

Crushing has been considered in conceptual reprocessing flowsheets at GGA.^(6,7) The block in this case is fed to a crusher without prior size-reduction. The advantage of this method is that any design of block can be crushed. This method is being developed at GGA using small-scale fuel elements.

Broaching and Splitting

This method of block disintegration has been considered and a few initial experiments have been carried out.

The scheme requires that a tube, with cutters fixed radially round the circumference, is pushed or pulled through the coolant channels. The

cutters, each 0.16-in. wide, cut grooves in the wall of the coolant channel. Six grooves are formed by a single pass of the broach. The grooves are cut in the graphite at points where the wall between the coolant channel and fuel channel are thinnest so that the bottom of the groove is only about 13 thousandths of an inch from the fuel channel.

After passing the broach through each coolant channel in the block, the thin graphite webs, dividing the fuel channel from the groove, are broken by splitting and the whole graphite structure breaks into pieces liberating the fuel from the fuel holes.

Splitting is carried out by pushing or pulling a splitter through the coolant channels. The splitter is a rod, equal in diameter to the coolant channel, and has six wedges, fixed radially on the circumference, which fit into the grooves. The wedge shape opens the groove and splits the thin web.

Trials carried out on 12-rod fuel elements have shown that broaching is easily performed. It was found that a groove can be cut to a depth of 0.16 in., in four steps each of 0.04 in. Each broach, therefore, had to be fitted with six radial cutters to groove to a depth of 0.04 in., six to groove to 0.08 in., six to groove to 0.12 in., and the final six to complete the groove. This system therefore required 24 cutters on each broaching bar.

The force required to pull the broach through the channel was 1600 lb.

Tool wear was determined by cutting a graphite cylinder on a lathe using a tool equivalent to a broach cutter. It was found that each cutter is good for 37,000 feet of groove length before the force required to pull the broach begins to increase.

Splitting the block after broaching was found to have the problem that as the webs broke, the block expanded. If the block was not restrained during splitting, it tended to fall apart and the wedges could jump out of the grooves.

This method was considered to require rather complex equipment but was sufficiently successful to be considered as a back-up method of graphite particle separation.

Blended Fuel Particles BISO/BISO or TRISO/BISO

The present reference fuel for an HTGR reactor is fissile BISO/Fertile BISO, contained in the fuel channels. The fuel particles, in this case, must be separated before burning if U^{236} and U^{233} are not to be mixed.

Sawing and Screening

A method described⁽⁸⁾ is to cut the block perpendicular to the length into two halves and shake each half with the open ends pointing downward. The particles are dislodged and screened to separate fissile and fertile. A disadvantage could be that if the particles agglomerate they may not be dislodged. It may be necessary to introduce an extra processing step to separate particles that are stuck together, e.g., washing in nitric acid. The probability of particles agglomerating cannot be assessed until blended particles have been irradiated to about 20% burnup.

Whole-Block Crushing

The whole block can be fed to a crusher and the crushed material screened. This method is very simple but as in the sawing and crushing concept, agglomeration could be a problem. Whole-block crushing experimental work being done on TRISO/TRISO fuel will be applicable to whole-block crushing BISO/BISO fuel.

LABORATORY SCALE RESULTS AND PROGRAM

Whole-Block Crushing

The crusher to be used for crushing the whole block should be robust, capable of operating for years without maintenance, break only few particles and produce a product which can easily be separated by screening into a fuel-rich fraction and graphite-rich fraction as indicated on Figs. 1, 2, and 3. Crushers considered suitable for this job are: jaw crusher, hammer-mill, and single-roll crusher.

The program to decide which crusher will be finally chosen is as follows: three sizes of fuel elements, i.e., 1-rod, 12-rod, and full size, will be crushed in small and large crushers of each type. The response from each experiment will be the fraction of broken particles and the fraction of particle-free burnable carbon (graphite, matrix, outer PyC) separated by subsequent screening. The results will show the effect of fuel element

size, and type and size of crusher on the responses and a decision will be made on which crusher is most suitable for crushing the full-size fuel element.

Experimental work to date has been done on crushing 1-rod fuel elements in a 3-in. jaw crusher and 1- and 12-rod fuel elements in a hammer-mill. Results of crushing and screening are shown in Tables 1, 2, and 3. The fractions of fines carbon and broken particles are shown in Table 4.

Crushing 1- and 12-rod fuel elements in a larger jaw-crusher is programmed to be done in the near future. A single roll crusher that will handle radioactive material has not yet been located.

Screening

The screen must be capable of separating the various size fractions from the crusher. Because dust will be created in the crusher and on the screen, it must be enclosed and its atmosphere extracted through a filter. In addition, broken and blinded screens must be detected early and be easily serviced. Vibrating or revolving screens would be capable of handling the job and inquiries are being made with a view to selecting and buying one for equipment testing.

Experimental work to date has been done using 3-in. or 8-in. diam. test screens.

Tumbling or Crushing

Tumbling has been used in the laboratory work as a step to break down oversize particle agglomerates, which are too large to feed to a fluidized bed, into $-1/8$ or $-1/4$ in. lumps without breaking down the graphite. A screening step then enabled the graphite and particles to be separated, the overflow graphite reporting to the particle-free graphite fraction and the particles to the fluidized bed feed. A disadvantage is that the process is not definite, i.e., some agglomerates have a chance of being carried along with the overflow graphite. The two processes, tumbling and screening, could be combined into one by operating a revolving screen with flights round the inside of the circumference to lift the feed and allow it to fall, i.e., a Bradford Breaker.

L.H.B. 7/23/69

Table 1
DISTRIBUTION OF CRUSHED PRODUCT WHEN CRUSHING
1-ROD FUEL ELEMENTS IN A 3-in. JAW CRUSHER

Run No.	Jaw Gap (in.)	Wt-% Product										Loss	Remarks ^(a)
		+5/8 in.	+1/2 in.	+5/16 in.	+1/4 in.	+1/8 in.	+300 μ m	-300 μ m	+250 μ m	-250 μ m			
2	1/2	---	46.7 ^(b)	22.1	---	12.4	18.3	0.3	---	---	0.10	TRISO/TRISO Bonded. Crushed at GGA	
12	1/2	-----	65.5 ^(b)	---	---	20.2	13.8	0.4	---	---	0.07	TRISO/BISO Bonded. Crushed at ORNL	
13	3/8	-----	49.9 ^(b)	---	---	28.4	20.7	0.9	---	---	0.09	TRISO/BISO Bonded. Crushed at ORNL	
14	1/4	---	22.1 ^(b)	---	---	46.1	29.4	2.5	---	---	0.01	TRISO/BISO Bonded. Crushed at ORNL	
15	1/8	---	0	---	---	54.1	38.1	7.8	---	---	0.04	TRISO/BISO Bonded. Crushed at ORNL	
24	1/2	61.1 ^(b)	---	---	---	29.0	---	---	9.6	0.3	0.09	TRISO/TRISO Bonded. Crushed at ORNL	
25	3/8	---	59.2 ^(b)	---	13.0	---	---	---	27.2	0.6	0.04	TRISO/TRISO Bonded. Crushed at ORNL	
26	1/4	---	31.7 ^(b)	---	31.0	---	---	---	35.5	1.8	0.11	TRISO/TRISO Bonded. Crushed at ORNL	
31	5/16	---	24.4 ^(b)	---	47.4	---	---	---	27.7	0.4	0.14	TRISO/TRISO Bonded. Crushed at GGA	

(a) The word before the oblique refers to fissile and after, to fertile

(b) No fuel particles in this fraction

Table 2

DISTRIBUTION OF CRUSHED PRODUCT WHEN CRUSHING
1-ROD FUEL ELEMENTS IN A 3-in. JAW CRUSHER

Run No.	Jaw Gap (in.)	Wt-% Product					Remarks
		+869 μ m	+375 μ m	+250 μ m	-250 μ m	Loss	
32	5/16	---	78.9 ^(b)	20.9	0.23	0.003	BISO fissile (-350 μ m) blended. Crushed at GGA
33	5/16	70.4 ^(a)	---	29.4	0.23	0.015	BISO fertile blended. Crushed at GGA
34	5/16	73.7 ^(a)	16.2	9.9	0.23	0.13	BISO/BISO blended. Crushed at GGA

13 (a) No fuel particles in this fraction

(b) Contained a few particles

Table 3

DISTRIBUTION OF CRUSHED PRODUCT WHEN CRUSHING 1-ROD AND
12-ROD FUEL ELEMENTS IN A HAMMER MILL
(TRISO/TRISO Bonded. Crushed at ORNL)

Run No.	Size of Fuel Element	Grate Spacing (in.)	Wt-% Product					
			+5/8 in.	+1/2 in.	+1/4 in.	+250 μ m	-250 μ m	Loss
27	1-rod	2-1/4	26.9 ^(a)	---	43.0	27.7	2.2	0.21
28	1-rod	3/4	---	10.6 ^(a)	37.4	47.5	2.1	0.25
29	12-rod	3/4	4.9	6.8	29.2	54.0	4.7	0.43
30	12-rod	2-1/4	20.1 ^(a)	12.8	25.4	39.1	2.4	0.27

(a) No fuel particles in this fraction

L.H.B. 8/4/69

Run No.		Table 4 FRACTION OF FINE BURNABLE CARBON AND BROKEN PARTICLES AFTER JAW CRUSHING AND HAMMER MILLING (-250 μ m)				Table 5 BROKEN PARTICLES AFTER TUMBLING AND AFTER CRUSHING AND TUMBLING		Table 6 DISTRIBUTION OF PARTICLE FREE BURNABLE CARBON AFTER CRUSHING AND TUMBLING			Table 7 WT-% BURNABLE CARBON ACCOMPANYING THE FUEL TO THE BURNER	
		Crusher Type(a)	Crusher Setting (in.)	Wt-% Burnable Carbon	Wt-% Broken Particles(b)	Tumbling Wt-%	Total of Crushing and Tumbling	Wt-% After Crushing	Wt-% After Tumbling	Total Wt-% C and T	As OPyC	As Graphite and Matrix
2	1-rod TRISO/TRISO Bonded	JC	1/2	0.39	0.08	—	—	58.7	22.3	81.0	8.0	11.0
12	1-rod TRISO/BISO Bonded	JC	1/2	0.50	0.09	—	—	78.3	5.6	83.9	9.8	6.3
13	1-rod TRISO/BISO Bonded	JC	3/8	1.00	0.17	—	—	59.7	21.6	81.3	9.8	8.9
14	1-rod TRISO/BISO Bonded	JC	1/4	2.81	0.75	—	—	26.5	48.2	74.7	9.8	15.5
15	1-rod TRISO/BISO Bonded	JC	1/8	8.00	2.24	—	—	0	66.1	66.1	9.8	24.1
24	1-rod TRISO/TRISO Bonded	JC	1/2	0.31	0.05	1.2 ^(c)	1.25	76.3	9.8	86.1	6.8	7.1
25	1-rod TRISO/TRISO Bonded	JC	3/8	0.66	0.19	0.8	0.99	73.8	9.2	83.0	6.8	10.2
26	1-rod TRISO/TRISO Bonded	JC	1/4	2.1	0.45	0	0.45	39.4	34.4	73.8	6.8	19.4
27	1-rod TRISO/TRISO Bonded	HM	2-1/4	2.4	1.22	0.4	1.62	33.5	43.2	76.7	6.8	16.5
28	1-rod TRISO/TRISO Bonded	HM	3/4	5.0	2.48	0.4	2.88	12.7	41.6	54.3	6.8	38.9
29	12-rod TRISO/TRISO Bonded	HM	3/4	5.8	0.94	0	0.94	6.3 ^(d)	30.1	36.4	7.6	56.0
30	12-rod TRISO/TRISO Bonded	HM	2-1/4	2.8	0.52	0.6	1.12	25.0	22.1	47.1	7.6	45.3
31	1 rod TRISO/TRISO Bonded	JC	5/16	0.05	0.06	0.6	0.66	33.1	39.2	72.3	6.8	20.9
32	1-rod BISO/ — Blended	JC	5/16	0.29	0.03	—	0.03	79.0	—	79.0	20.8 ^(e)	0.2
33	1-rod — /BISO Blended	JC	5/16	0.32	0.01	—	0.01	70.2	—	70.2	28.9 ^(e)	0.9
34	1-rod BISO/BISO Blended	JC	5/16	0.30	0.02	—	0.02	73.7	—	73.7	25.6 ^(e)	0.7

(a) JC = Jaw Crushing; HM = Hammer Milling

(b) Corrected for graphite ash

(c) Screened 1/8-in. mesh

(d) Contained 1/2-in. of rod

(e) The particles burned to powder being BISO

The tumbler illustrated in Fig. 4 has been used in the laboratory work and has shown the feasibility of the method.

The rate of attrition of particle agglomerates and graphite is shown in Fig. 5. It is obvious that the rate of attrition of agglomerates is very much greater than that of graphite. Particle breakage during tumbling is small but definite as is seen from the results in Table 5 together with the total for crushing and tumbling.

The total fraction of particle-free graphite obtained by crushing and tumbling is shown in Table 6.

An alternative to tumbling is to crush the agglomerates and graphite in a crusher and feed all the product to the fluidized bed. This has the effect of increasing the heat load on the fluidized bed. A possible candidate is a small jaw crusher.

Burning

This step is to remove burnable carbon to prepare the TRISO/TRISO and TRISO/BISO fuel particles for separation by screening. In the case of BISO/BISO particles, the purpose is to remove burnable carbon from the fertile particles in preparation for leaching. The equipment considered most suitable for this step is a fluidized bed.

Burning has been carried out to date in Inconel trays in an air purged muffle furnace at 750°C. The results, Table 5, show that particle breakage, i.e., rupture of the SiC coat, during crushing and burning is low. The fraction of burnable carbon that can be expected to accompany the fuel to the particle burner is shown in Table 7. The remainder, Table 6, is fed to the particle-free graphite burner.

A 2-in. fluidized bed has been constructed and will be operated in the near future. This apparatus will be used to burn the feed of particles/graphite from the screening step and to determine the throughput, residence time, maximum lump size in the feed, particle breakage, optimum temperature, CO₂/O₂ ratio in the feed gas, alumina particle size range, alumina particle breakdown, and fraction fines in the off-gas. This data will enable a larger fluidized bed unit to be constructed and operated.

Fig. 4. 24-in. tumbler

Fig. 5 Attrition by tumbling

The particle-free graphite stream can be burned in either a shaft kiln⁽¹³⁾ or a fluidized bed. The decision will depend upon a feasibility study of each method. The ash will be returned to the main stream at the leaching stage so that small quantities of uranium, if any, will be recovered.

Alumina/Product Separation

The product from the fluidized bed depends on the feed and is a mixture of SiC coated particles/alumina, fines/alumina, and SiC coated particles/fines/alumina when a TRISO/TRISO, BISO fertile and TRISO/BISO feed is used, respectively. It is possible to screen the mixture but a disadvantage could be screen wear by alumina particles. Alternatives are to use spherical alumina or thoria, or to elutriate the mixture and collect the elutriated fraction. The latter has already been shown to be feasible.⁽⁵⁾

Both a screen and elutriation will be used for trial separations and a flowsheet will be drawn up.

It is possible to leach all the material taken from the fluidized bed as illustrated in the conceptual flowsheet in Ref. 14. In this case, the alumina has to be dried, screened, and returned to the fluidized bed.

Work to date on particle/alumina separation has shown that the irregular shaped alumina particles very easily become wedged in the screen and are difficult to remove. Work on particle/particle separation has shown that recovery of fissile and fertile particles from fuel rods is quite efficient as shown in Table 8. Some experiments were done to measure the efficiency of separation of particles which were sized before mixing and screening. The results are shown in Tables 9, 10, and 11. These results show that the size gap between the largest fissile and smallest fertile should be 25 μm or greater. A further experiment was done to measure the rate of separation of mixed fissile and fertile particles. The results are shown in Table 12.

In order to test the ease of separation of irradiated particles, a portion of each of four rods taken from capsule FR-1 was burned, photographed, screened and photographed. The number of broken particles in each portion was counted and the number of fertile particles was counted after separation from the fissile. The results of this experiment showed that the SiC coat

Table 8
SEPARATION OF FISSILE/FERTILE MATERIAL
BY SCREENING AND CROSSOVER OF
ONE TO THE OTHER
(420 μ m Screen)

Run No.	Recovery Weight (g)		Crossover (%)		Remarks
	Fissile	Fertile	Fissile to Fertile	Fertile to Fissile	
2	16.00	34.78	3.0	---	TRISO/TRISO Bonded
3	17.34	34.31	2.0	---	TRISO/TRISO Bonded
4	17.52	32.81	1.7	0.35	TRISO/TRISO Bonded
6	17.64	34.90	4.1	0	TRISO/TRISO Bonded
Expected	17.03	35.26	---	---	---
12	23.43	18.00	< 0.8 ^(a)	(b)	TRISO/BISO Bonded
13	23.36	18.21	< 0.8 ^(a)	(b)	TRISO/BISO Bonded
14	23.46	17.75	< 0.8 ^(a)	(b)	TRISO/BISO Bonded
15	21.95	17.80	~ 0.8	(b)	TRISO/BISO Bonded
Expected	21.78	17.82	---	---	---
24	31.53	43.26	---	---	TRISO/TRISO Bonded
25	32.71	42.68	---	---	TRISO/TRISO Bonded
27	30.34	46.47	---	---	TRISO/TRISO Bonded
28	33.04	43.72	---	---	TRISO/TRISO Bonded
31	31.32	43.83	---	---	TRISO/TRISO Bonded
Expected	32.5 $\pm$ 1.0	42.1 $\pm$ 1.2	---	---	---
34	35.48	56.67	---	---	BISO/BISO Blended
Expected	35.44	57.97 ^(c)	---	---	---

- (a) Limit of detection of uranium in thorium (in the particular method of analysis used).
 (b) All the fertile material burned to powder and was separated from the fissile particles.
 (c) Contained graphite because the mixture was screened before burning.

Table 9
 PARTICLE SEPARATION EFFICIENCY USING A 589 μ m SCREEN
 Zero Initial Size Gap Between the Two Batches

	Weight (g)		Cross-over of Large to Small (%)
	+589 μ m Particles	-589 μ m Particles	
Before screening	3.576	5.570	2.4
After screening	3.440	5.706	
Before screening	13.579	14.500	4.4
After screening	12.893	15.186	

Table 10
 PARTICLE SEPARATION EFFICIENCY USING A 470 μm SCREEN
 25 μm Initial Size Gap Between the Particles

Weight After Screening (g)		Cross-over of Large to Small (%)
+470 μm Particles	-470 μm Particles	
Initial Batch Size: +495 μm , 17.116 g -470 μm , 11.119 g		
17.145	11.089	-0.27
17.118	11.116	-0.03
Initial Batch Size: +495 μm , 16.881 g -470 μm , 11.119 g		
16.904	11.096	-0.21
16.918	11.086	-0.30
16.907	11.093	-0.24
16.908	11.092	-0.24
Initial Batch Size: +495 μm , 16.707 g -470 μm , 11.092 g		
16.722	11.078	-0.13
16.712	11.088	-0.05
16.721	11.077	-0.13
16.721	11.076	-0.15
16.718	11.072	-0.18

Table 11

PARTICLE SEPARATION EFFICIENCY USING A 470 μm SCREEN119 μm Initial Size Gap Between the ParticlesInitial Batch Size: +589 μm ; 10.783 g-470 μm ; 5.123 g

Weight After Screening (g)		Cross-over of Large to Small (%)
+470 μm Particles	-470 μm Particles	
10.792	5.113	-0.20
10.788	5.108	-0.29
10.785	5.123	0
10.789	5.119	-0.08
10.779	5.130	+0.13
10.790	5.120	-0.06

Table 12
SEPARATION OF MIXED PARTICLES
BY SCREENING

Weight of -400 μm : 90.91 g
 Weight of +450 μm : 125.25 g
 Screen Aperture: 420 μm
 Screen Area: 50 sq. in.

Time (min.)	Wt-% of -420 μm Particles		
	Without Tapping, 1.	Without Tapping, 2.	With Tapping
0	0	0	0
1/2	99.25	99.31	99.48
1	99.48	99.47	99.56
2	99.56	99.59	99.64
3	99.60	99.64	99.69
5	99.67	99.72	99.85

Scale-up

Weight of fissile particles per hour: 2.62 kg
 Weight of fertile particles per hour: 38.78 kg
 Rate depending on fertile particles: $0.125 \times 30 = 3.75 \text{ kg/hr}$
 $\therefore$ Area is $38.78 / 3.75 \times 50 \text{ in.}^2 = .520 \text{ sq. in. (1 ft. x 4 ft.)}$

did not burst during the burning operation, the particles were not agglomerated thus presenting no problem to the screening step, and the number of broken SiC coats was less than 0.8%.

Grinding

Grinding is used only when SiC coats have to be cracked open prior to oxidation, i.e., in the TRISO/TRISO route or the recovery of U^{235} and U^{236} from TRISO fissile particles.⁽¹⁵⁾ ORNL have designed a smooth double-roll crusher but the Germans⁽¹⁶⁾ say that roll wear is unacceptably bad on this type of machine and prefer a disk grinder coated with diamond powder. The types of machine which guarantee to break the SiC coat in one pass are limited to those with a diminishing gap. Those which depend on chance, e.g., pin mill or micronizer, would require a recycle of material to ensure that the feed to the next stage contains no uncracked particles.

Laboratory experiments to date have been carried out using batches of 20 g of burned SiC-coated particles. The equipment used is a smooth double-roll crusher with the gap set at 275 μ m. Each roll is 3-in. diam. and 1-in. wide. The machine works well but the real test will be the deterioration of the roll surface with prolonged operation.

Leaching

The objective of leaching is to dissolve the uranium. The liquor, however, also dissolves the thorium and most of the fission products and these are separated in the subsequent solvent extraction step.⁽¹⁷⁾ The residue contains SiC hulls and alumina.

The type of equipment required is a tank which will receive the liquor and solids, and means of agitation for batch dissolution. It is planned to construct a stainless-steel leacher to carry out equipment trials.

A small scale glass leacher has been constructed but not yet operated. Results from experiments on this unit will be used to design the stainless-steel leacher referred to above.

Filtration

This step is intended to clarify the leach solution before solvent extraction. Some form of filter cloth will be required and it may be incorporated in the leacher.

When the glass leacher has been operated, some filtration scoping trials will be carried out to define the mesh size, pressure drop, area, and effects of corrosion. No work has been done to date.

DISCUSSION

The laboratory work done so far has shown that the conceptual flow-sheets proposed by GGA and ORNL are feasible. Briefly, crushing breaks an acceptably small fraction of particles and enables a large fraction of burnable carbon to be separated from fuel particles. Tumbling increases the fraction of burnable carbon with a small increase in the broken fuel particle fraction. The three conceptual flow-sheets shown in Figs. 6, 7, and 8 have been derived from results obtained to date. A jaw-crusher appears to give a lower particle breakage and a higher fuel particle free graphite fraction. A more definite conclusion will be drawn when the results from the larger jaw-crusher experiments are known.

Burning enables the fissile and fertile particles to be separated efficiently by screening providing there is a size gap of greater than 25 μm between the fissile and fertile particles. If BISO/BISO blended fuel is fed to the plant, the flowsheet is simpler providing the fuel particles do not agglomerate.

Laboratory work on elutriation, grinding, leaching, and filtering will be continued to collect the data necessary for scale-up.

The future program lies in the direction of equipment testing at full-scale where economically possible or smaller in some unit processes. A full-scale vibrating screen is about to be purchased; a half-scale Bradford breaker will be constructed and operated; a large fluidized bed burner will be designed and built when the 2-in. fluidized bed produces results; an elutriator will be built for fuel particle/alumina or fuel powder/alumina separation; a large leacher and filter will be built using data from the laboratory work. No program of larger scale grinding will be prepared until SiC coated fuel particles are announced as fact.

L.H.B. 7/21/69

28

Fig. 6 Simplified flowsheet of the crushing and separation of blended BISO/BISO using a jaw crusher. Run 34

L.H.B. 7/21/69

Fig. 7 Simplified flowsheet of the crushing and tumbling concept using a jaw crusher. Run 25

L.H.B. 7/21/69

30

Fig. 8 Simplified flowsheet of the crushing and tumbling concept using a hammer mill. Run 30

REFERENCES

1. S. L. Reese, P. Burn, S. E. Donelson, and K. G. Steyer, "Feasibility Study for Reprocessing HTGR Fuels Based on Using the Nuclear Fuel Services, Inc. Reprocessing Plant," USAEC Report GAMD-6360, General Dynamics, General Atomic Division, December 15, 1964.
2. J. J. Shefcik, "Reprocessing HTGR Hex Block Fuels at the Nuclear Fuel Services Plant," USAEC Report GAMD-7657, General Dynamics, General Atomic Division, January 23, 1967.
3. Private communication, Nuclear Fuel Services Inc. to General Dynamics, General Atomic Division, December 15, 1964.
4. D. E. Ferguson, et al., "Chemical Technology Division Annual Progress Report for Period Ending May 31, 1968," Section 3, USAEC Report ORNL-4272, Oak Ridge National Laboratory, September 1968.
5. R. S. Lowrie, "Fuel Cycle Studies Monthly Report," Oak Ridge National Laboratory, April 1969.
6. J. J. Shefcik, "HTGR Fuel Reprocessing. Effects of Including a Silicon Carbide Coating on Fertile Fuel Particles," Gulf General Atomic Report GAMD-8661, May 1968.
7. J. J. Shefcik, "HTGR Fuel Reprocessing," Gulf General Atomic Report GAMD-8759, July 1968.
8. H. E. Stelling and K. G. Steyer, "Design Study and Economic Evaluation of a Head-End Reprocessing Facility Integral with an HTGR Power Plant," Gulf General Atomic Report GAMD-7294, August 1966.
9. "National HTGR Recycle Development Program Plan Subtask 5100: Reprocessing Development," Oak Ridge National Laboratory, May 29, 1969.

10. "HTGR Base Program Quarterly Progress Report for the Period Ending May 31, 1968," Gulf General Atomic Report GA-8662, June 28, 1968.
11. "HTGR Base Program Quarterly Progress Report for the Period Ending May 31, 1967," General Dynamics, General Atomic Division Report GA-7981, September 29, 1967.
12. J. A. Paget, Private Communication.
13. "Development of Reprocessing Procedures for Thorium-Containing Fuel Elements," International Atomic Energy Agency, Panel on Reprocessing of Highly Irradiated Fuels, Vienna, May 1969.
14. J. J. Shefcik, "Central Reprocessing Plant for HTGR Fuels," Gulf General Atomic Report GAMD-8382, October 1967.
15. J. J. Shefcik, "HTGR Fuel Reprocessing - Recovery of Uranium from Silicon-Carbide-Coated Fissile Particles," Gulf General Atomic Report GAMD-8615, March 1968.
16. Dr. Merz, Jülich, Germany. Private Communication at a Meeting Held at Gulf General Atomic on 6 June 1969. The subject was "Reprocessing of Thorium-Graphite Fuels."
17. M. J. Bradley and L. M. Ferris, "Recovery of Uranium and Thorium from Graphite Fuels. 1. Laboratory Development of a Grind-Leach Process," ORNL-2761, Oak Ridge National Laboratory, April 1960.