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ECONOMICS AND TECHNOLOGY OF HTGR FUEL RECYCLE

A. L. Lotts and R. G. Wymer

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A. L. Lotts and R. G. Wymer

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ABSTRACT

The attainment of low power cost and high fuel utilization in high-temperature gas-cooled reactors (HTGR's) using the thorium-^{233}U fuel cycle depend to a great extent upon the ability to economically recycle fuel. Accordingly, work is being conducted by the Oak Ridge National Laboratory and others to establish the technical requirements and economics for HTGR fuel recycle. The development effort, which is based on a fuel element having a pyrolytic carbon and silicon carbide coated ^{233}U-^{235}U-thorium fuel in graphite prismatic blocks, includes the development of processes and equipment for head-end processing, for chemical reprocessing, for sol-gel preparation of microspheres, and for refabrication of fuel elements. The ultimate objective is to furnish engineering data for the design and operation of a large commercial recycle plant. In addition, the recycle economics and the effects on economics of various alternatives in the recycle processes are being studied. This paper describes the status of the recycle effort and presents recent projections of the cost of recycle HTGR fuel as well as the cost penalties associated with delay in recycling fuel in a growing HTGR economy.

At present, the various processes are in the stage of laboratory development and conceptual design of pilot-scale equipment. The status of all steps in the recycle processes is discussed and the pilot facility, the Thorium-Uranium Recycle Facility (TURF), is briefly described. TURF has been constructed and is ready for installation of equipment for HTGR recycle work. The reference fuel design and the status of the work under way, including irradiation performance tests of coated fuel particles, are also discussed.

INTRODUCTION

High-temperature gas-cooled reactors using the thorium-^{233}U fuel cycle are under development in the United States by Gulf General Atomic because of their potential to achieve low power costs and high fuel utilization. In a 1963 paper, Pahler [1] stated that the attainment of low power costs depends to a great extent upon the development of technology for the economical recycle of fuel from HTGR's, particularly coated particle fuels. This situation has not changed significantly during the last five years in the United States. Indeed, a complete fuel cycle for HTGR's remains to be demonstrated.

Previously the ORNL staff reported on HTGR chemical processing development [2,3] and on their preliminary steps for development of a refabrication technology for HTGR fuel. [4–6] Gulf General Atomic has also described a concept for a head-end reprocessing facility. [7] Since that time ORNL and others in the United States have been conducting work to establish the technical requirements and economics for HTGR fuel recycle. This development has included head-end processing, chemical processing, sol-gel preparation of microspheres, and refabrication of fuel elements. This work has placed us in a better position to assess the economic value of HTGR fuel recycle and, correspondingly, the cost of delaying recycle or not doing it. Two questions arise: (1) what is the status of the technical development for HTGR recycle and (2) what is the economic penalty for either delaying or even not carrying out the thorium-^{233}U recycle. We shall deal with these two questions in this paper.

First, we shall present the basis for a thorium-based HTGR fuel recycle program; second, a statement describing how the work is organized and scheduled; third, the current status of recycle technical development; and fourth, the economic projections and penalties associated with delay in recycle in a growing HTGR economy.

PROGRAM BASIS

The objective of our work is to furnish the necessary technology to enable a central reprocessing and refabrication plant to be constructed and placed into operation. To meet this goal, it was necessary to select reference fuel and recycle processes for development and pilot plant demonstration. The development references and the required developments are as follows.

Reference Fuel Element Design for Recycle Development

The reference recycle fuel element is very similar to that specified by Gulf General Atomic for the Public Service of Colorado (PSC) Fort St. Vrain Reactor and for the near-term 1000-Mw (electrical) HTGR's that are planned by Gulf General Atomic. The principal difference is in the details of the coated particles. The Fort St. Vrain Reactor will contain 1260 standard fuel elements and 222 fuel elements with control rod holes. The fuel elements are hexagonal blocks of graphite approximately 30 in. long × 14 in. across the flats, as shown in Fig. 1. Each element contains 102 coolant holes and 210 fuel holes. The fuel holes are approximately 0.45 in. in diameter. The recycle fuel for these reactors will consist of three different types of particles: (1) one containing ^{233}U and thorium, (2) one containing makeup ^{235}U, and (3) one containing thorium. The particles, which are coated with layers of SiC and pyrolytic carbon, are sized to facilitate selective separation by screening to simplify reprocessing. The particles are mixed and molded into a bonded fuel stick and inserted into carbon fuel blocks during fabrication.

Although the reference recycle fuel will consist of the particles previously indicated, the recycle process should be capable of handling a broader range of particles and fuel body types. Table I describes eight particles and groups them to satisfy four different situations: a reference fuel for both initial and recycle core loadings and a long-term fuel for both initial and recycle core loadings. We assume that head-end processing steps may have to accommodate hot-pressed fuel compacts as well as the bonded fuel particles of future HTGR's. Such hot-pressed fuel compacts are used in the only operating HTGR in the United States, the Peach Bottom Reactor located near Delta, Pennsylvania. The reference fuel listed in Table I is amenable to recycle by current technology. The advanced fuel appears most economically desirable for future HTGR fuel systems. A discussion of the various particles listed in Table I follows.

Reference Fuel Particles. — The reference fuel particles are of four types. Carbides were selected for the initial core loadings because Gulf General Atomic now has the technology necessary to fabricate these particles in large quantities.

To produce an instantaneous Doppler coefficient in the initial core, some or all of the ^{235}U will be formed into thorium-uranium dicarbide particles (type 2). In the Fort St. Vrain Reactor all of the ^{235}U is mixed with the thorium, but in the current 1000 Mw (electrical) HTGR design only half of the initial fuel elements contain thorium-uranium particles, and the remaining half use uranium dicarbide particles (type 3). Since two of the four initial segments in the 1000 Mw(electrical) reactor achieve less than four years of burnup, the ^{235}U depletion is less than normal. These segments contain the thorium dicarbide and the thorium-uranium dicarbide particles. To reduce the complexity of recycle, no particle separation will be done on these segments; thus, all uranium isotopes will be recycled together. The remaining two core

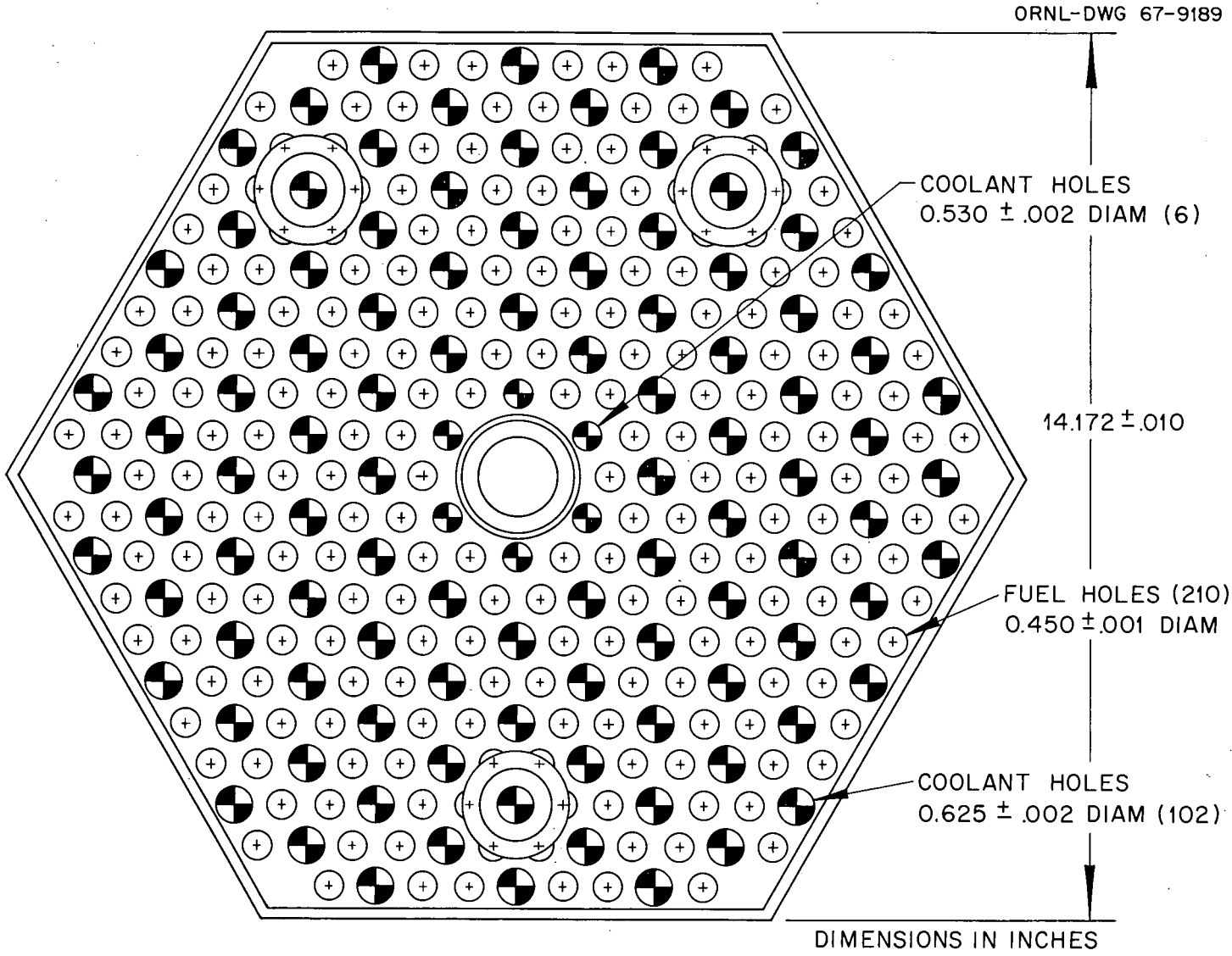

Fig. 1. Fort St. Vrain Reactor Fuel Element.

Table I. HTGR Fuel Particle Descriptions[a]

	Fresh Fuel			Recycle Fuel[b]
	Fertile Particle	Fissile Particle 1	Fissile Particle 2	Fissile Particle 1
	Reference Fuel			
Particle type	1	2	3	4
Kernel composition	ThC_2	$(Th,^{235}U)C_2$	$(^{235}U)C_2$	$(Th,^{233}U)O_2$
Kernel diameter, μ	400 ± 100	200 ± 50	100 ± 10	400 ± 100
Buffer carbon thickness, μ	50 ± 10	50 ± 10	100 ± 10	50 ± 10
Inner isotropic pyrolytic carbon thickness, μ	20 ± 5	20 ± 5	20 ± 5	20 ± 5
Silicon carbide thickness, μ	20 ± 3	20 ± 3	20 ± 3	20 ± 3
Outer pyrolytic carbon thickness, μ	40 ± 5	30 ± 5	30 ± 5	40 ± 5
Total particle diameter, μ	660 ± 146	440 ± 96	440 ± 56	660 ± 146
	Advanced Fuel			
Particle type	5	6	7	8
Kernel composition	ThO_2	$(Th,^{235}U)O_2$	$(^{235}U)O_2$	$(^{233}U)O_2$
Kernel diameter, μ	400 ± 100	200 ± 50	100 ± 10	200 ± 50
Buffer carbon thickness, μ	50 ± 10	50 ± 10	100 ± 10	150 ± 10
Outer pyrolytic carbon thickness, μ	80 ± 10	70 ± 10	70 ± 10	80 ± 10
Total particle diameter, μ	660 ± 140	440 ± 90	440 ± 50	660 ± 90

[a]Particles will be bonded into fuel sticks for insertion into hexagonal graphite fuel elements. Natural boron carbide sticks containing about 15% B_4C in a graphite matrix will be used in first-core fuel elements as a burnable poison.

[b]Fertile and fissile 2 particles are the same as for fresh fuel.

segments contain thorium dicarbide and uranium dicarbide particles. For these fuel elements particles are separated by the size difference between particles 1 and 3. This is done to avoid recycle of ^{236}U and ^{237}U with the ^{233}U. Particle 3 is diluted with carbon or voids to accommodate swelling caused by its very high burnup. The dilution is assumed to be accomplished by increasing the thickness of the buffer carbon layer adjacent to the kernel.

Recovery of ^{235}U from particle 3 may be economically warranted since the discharged uranium is about 30% enriched and contains a significant quantity of ^{237}Np and ^{238}Pu.

Particle 4 is representative of the fuel for early recycle cores. This particle contains ^{233}U and thorium in the form of oxide prepared by the sol-gel process. The spent fuel particles are assumed to be separable by size difference. Although thorium is blended with ^{233}U in this particle, development of the sol-gel process for making pure UO_2 particles makes it possible to use ^{233}U particles if desired.

Advanced Particles. — There are also four types of advanced particles. The particles used in the initial reactor charges (particles 5, 6, and 7) are designed on the same basis as particles 1, 2, and 3. The significant difference is the elimination of the SiC coating and the use of oxides rather than carbides. This change anticipates demonstration of the adequacy of two isotropic BISO coatings for satisfactory fission-product retention and the development of sol-gel methods for fuel fabrication to the point where future large-scale plants employing sol-gel methods would have a competitive advantage over present methods of fabricating non-recycle fuels.

The advanced recycle ^{233}U particle (particle 8) contains no thorium. With thorium omitted, the volume of the ^{233}U refabrication process streams can be reduced 80% to save costs. The reduction arises because particles 2 and 4 contain four times as much thorium as uranium.

The reprocessing plant must be able to handle all the particle types. Only the ^{233}U-bearing particles (4 and 8) must be prepared in the recycle facilities; however, the fuel rod and assembly processes must be capable of producing fuel elements using combinations of recycle fuel particles as indicated in Table I.

Generalized Fuel Cycle Flowsheets

The reference and advanced recycle process flowsheets, which are based on the particles shown in Table I, are given in Fig. 2. The main differences in the flowsheets are as follows. In the reference flowsheet the fuel is removed from the spent fuel blocks by crushing and burning the entire block, whereas in the advanced flowsheet the fuel particles are first removed from the blocks. Also, in the advanced flowsheet the fuel particles are not crushed because silicon carbide

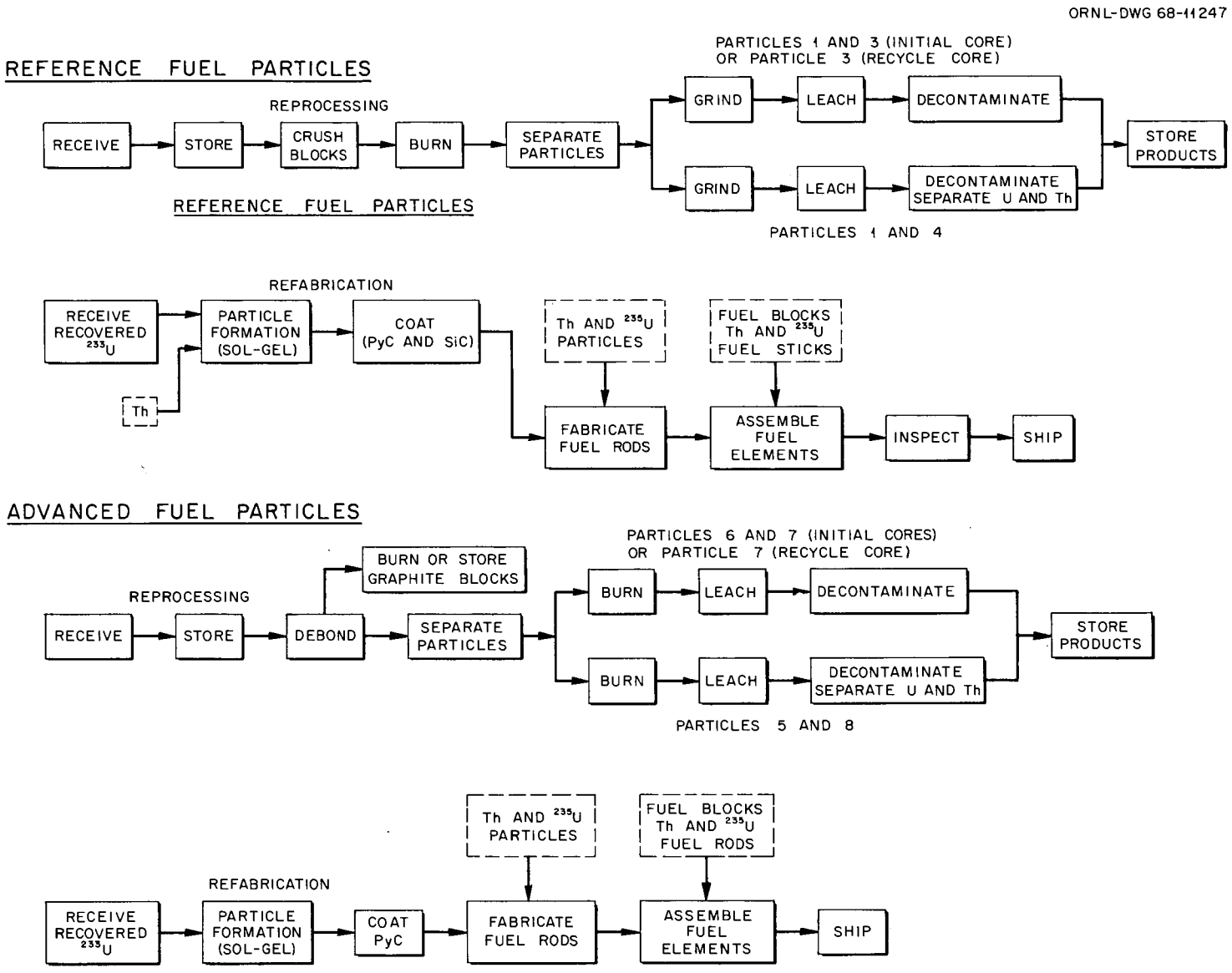

Fig. 2. Recycle Process Flowsheets.

coatings are not present. The key problem areas of these flowsheets, and therefore the areas that need the most technical effort, are as follows: (1) a method must be developed for removal of particles from the fuel blocks and separation of the removed particles; (2) systems must be developed for treating and disposing of fission-product gases; and (3) systems must be developed for the remote operation and maintenance of equipment for microsphere formation, particle coating, mixing the particles before bonding into fuel sticks, and bonding the particles into fuel sticks. The approach at ORNL is primarily based on the burn-leach method for head-end processing, aqueous methods for chemical processing of fuel, sol-gel methods for preparation of recycle fuel particles, and conventional coating and bonding methods for the refabrication of fuel.

Schedule for Recycle Development

Economic studies, which will be presented later in this paper, indicate that the most desirable time to begin large-scale HTGR fuel recycle is during the 1980–1982 period. For the purpose of presenting a schedule in this paper, we shall select 1981. Before large-scale plants can be built and placed into operation to process and fabricate ^{233}U fuels by 1981, a comprehensive program for development of HTGR fuel recycle technology must be completed. Our schedule for HTGR recycle development is presented in Fig. 3. The schedule has been divided into two broad areas: (1) fuel reprocessing development, which includes head-end and solvent extraction processing, and (2) fuel refabrication development, which includes both preparation by the sol-gel process and manufacture of fuel elements by pyrolytic carbon coating, bonding of fuel sticks, and assembly of the sticks into graphite blocks. Development in the two areas culminates in the design and construction of a central plant, the construction schedule for which is included in Fig. 3. Approximately 4.5 years are needed to complete the process development. The last year of that period will be used to complete and prove the reliability of equipment before processing of radioactive material begins. About 1.5 years of hot operation will be required to obtain adequate experience and information to design, construct, and test the large-scale commercial plant. [8] We estimate that 5.5 years are required to go from conceptual design to hot operation of the commercial recycle plant.

Probably a smaller and diminishing development effort will be required during the latter period to solve problems related to scaleup and to further refine the processes for recycle.

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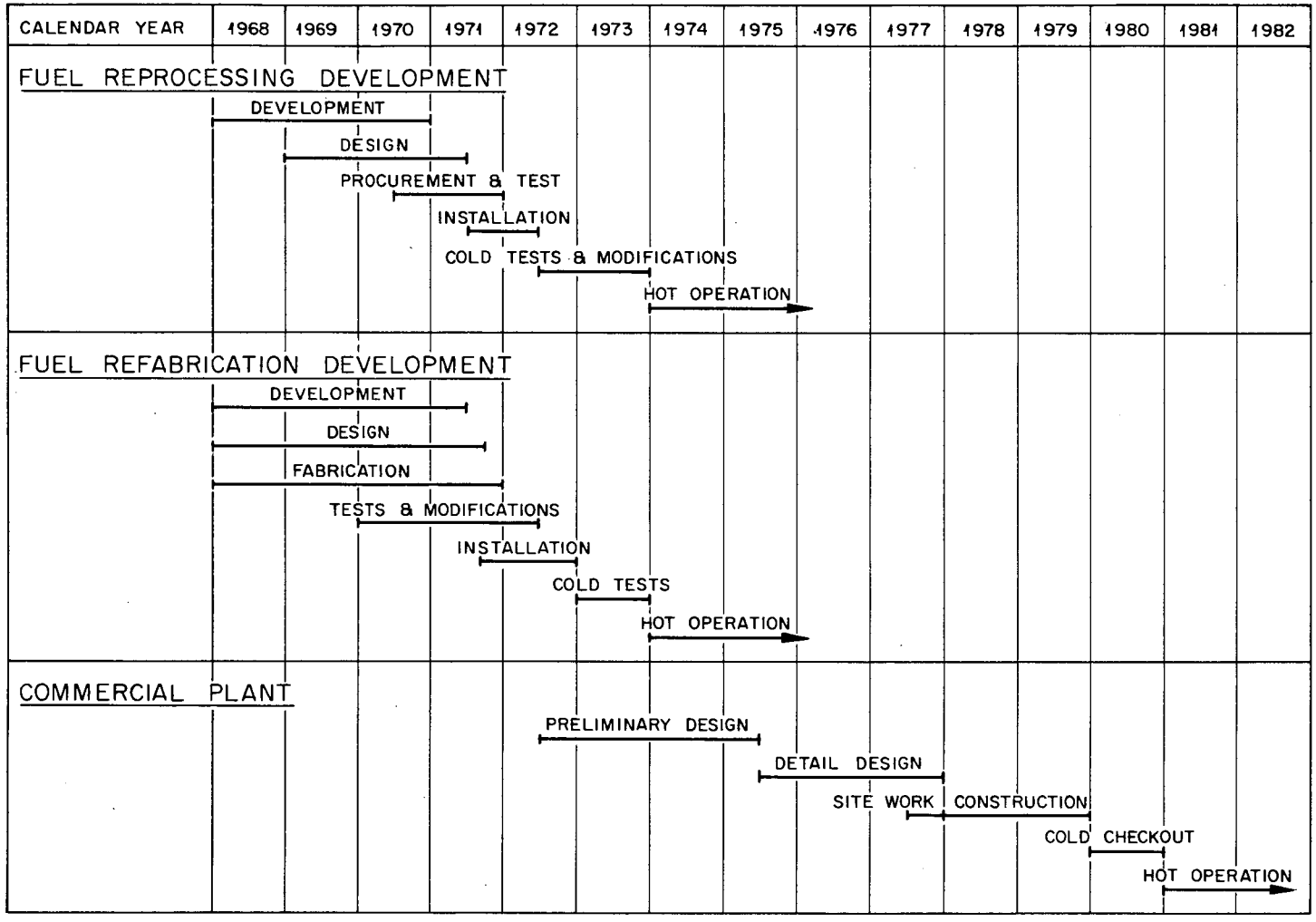

Fig. 3. Development Leading to Operating Commercial Recycle Plant.

STATUS OF RECYCLE DEVELOPMENT

Papers by Nicholson [2] and Lotts [4] and their co-workers in 1965 summarized the status of HTGR recycle development in the United States. Before proceeding with a discussion of the status of recycle development in the various processing technologies, it seems useful to briefly summarize what has been done on reprocessing and refabrication of ^{233}U-thorium fuels. Only a limited amount of technology is presently available for the recycle of ^{233}U-containing graphite matrix fuel elements. Commercial facilities such as the plant of the Nuclear Fuels Services, Inc., a subsidiary of W. R. Grace Company, and Midwest Fuel Recovery Plant, currently proposed by General Electric, as well as other proposed chemical processing facilities, are primarily concerned with the extraction of ^{235}U and plutonium from water reactor fuels. Also the government-owned plants at Hanford and Savannah River are not concerned with reprocessing of ^{233}U graphite matrix fuels.

Preliminary head-end treatment work has been done on ^{233}U graphite matrix fuel. [7,9,10,11] Reprocessing of irradiated thorium metal was demonstrated in the ORNL Thorex Pilot Plant [12] in 1956, and ^{233}U has been recovered from irradiated thoria. [13] The vibratory compaction of $(ThO_2-3\%\ U)O_2$ shards into Zircaloy-clad fuel rods was demonstrated in the Kilorod Plant [14] in 1964.

There is essentially no experience with power reactor graphite matrix fuels containing thorium and ^{233}U. Little has been done on the separation of fuel particles containing ^{233}U from those containing ^{235}U as is required by the reference recycle process flowsheet previously presented.

Laboratory studies have been reported on head-end processing, chemical processing by solvent extraction, and particle fabrication at ORNL. [15–17] The various steps in chemical reprocessing are well delineated. Cold fabrication of the reference fuel has been performed on a developmental engineering scale at ORNL and on a production scale at Gulf General Atomic. This fabrication development includes making microspheres by sol-gel forming techniques, [18] coating with pyrolytic carbon and silicon carbide, [19] and combining the microspheres into fuel sticks. [20] The sol-gel process for preparing ThO_2-UO_2 and UO_2 microspheres has been demonstrated in directly maintained engineering equipment on a greater than 10 kg/day scale. [21] Substantial progress has been made in developing methods for performing the same operations remotely. This work, along with related coating studies, is being done in the Coated Particle Development Laboratory, a laboratory designed for this task. [18] Also, some equipment for performing these operations in a remote facility has been conceptually designed. [18]

Although the above discussion gives general information on the problems in recycling thorium-^{233}U and implies specific data and technology on certain steps associated with the recycling of HTGR fuels, it also shows that much remains to be done before accomplishing the fully

integrated recycle demonstration required for the design of a commercial recycle plant. To assess in detail the present status of the various technological areas and to determine more precisely what remains to be done, the following comments give the present status of recycle development for the various areas: reprocessing, including head-end processing and solvent extraction, and fuel refabrication, including preparation of microspheres and fabrication of fuel elements.

Fuel Reprocessing

Reprocessing development tasks include the development of head-end processes and the application of existing technology for aqueous processing.

Head-End Process. — The head-end reprocessing development program involves cold engineering-scale work on graphite fuel block treatment and on fuel particle separation, as well as hot-cell experiments on irradiated fuel capsules. The objective of the engineering work is to find means for separating coated fuel particles from the bulk of the graphite fuel matrix and separating ^{233}U-containing particles from ^{235}U-containing particles. A flowsheet for head-end processing of the reference fuel element is shown in Fig. 4. The hot-cell experiments are to provide information on fuel stick and coated particle properties and behavior when they have been irradiated to anticipated HTGR conditions and on the effect such behavior will have on fuel particle recovery, separability, and dissolution in acid. Information will also be obtained on radioactive gas behavior and treatment.

The very large amount of graphite in the block relative to fuel particle mass in the fuel element makes mechanical separation of the matrix from the fuel particles highly desirable. If the matrix material cannot be separated from the fuel particles, the entire fuel block must be burned. This requires breaking the block into pieces which will burn in equipment of reasonable size. It also leads to large volumes of gas containing oxides of carbon, oxygen, perhaps a chemically inert diluent gas, and small but significant amounts of the radioactive gases tritium and ^{85}Kr. The radioactive gases must be removed from the waste gas stream before its permanent disposal. Because of the anticipated problems of burner operation, gas handling and disposal, methods are being studied for the removal of the fuel stick. These include: erosion of the stick bonding material away from the fuel particles by a water jet directed into the fuel holes in the graphite block; drilling or "brushing" the fuel particles and stick bonding material out of the fuel holes; cutting around the fuel holes so as to free the fuel stick in a thin-walled graphite tube; and finally, burning away the stick bonding material preferentially from the fuel particles by directing a flame lance into the fuel holes. This latter approach is perhaps the most promising. Its success depends on the fact that the graphite block matrix, the stick bonding material, and the pyrolytic carbon coatings on the particles all burn at a different rate; the stick bonding material is the easiest to burn.

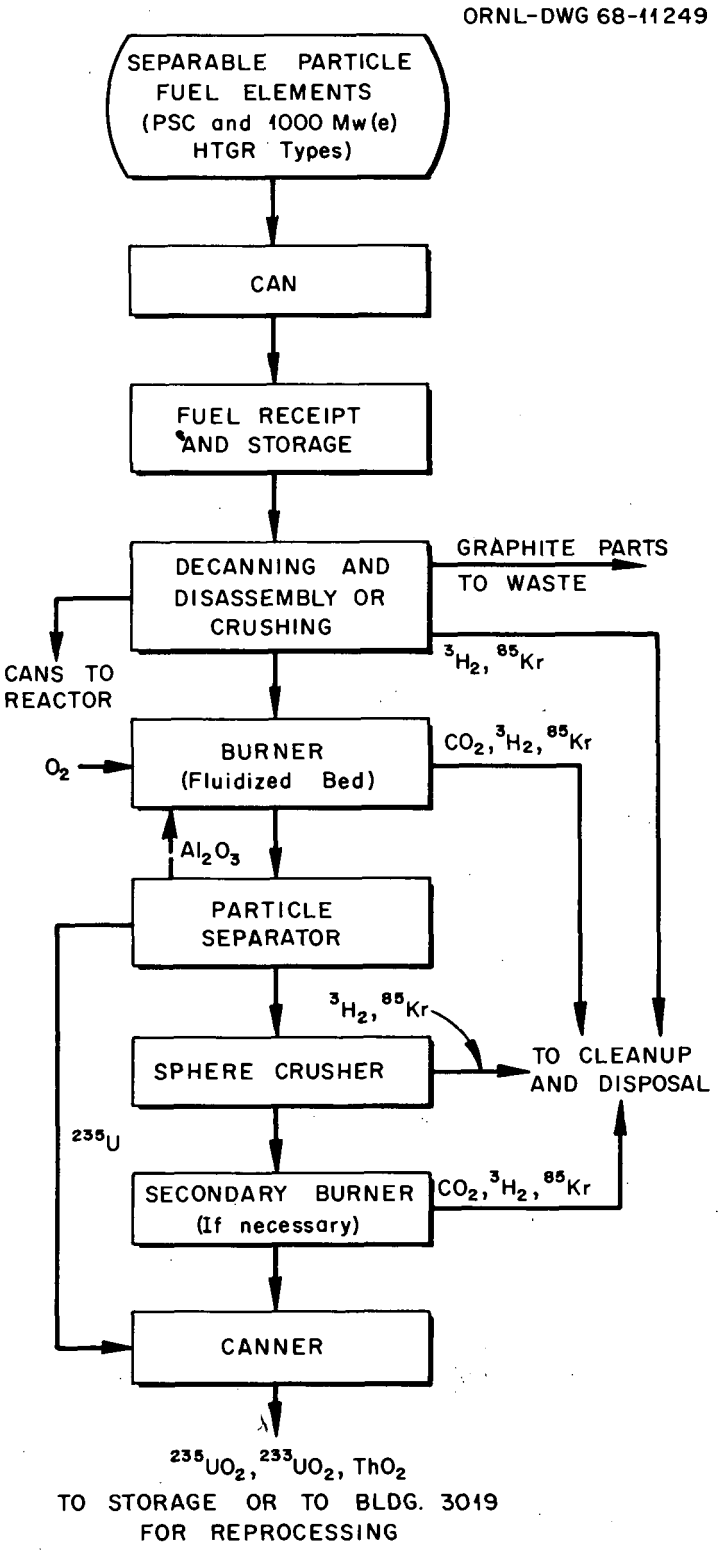

Fig. 4. HTGR Head-End Reprocessing Schematic Flowsheet.

Notwithstanding the desirability of mechanical stick removal, the only approach to head-end reprocessing that seems certain to work is block burning. This approach relies on the SiC coatings on the three different particles retaining their integrity during block crushing and burning. Important answers to be found in the engineering tests are how to crush the graphite blocks; what fraction of the particles will break during block crushing and the resultant extent of cross contamination between the two uranium isotopes; optimum burner size, configuration and operating conditions; composition, nature, and best treatment of burner off-gas; the degree of particle breakage during burning and the extent of uranium isotope cross contamination from SiC coating failure; how to separate components in the burner product, which will consist of two different sizes of SiC-coated particles, and probably of alumina as well, since a fluidized-bed burner using alumina as the fluidized material is likely to be the best type of burner; and finally, how to treat the separated fuel particles to prepare them as suitable feed for solvent extraction. This includes how best to break the SiC coating to permit dissolution of the fuel particle kernels and whether the inner buffer coating needs to be burned off.

At present, full-scale prototype HTGR fuel blocks have been fabricated, and some methods of block crushing and mechanical removal processes for freeing fuel particles from the graphite matrix have been tested. [22] Small-scale burner studies have also been done. [23]

Irradiated loose particles and bonded particles will be tested in a hot cell. Tests with the irradiated loose particles will provide information on SiC coatings and on fission-gas release from the fuel particles during various stages of head-end processing. The tests include the burnoff of the outer pyrolytic carbon coatings, the breaking of the SiC coating, probably by grinding, subsequent burning (if necessary) of the buffer coat, and dissolution of the fuel kernels in acid. The points at which fission product gases are released and their amounts will have an important bearing on design of the off-gas facilities, the burner, the SiC coating grinder, and the kernel dissolving equipment.

Tests with the irradiated fuel sticks will provide information on effects of irradiation on the bonding matrix material and thus will help determine if promising mechanical means of matrix removal are likely to be helped or hindered by irradiation. Also, crushing and burning of the irradiated fuel sticks contained in graphite sleeves will help determine if cold engineering tests are valid, or if irradiation-induced changes necessitate modified or new approaches to crushing and burning.

Solvent Extraction. — Thorium- and uranium-bearing material from the burner will be dissolved to form a solution corresponding to Thorex Process [12] requirements. The Thorex Process or a very similar solvent extraction process will be used to separate ^{233}U from thorium and from fission products. This process is based on solvent extraction of metal ions from nitrate media with a solution of tributyl phosphate (TBP) in a high-boiling organic diluent, such as dodecane. The chemical flowsheet

which will very likely be emphasized is shown in Fig. 5. The uranium from the ^{235}U-containing particles may also be recycled to the reactor because of economic considerations. However, because it need not be refabricated remotely, it is not being considered in this part of the recycle development.

Of considerable importance in developing the solvent extraction purification of HTGR fuels is the presence of ^{233}U in concentrations exceeding the critically safe maximum. Soluble and fixed poisons in solutions and tanks will be used along with geometry and mass control to ensure that no possibility exists for a critical accident.

The Thorex Process has been tested on a pilot plant scale. A total of 40 metric tons of irradiated thorium metal and oxide were processed. In part of the pilot plant demonstration, thorium that had been irradiated to produce 4 kg ^{233}U per metric ton of thorium and cooled only 28 days was processed. Recently, both the Hanford and Savannah River sites employed the technology derived from the Thorex Process pilot plant demonstration to process many tons of ThO_2 that had been lightly irradiated to produce ^{233}U.

After the Thorex Process pilot-scale demonstration, the equipment and facilities have been used to make ORNL the national ^{233}U depository. Thus facilities are available for storing 100 kg of this isotope. Much of the solvent extraction plant necessary to carry out the HTGR fuel reprocessing part of the recycle demonstration is also available at ORNL in the Pilot Plant Building; however, some additional off-gas treatment facilities, as well as other equipment needed specifically for the recycle demonstration, may have to be added to the plant. An equipment flowsheet is shown in Fig. 6. Shaded equipment pieces must be added to the plant. Not shown is the off-gas treatment facility that will probably be required.

Fuel Refabrication

The refabrication developmental effort includes the preparation of fuel material, the coating of fuel particles, and the fabrication of fuel sticks and fuel elements. We are emphasizing development of refabrication capabilities for fuel for the PSC Fort St. Vrain and the proposed 1000 Mw reactors. The initial cores of these reactors will consist of thorium-uranium dicarbide particles coated with pyrolytic carbon and SiC, and of thorium dicarbide and uranium dicarbide particles coated similarly. Recycle cores will have particles consisting of ThC_2, $^{233}UC_2$, and $(^{233}U,Th)O_2$ coated with SiC and pyrolytic carbon. Only the sol-gel processes will be considered initially for production of the mixed oxide fuel kernels.

Sol-Gel Process. — Sol formation is the first operation in forming recycle fuel particles after spent fuel reprocessing. The solution received from the Thorex pilot plant is transformed to sol by a continuous remotely operable solvent extraction process. This process is shown schematically in Fig. 7.

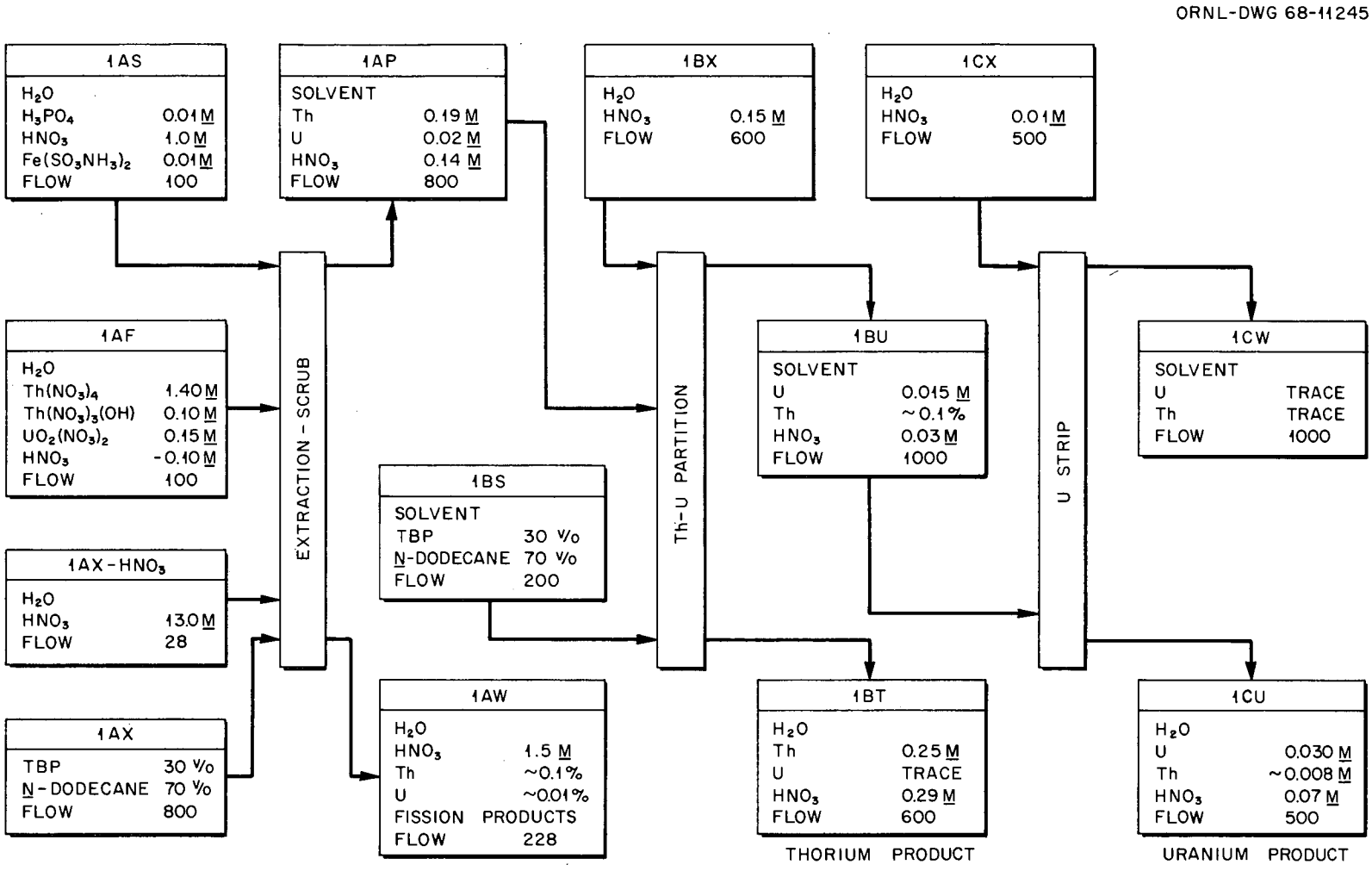

Fig. 5. Solvent Extraction Chemical Flowsheet for HTGR Fuel Purification.

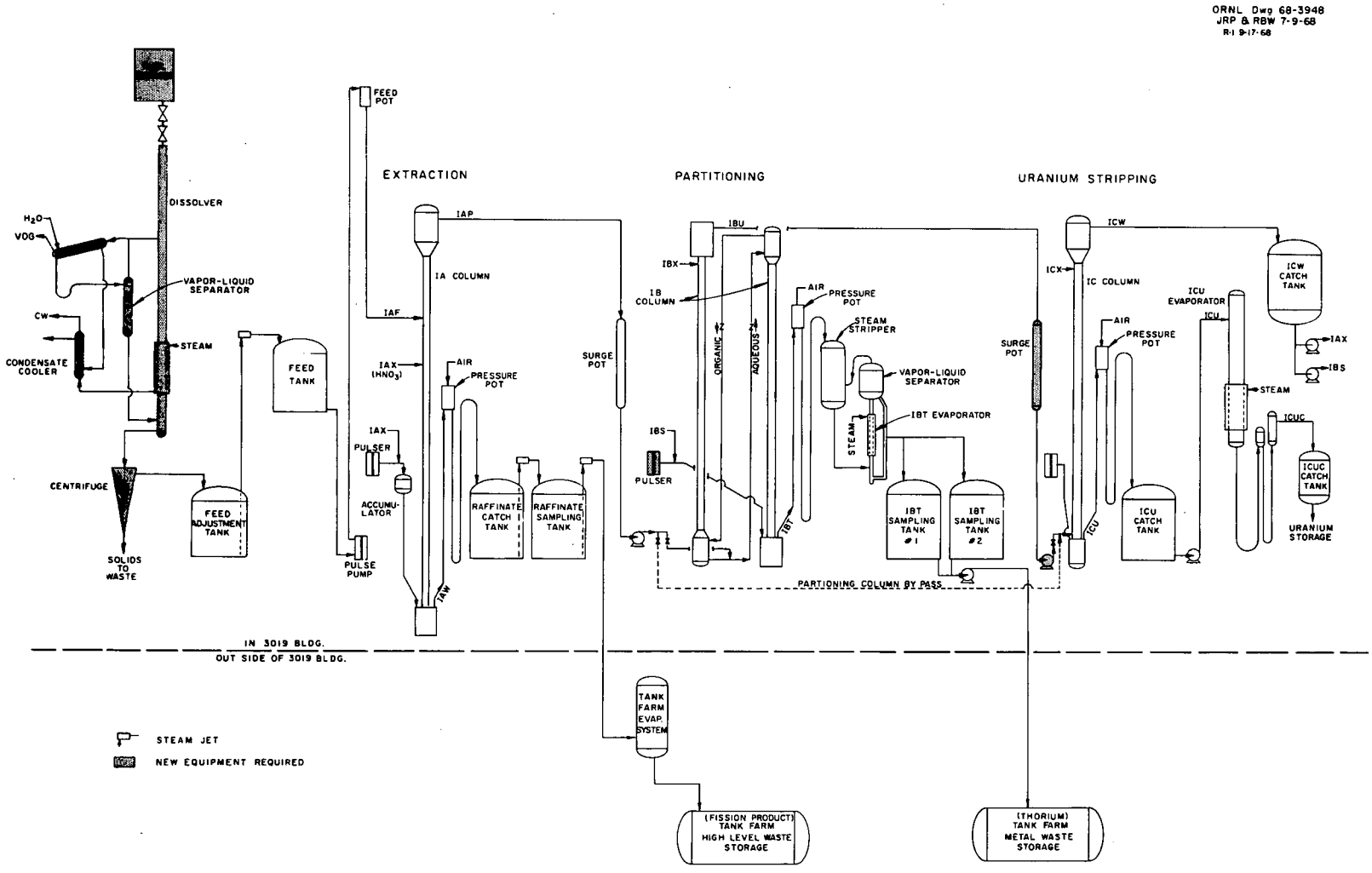

Fig. 6. Proposed HTGR Equipment Flowsheet for Building 3019.

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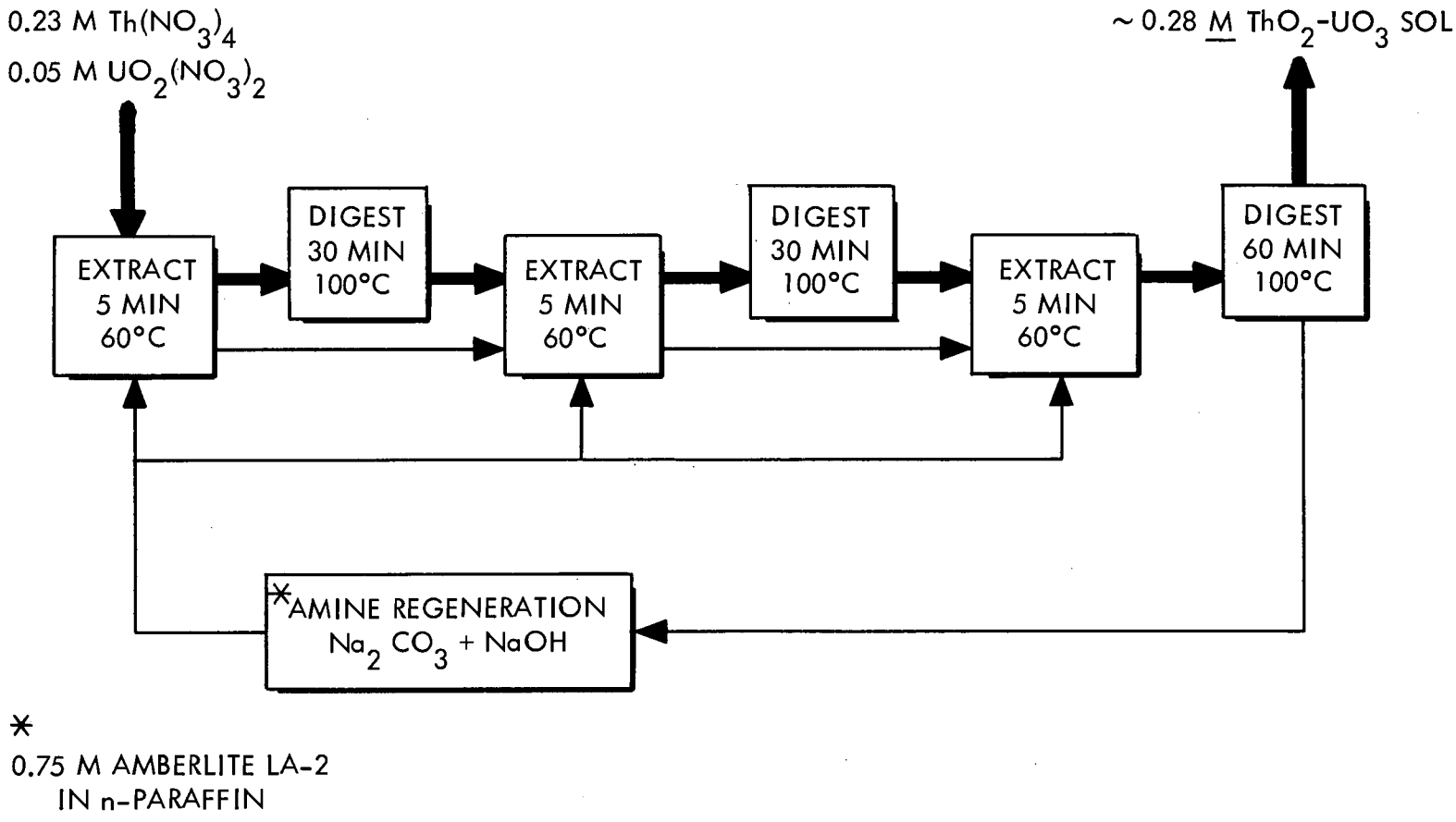

* 0.75 M AMBERLITE LA-2 IN n-PARAFFIN

Fig. 7. ThO_2-UO_3 Sol Preparation by Solvent Extraction.

Sol is formed into gel microspheres of controlled size in a remotely operable sphere-forming column. The gel microspheres are then dried and fired to form the final oxide particles. The only particle that will be formed in this way is the ThO_2-$^{233}UO_2$ particle. The presence of several hundred parts per million ^{232}U in the recycle ^{233}U HTGR fuel dictates that it not be handled directly. The other particles do not require remote fabrication.

Engineering-scale equipment for forming sols of mixed thorium and uranium is in use as a hooded operation. Further operating experience is needed before the remotely operable equipment can be designed. All ThO_2-UO_2 ratios of interest can be made from solutions of thorium nitrate and uranyl nitrate. Such solutions are the normal product of conventional solvent extraction processes for uranium and thorium purification, for example, the Thorex Process.

Gel microsphere-forming column operations are in advanced stages of development, although much of the work is empirical and the underlying principles are imperfectly understood. Gel sphere drying is also still imperfectly understood. More work is needed on these steps. Work also remains to be done on developing remote handling and inspection techniques for the sphere-forming operations. The sphere-forming equipment is more flexible than other types of equipment used in refabrication because of the inherent ease of changing from one microsphere size to another and because the sphere handling and inspection equipment can be designed to deal with a range of sizes and a variety of materials (e.g., ThO_2-UO_2 and UO_2). The requirement for carrying out the operations remotely imposes problems and constraints that must be studied further through cold engineering development work in prototype equipment.

Fabrication of Fuel Elements. — The flowsheet for refabrication of the reference fuel elements is shown in Fig. 8. This figure shows primary steps for inspection of bare particles containing ^{233}U, coating of these particles, and inspection of the coated particles. The ^{233}U-bearing particles are mixed with separate particles bearing the thorium and the ^{235}U and molded into fuel sticks. The fuel sticks are polymerized, loaded into graphite blocks, and carbonized *in situ*. Operations are also shown for inspection of fuel sticks and fuel element loading and inspection.

Particle Coating. — A 5-in.-diam, spouting-bed furnace similar to the coating furnace planned for refabrication has been extensively used for pyrolytic carbon coating. [19] A 3-in.-diam, spouting-bed furnace has been used for silicon carbide coating. [24] Coatings of the required types have been produced on semi-production scale.

The buffer coating is applied from a mixture of acetylene and inert gas. The isotropic coating is presently applied with propylene but can be applied with either propane or methane. The silicon carbide coating is done with methyltrichlorosilane and hydrogen.

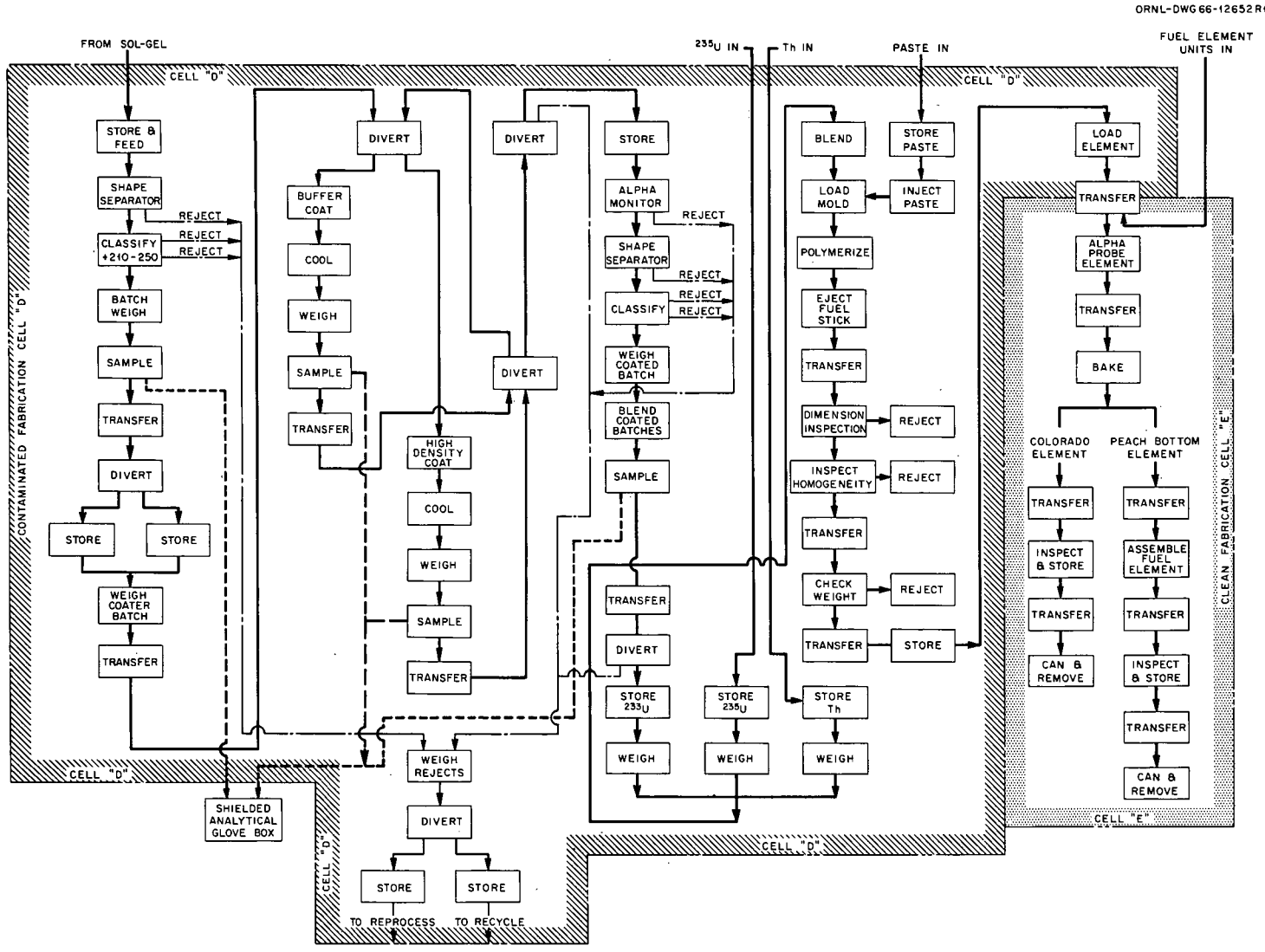

Fig. 8. Process Flow Diagram for Recycle HTGR Fuel in TURF.

All coating operations involve the production of large volumes of off-gas, consisting of hydrogen or hydrogen and HCl, depending on which coating is being applied. In addition, all off-gas must be filtered to remove any radioactive material. Countercurrent caustic solutions remove HCl. During hydrocarbon coating, no HCl will be in the off-gas but large quantities of soot may be present. Hence, the off-gas first passes through a bag filter, where the soot is removed. For either coating process the gas is passed through absolute filters before being ejected into the atmosphere. Special precautions are used to prevent explosive mixtures of hydrogen and oxygen from occurring in the off-gas equipment.

A prototype of the remote coating furnace is currently being constructed. On completion, the effect of operating variables on the coatings produced will be determined, and the remote operation and maintenance capabilities of the prototype will be checked. Next, a system for coating in the refabrication pilot plant will be designed and installed.

Particle Handling and Inspection. — Particles must be remotely transferred between operations in the pilot demonstration. The transfer method must not damage the particles and must require only limited maintenance. The particles, both before and after coating, must be inspected to ensure that specifications are met.

Considerable progress has been made on the development of equipment and processes for particle handling and inspection. After the microsphere production step, the dried and sintered microspheres must be fed into a hopper. This hopper has been designed and tested. The feed mechanism on the hopper is a cut-off valve also designed and tested at ORNL. This valve consists of a flexible tube that can be externally pressurized. When pressurized, the tube buckles inward, thereby shutting off the flow of microspheres through it. Particles are fed from one location to another by gravity feeding and pressurized pneumatic feeding. Tests made at ORNL indicate that pneumatic feeding can be used to transfer particles over distances of more than 50 ft with increases in elevation of at least 15 ft. Negligible abrasion occurs even on the low-density acetylene particle coatings.

Microspheres next must be shape-separated to eliminate any particles with a maximum-to-minimum diameter ratio of greater than 1.3. The shape separator developed at ORNL consists of a flat plate, which is tilted and vibrated to separate the nonspherical particles from the microspheres. The microspheres then must be fed to a size classifier which, as presently conceived, consists of two screens to eliminate the oversize and undersize in one operation. At this point, the acceptable material must be weighed and then dispensed into a sampler. We presently use Vezin samplers, which appear ideal for the pilot demonstration. The sample and the main batch material then must be fed to storage hoppers. The sample at this time must be inspected for size distribution. If necessary particle sphericity and density may also be inspected. The microspheres

to be inspected for size distribution are transferred to the particle size analyzer. In this instrument, a light shines across a transparent rectangular tube through which the microspheres are passed one at a time. The light intensity is observed by a photodetector as a microsphere passes through the channel; the photodetector records a dip in current proportional to the size of the microsphere, which is then recorded by a pulse-height analyzer. Presently, a sample can be recorded in approximately 3 min. No real effort has been made to speed the operation; hence, considerably faster counting may be possible.

If density must be measured, a mercury pycnometer can be used. This apparatus measures the volume of the particle by measuring the amount of mercury displaced by the particles. Knowing the volume displaced, we can weigh the microspheres and accurately determine density.

The qualified material then must be weighed to the proper batch size for coating. After each coating operation, the coating thickness has to be measured by the particle size analyzer, and the density in conjunction with a weight measurement. In addition, the anisotropy must be occasionally checked on the pyrolytic carbon coatings. The method used is based on the reflection of polarized light. When sectioned specimens of coated microspheres are examined under polarized light, highly anisotropic coatings show a maltese cross, but isotropic coatings show no such effect. The appearance of the maltese cross can be calibrated with x-ray determinations of anisotropy.

Microspheres can be transferred from the furnace by gravity feeding through the bottom of the cone to receiving hoppers. At this point the microspheres must be fed to weighers and samplers, from which they go to the inspection line. The microspheres would next be either returned to the coating furnaces if more coating is required or sent on to the next step if the coating operations are complete.

To facilitate these various transfers, diverter valves have been developed to be used in conjunction with storage hoppers. These diverter valves operate by material entering the top and traveling through a flexible tube, which is then connected to one of two exit tubes by a pneumatic cylinder and cam device. The design and testing of this device has been completed.

After all coating operations are finished, the microspheres must be sent to an alpha monitor for inspection of surface contamination. This alpha monitor consists of a feeding mechanism that produces a monolayer of microspheres traveling across a 6-in.-wide plate, observed by an alpha monitor. The level of contamination provides an indication of the quality of the coating operations. A prototype of the alpha monitor has been completed. After qualifying in this inspection the microspheres must be sent to another shape separator and classifier. Again all products have to be weighed and any abnormal amount of reject material is then inspected. At this point in the operation numerous batches of microspheres are accumulated. Batches are then blended to produce large

homogeneous batches of microspheres needed for the succeeding steps. This batch blender has been conceptually designed. The homogeneous batch must then be sampled for quality control and weighed to the precise amount needed for production of fuel sticks.

Particle Blending, Fuel Stick Molding, and Inspection. — To facilitate inspection, the fuel particles are blended and cast into sticks rather than directly into graphite blocks. Fuel particles will be bonded so that if an irradiated fuel block is broken, the spread of contamination would be limited to the immediate area.

The particle blending step must blend the ThO_2-$^{233}UO_2$ particles with the ThC_2 and $^{235}UC_2$ particles to produce fuel sticks with less than 10% variance of uranium and thorium in any 1-in. length and less than 5% U and 6% Th variance from the desired loading in the entire fuel element. The operation must have sufficient loading accuracy so that the uranium and thorium loadings of the entire core will not vary more than 1%.

Conceptual design of a particle blender-filler device has been completed. This device will blend three types of particles while filling a fuel stick mold. Difficulty is expected in remotely adjusting the feeder orifices so that all three particle type feed streams will end concurrently.

Fuel sticks have been made at both GGA and ORNL. However, only limited information is available on the effect of irradiation on the fuel stick matrix material. To combat matrix shrinkage during irradiation, which may lead to cracking of the matrix, a bonding agent with greater amounts of stable carbon can be used. However, when more carbon is added to the bonding agent, the resin becomes more viscous, thereby lengthening the time required to inject it into the molds.

Little work has been done on fuel stick inspection. A combination of attenuation (gamma or x-ray) and gamma emission most likely will be used to determine the relative amounts of ^{233}U, ^{235}U, and thorium present as a function of position along the fuel sticks.

The prototype of the filler-blender device must be detail-designed, fabricated, and tested. A bonding method must be developed that can be used in a remotely operated production facility to produce a fuel stick matrix sufficiently stable to irradiation. Inspection methods must be developed to nondestructively test for the required loading tolerances.

If heavy mixtures of bonding resin and carbon are required to obtain a matrix sufficiently dense to withstand irradiation, other methods of loading the fuel stick molds must be developed to obtain the production rate necessary for a large facility.

Fuel Element Assembly, Fuel Stick Carbonization, and Fuel Element Inspection. — The fuel element must be loaded with approximately 200 fuel sticks of one or more fuel mixtures and fuel stick diameters. Various sticks must be matched to the proper holes in predrilled carbon blocks. The fuel sticks must be carbonized at 900°C; heating and degassing the blocks at the maximum operating temperature of the fuel particles may be required. The fuel element must be inspected for transferable contamination to limit buildup of contamination in the reactor.

Some conceptual design of equipment for fuel stick loading, element transfer, element inspection, and element bakeout has been done. The stick loader is envisioned to be a numerically controlled positioner, which will align the element under it. After being loaded, the fuel element will be transferred between cells in TURF. During this transfer the surfaces of the element will be inspected for contamination. If any is present, the cooling holes will be reinspected to assess the level of contamination. When the element is found to be clear of transferable contamination, it will be placed in the bakeout furnace. After bakeout, the element will be canned and placed in the casks used to ship the spent elements to head-end processing.

STATUS OF RECYCLE DEMONSTRATION

To demonstrate a technology for HTGR recycle, it is necessary to complete the following tasks: the supporting facility design and construction for pilot plant demonstration, the design and development of engineering-scale equipment for use in the pilot-scale facility, and finally the operation of an integrated reprocessing and refabrication system in the pilot-scale facility. Although it has not been specifically shown on the schedule that has been presented, we have constructed a facility for the development of the various steps involved in the recycle of ^{233}U-thorium for HTGR's. The facility, known as the Thorium-Uranium Recycle Facility (TURF), furnishes the necessary space, shielding, and facility equipment to perform all the operations required. The facility has been sized to accommodate complete recycle processes with equipment scaled down from anticipated production units so that a realistic and reliable basis for technical and economic analyses will exist. In fact, the facility has the capability of handling full-size fuel elements of the type used as reference in this paper. A photograph of the facility is shown in Fig. 9, a cutaway view in Fig. 10, a plan view of the first floor in Fig. 11, and a section elevation through the processing cells in Fig. 12. The primary zone of containment of the facility, consisting of six shielded cells and associated glove maintenance room and air lock, are depicted in Fig. 13. More details of the facility are reported in the literature. [25]

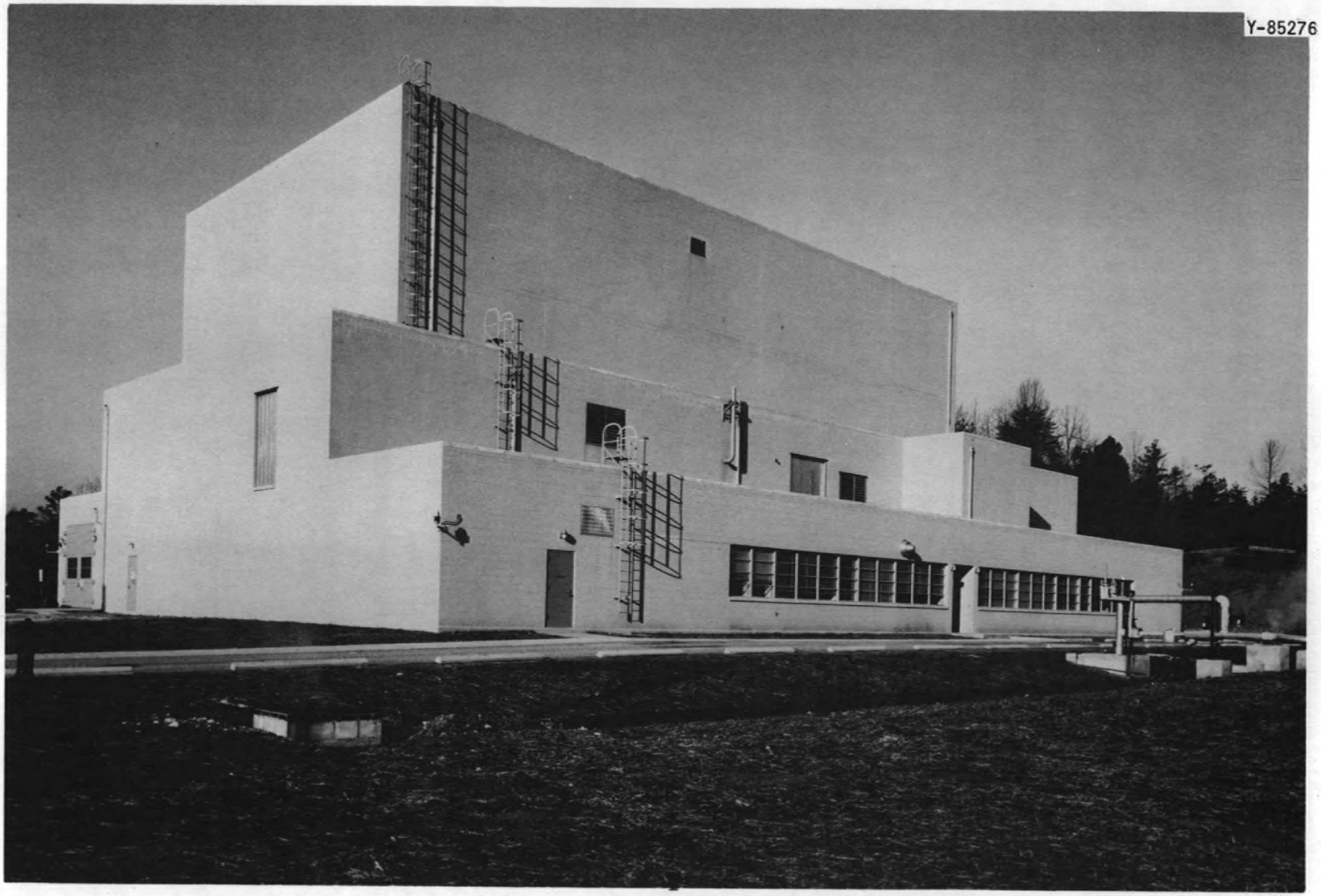

Fig. 9. Thorium-Uranium Recycle Facility.

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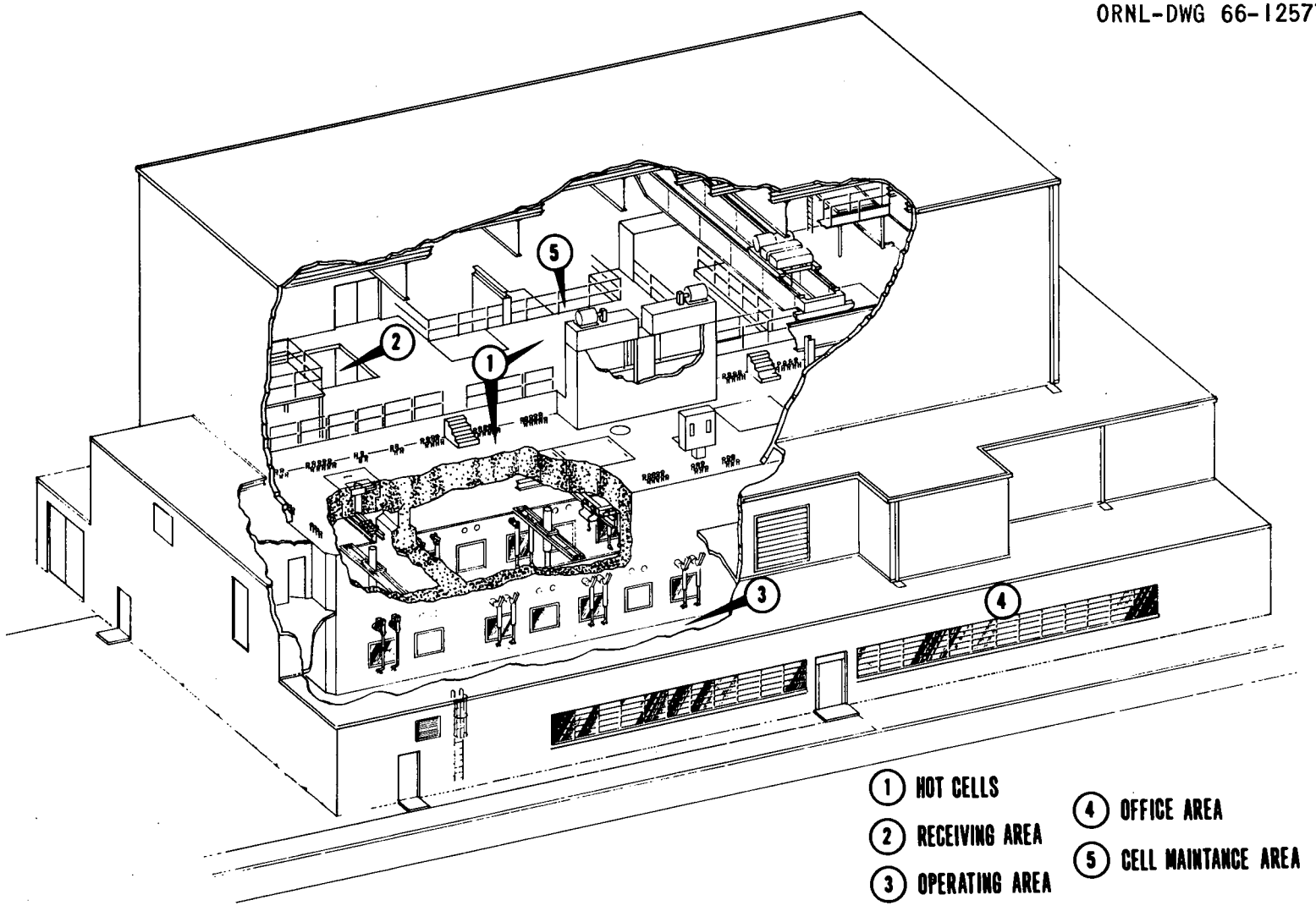

Fig. 10. Cutaway View of TURF.

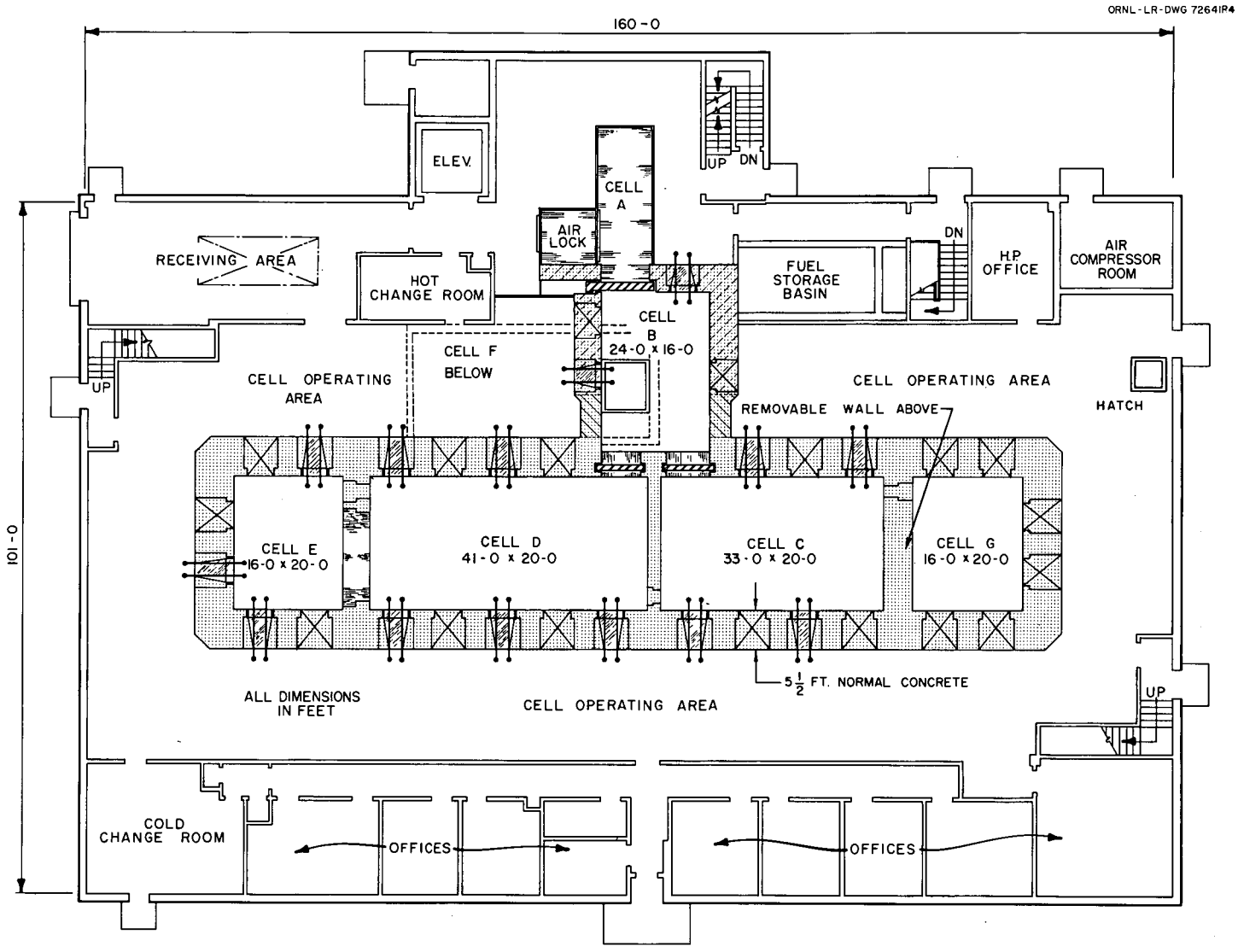

Fig. 11. Plan View of First Floor of TURF.

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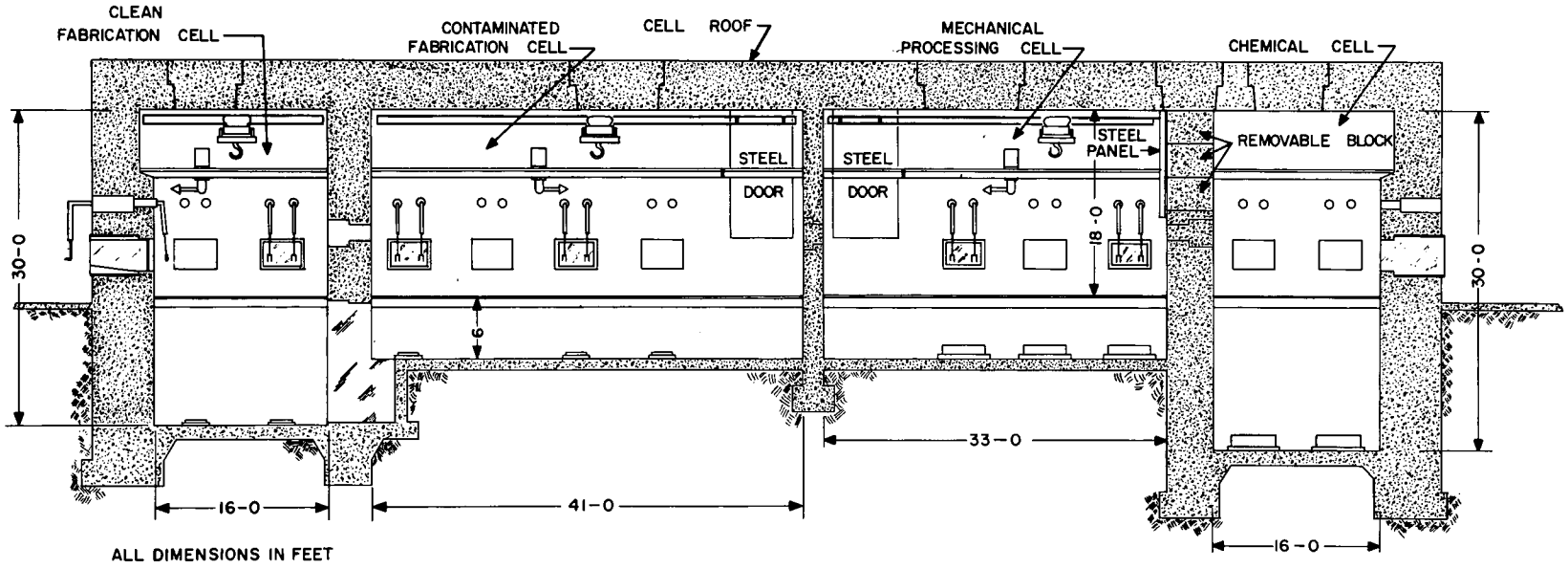

Fig. 12. Section Through TURF Processing Cells.

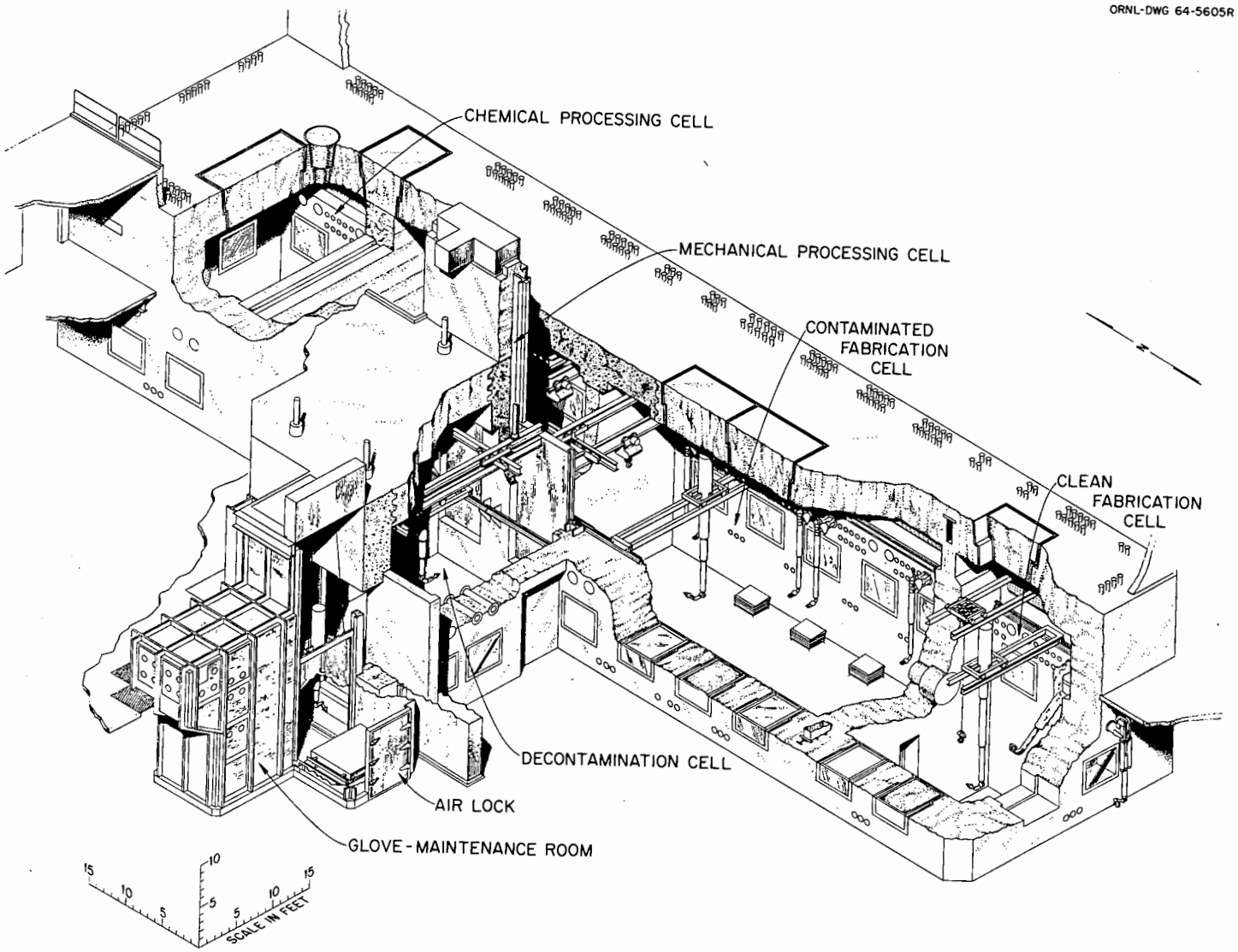

Fig. 13. Isometric of TURF Hot Cell Complex.

Although the facility has been placed into operation, the only task accomplished in it has been the preparation of ^{233}U-bearing salt for the Molten Salt Reactor Experiment. [26] No equipment for HTGR recycle has been constructed. With the exception of solvent extraction processing, none of the equipment for remote operation has progressed beyond the stage of conceptual design. Certain prototype equipment for engineering-scale development has been built, and some of it operated as indicated in the previous discussion. The conceptual designs for the various processes have been reported in several places. [3,4,18,19]

STATUS OF FUEL IRRADIATION PERFORMANCE

The extensive development of fluidized-bed coating technology has enabled one to define conditions for deposition of pyrolytic carbon coatings that have a wide range of properties. [27] While the systematic study of conditions for deposition of SiC has been less extensive, coatings with suitable structure and density can be readily obtained. Results from irradiation testing of coated particles and of strip specimens of coatings indicate that to withstand high fast-neutron exposures carbon coatings should have densities between 1.8 and 1.95 g/cm^3 and must be isotropic or very nearly so. [28,29]

Mathematical models have been developed that incorporate the data from observed behavior of fuel and coating materials and enable one to predict the performance of coated particles in a given environment. [30,31] Good correlations between observed and predicted performance have been noted at fast-neutron exposures as high as 5×10^{21} neutrons/cm^2 (>0.18 Mev) and burnups greater than 30 at. % heavy metal, although not in the same experiment. These results suggest confidence in the models and in extrapolations that enable one to design coated particles to withstand the full reference HTGR exposures, that is, 20 at. % heavy metal burnup at fast neutron fluences up to 8×10^{21} neutrons/cm^2. Tests in progress by GGA and ORNL on systematically selected coated fissile and fertile particles should demonstrate whether these extrapolations to full exposure are justified.

Injection techniques have been developed for bonding coated particles into fuel rods that have acceptable properties, but few results are available from irradiation testing of bonded fuel. In two sweep capsule experiments at ORNL on bonded carbon-coated UO_2 particles, burnups greater than 30 at. % were observed without coating failure, as shown in Fig. 14 and as predicted by the mathematical model. [32] However, maximum fast-neutron exposures in these experiments were only approximately 1.2×10^{21} neutrons/cm^2. Results from a recent experiment in the HFIR target region, in which bonded coated particles were irradiated to fast-neutron exposures as high as 6×10^{21} neutrons/cm^2, showed that the binder carbon shrinks about 25 vol %, after which the bonded rods have little if any strength. The severe degradation of the bonded specimens is illustrated in Fig. 15. These initial experiments were conducted on

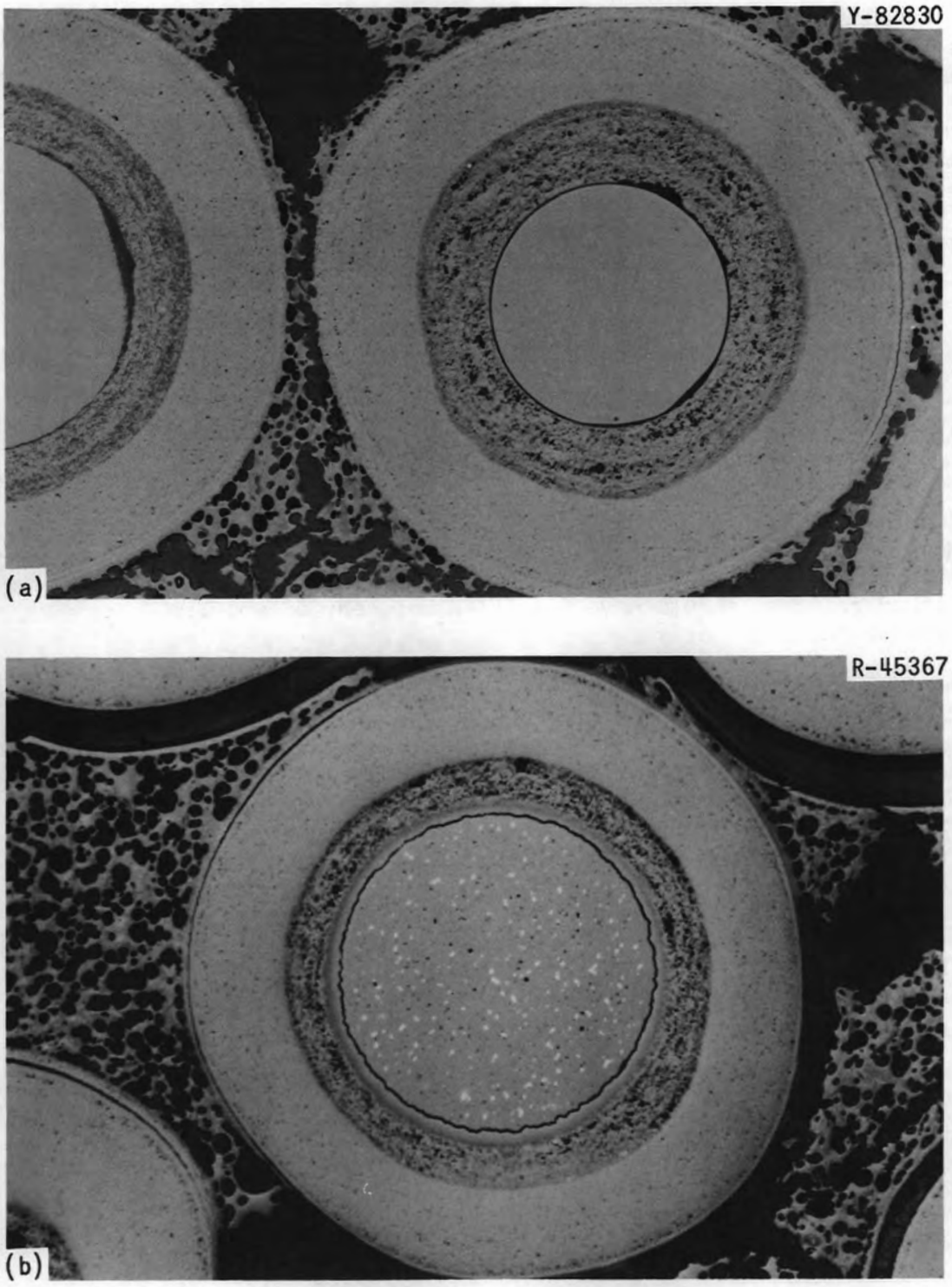

Fig. 14. Polished Sections of Bonded Fuel Rods Containing UO_2 Microspheres Coated with Buffer and Low-Temperature Isotropic Coatings Plus Additional Nonbonding and Sacrificial Layers. (a) Unirradiated. (b) Irradiated to 35 at. % burnup of heavy metal at 1250°C in experiment A9-15. Note that the sacrificial layer separates from the nonbonding layer and remains bonded to the carbon matrix. As polished. 200×.

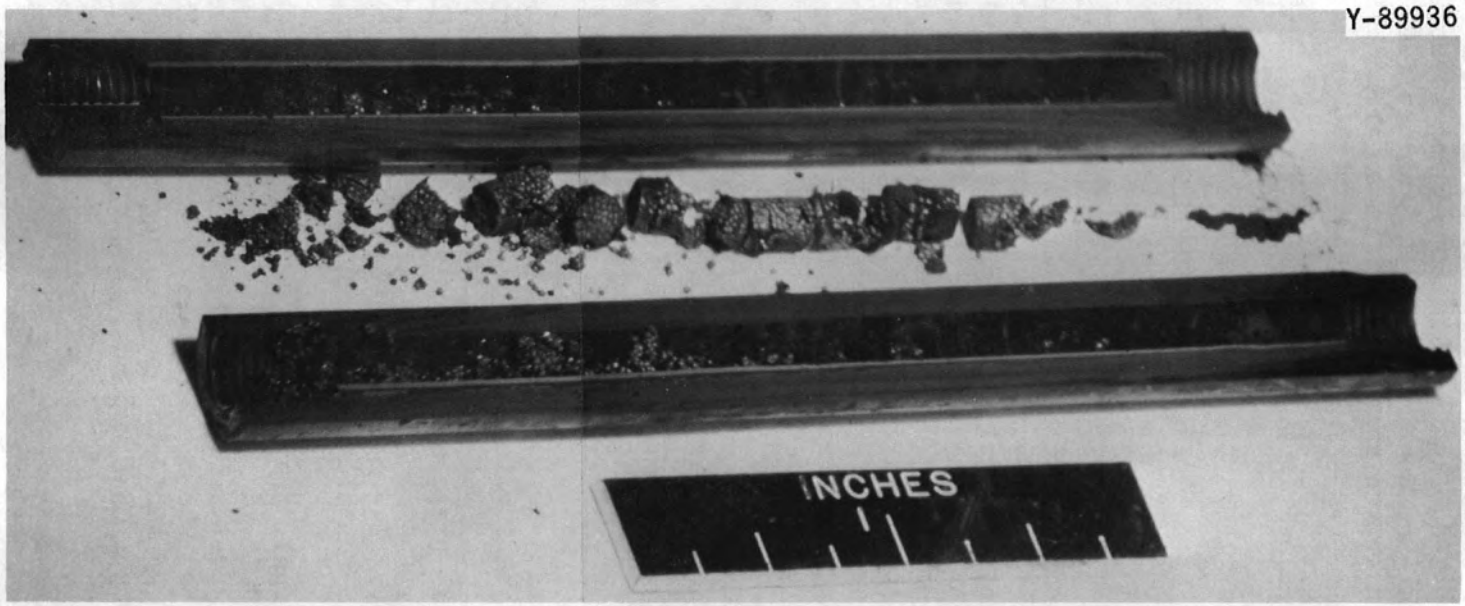

Fig. 15. Bonded Coated Particle Specimens and Graphite Holder After Irradiation to Fast-Neutron Fluence of 5×10^{21} neutrons/cm^2 at Approximately 750°C. All specimens were very friable, but those bonded with a resin-graphite mix containing 25% rather than 15% graphite were stronger.

coated particles that had a nonbonding layer in the coating. Further tests on fuel rods bonded by alternate procedures are in progress by ORNL and GGA. The results to date indicate that interaction of the binder with coatings during irradiation may impose severe stresses in the outer layers of the coatings, and that bonded fuel rods cannot be expected to retain appreciable strength after irradiation to high fast neutron exposures. However, the limited experience available indicates that the bonded particles were retained within the graphite sleeve in which they were irradiated to high fast-neutron exposure.

PROJECTED ECONOMICS

Although the present state of development on an engineering scale for the various recycle processes is limited, we have attempted to project the economics for various modes of recycle operations. Recycle operations can provide considerable cost savings in refueling reactors. However, if only one reactor exists, the cost of construction and operation of the complex facilities necessary for recycle would obviously not be justified. Therefore, it is appropriate to consider the cost of the various steps involved in both the non-recycle and recycle cases and to determine on the basis of these costs the optimum delay period before recycle operations should begin in a growing HTGR economy. We have examined this in some detail at ORNL, using as a reference the 1000-Mw reactor recently proposed by GGA and the reference flowsheet presented in this paper. [33]

Cost of Various Steps in the HTGR Fuel Cycle

The cost of the various steps involved in the fuel cycle for HTGR's may be applied according to whether or not fuel is recycled. If not, costs are incurred for the ore, its conversion to UF_6, the separation costs, and material preparation and fabrication. In addition, in non-recycle operation, the long-term storage of spent fuel blocks must be taken into account. The material preparation, fabrication, and block storage costs as a function of plant capacity are given in Fig. 16.

For recycle fuel, the costs listed under the non-recycle case are also incurred; and in addition, costs for chemical processing, reconstitution of fuel, refabrication of fuel, and waste storage are involved. Figure 17 summarizes the cost of these steps as a function of throughput or capacity. Most of the costs as shown in these two figures have been given in various ORNL reports, as well as the basis for these costs. [4,33–37]

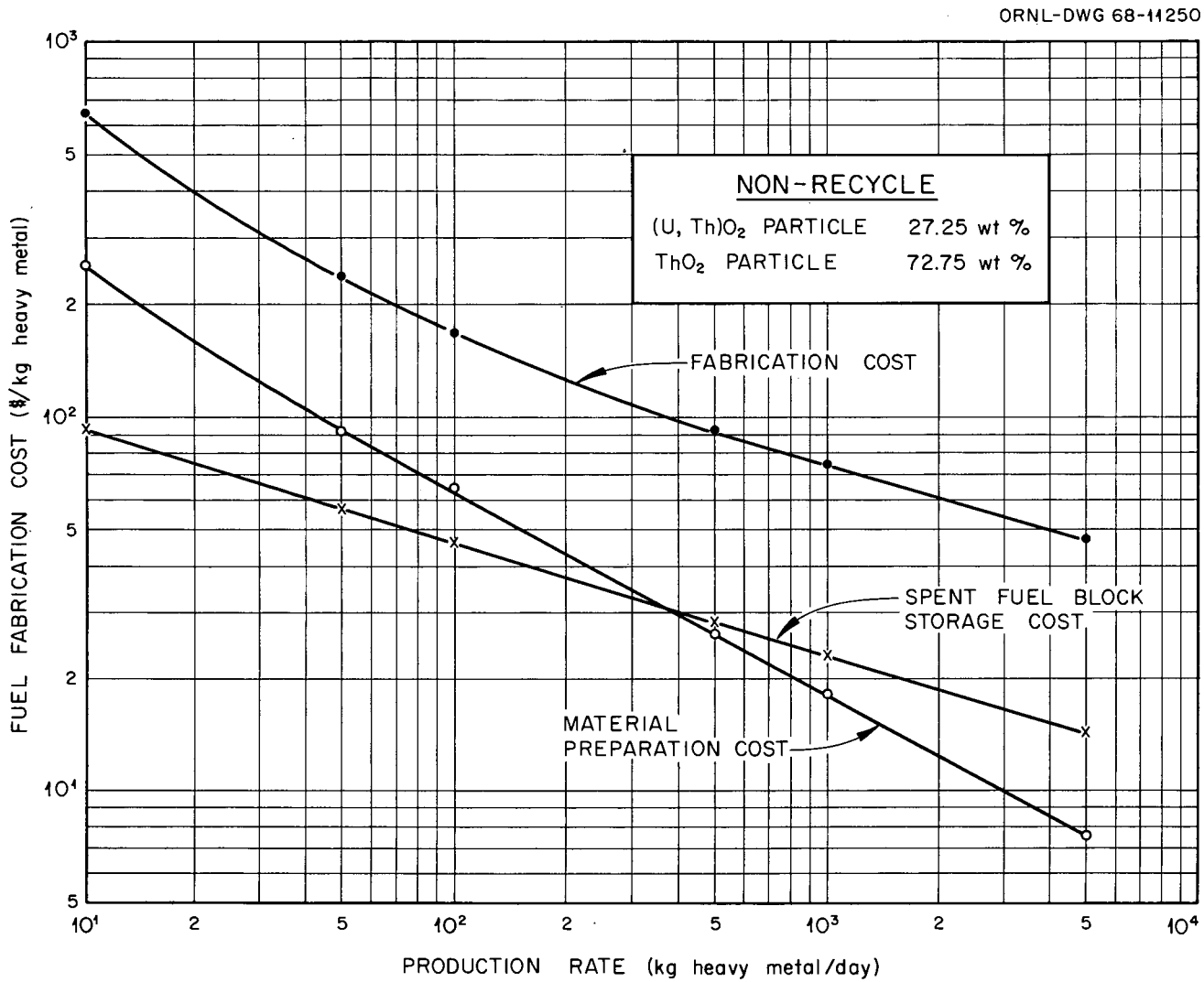

Fig. 16. Costs of Steps in Non-Recycle Process.

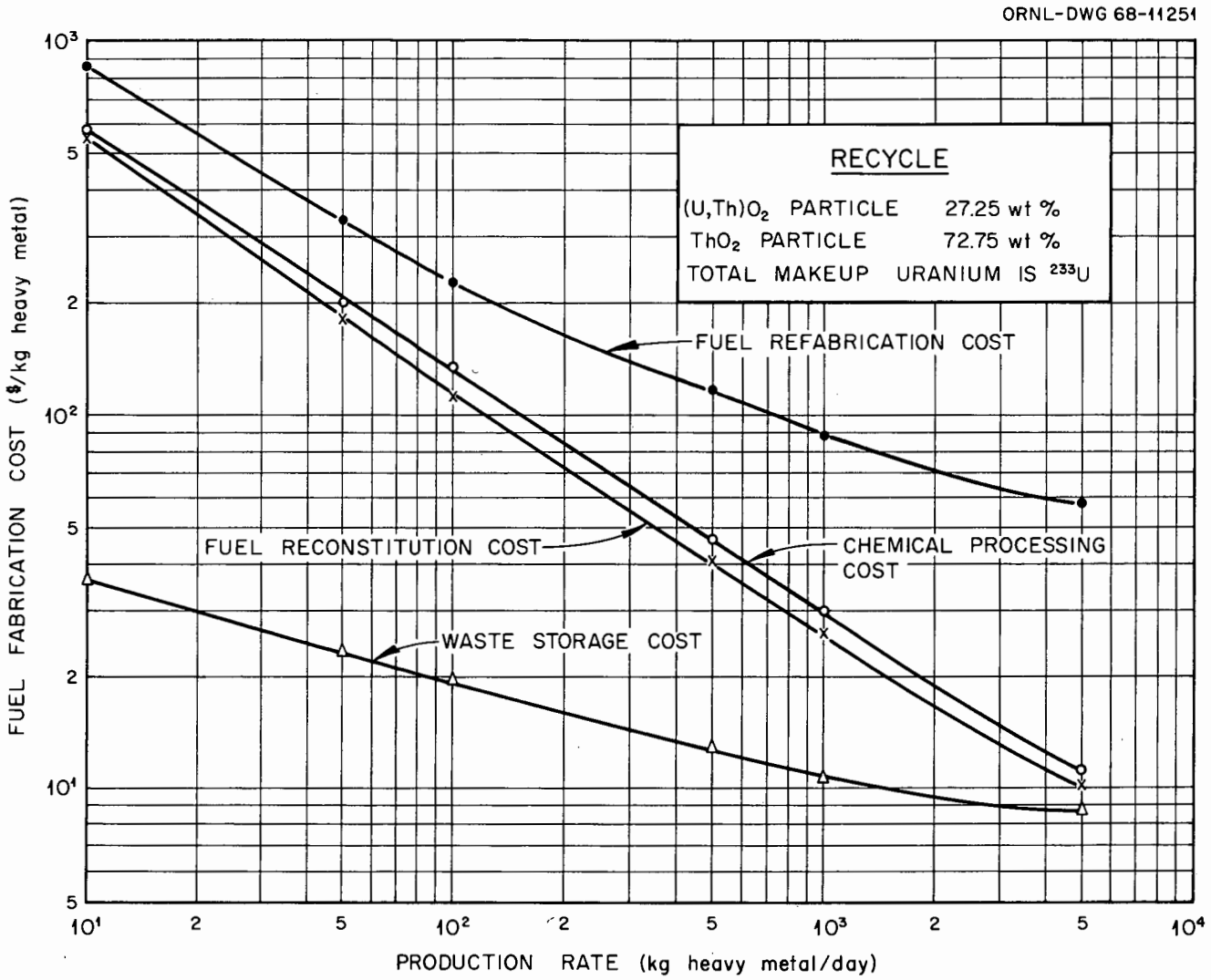

Fig. 17. Costs of Steps in the Recycle Process.

Integrated Costs

The above cost data can be combined to compare the non-recycle case with the recycle fuel as a function of throughput. This comparison is provided in Fig. 18. In formulating these curves, we assume that the various processing facilities and storage facilities are operating in an equilibrium cycle for the given production rate. This is not true in a growing HTGR economy, but is just an assumption to simplify the presentation of Fig. 18.

Calculation of Optimum Delay for Recycle

To calculate the economically optimum time for recycle operation startup, the complete fuel cycle for both non-recycle and recycle modes of reactor operation, must be well understood. Figure 19 presents, in simplified schematic form, the description of the steps in both modes of operation.

We can arrive at the economically optimum time for recycle operations to start by calculating the difference, in cost to the total economy, between various delays in recycle operations compared with the startup of recycle operations when the first 1000-Mw HTGR is refueled. The advantage of this approach is that certain items that are difficult to estimate accurately cancel. One such item is shipping. Whether the reactor is being recycled or not, the used fuel blocks must be shipped, either to long-term storage or reprocessing. Another item that can be canceled is the cost of thorium. This cost would be extremely difficult to predict for a long range study. We assume that the cost of recycling thorium (after a 15-year storage period) would be comparable to purchasing fresh thorium. Any error in this assumption is minimized when the costs are considered on a present worth basis. Hence, the thorium costs vary an insignificant amount when the recycle startup date is varied and thus can be neglected in the calculation.

Other items, such as coating, cannot be neglected in the calculations. Although the coatings required for non-recycle and recycle operations are very similar, the radioactivity of the materials being coated is sufficiently different to require hooded operation in the non-recycle case and remote operation in the recycle case. [38,39]

To simplify calculations, all operations are grouped into four categories:

1. ^{235}U fuel element fabrication (non-recycle element fabrication),
2. ^{233}U reprocessing and refabrication (recycle element fabrication),
3. irradiated fuel block storage,
4. reprocessing by-product storage.

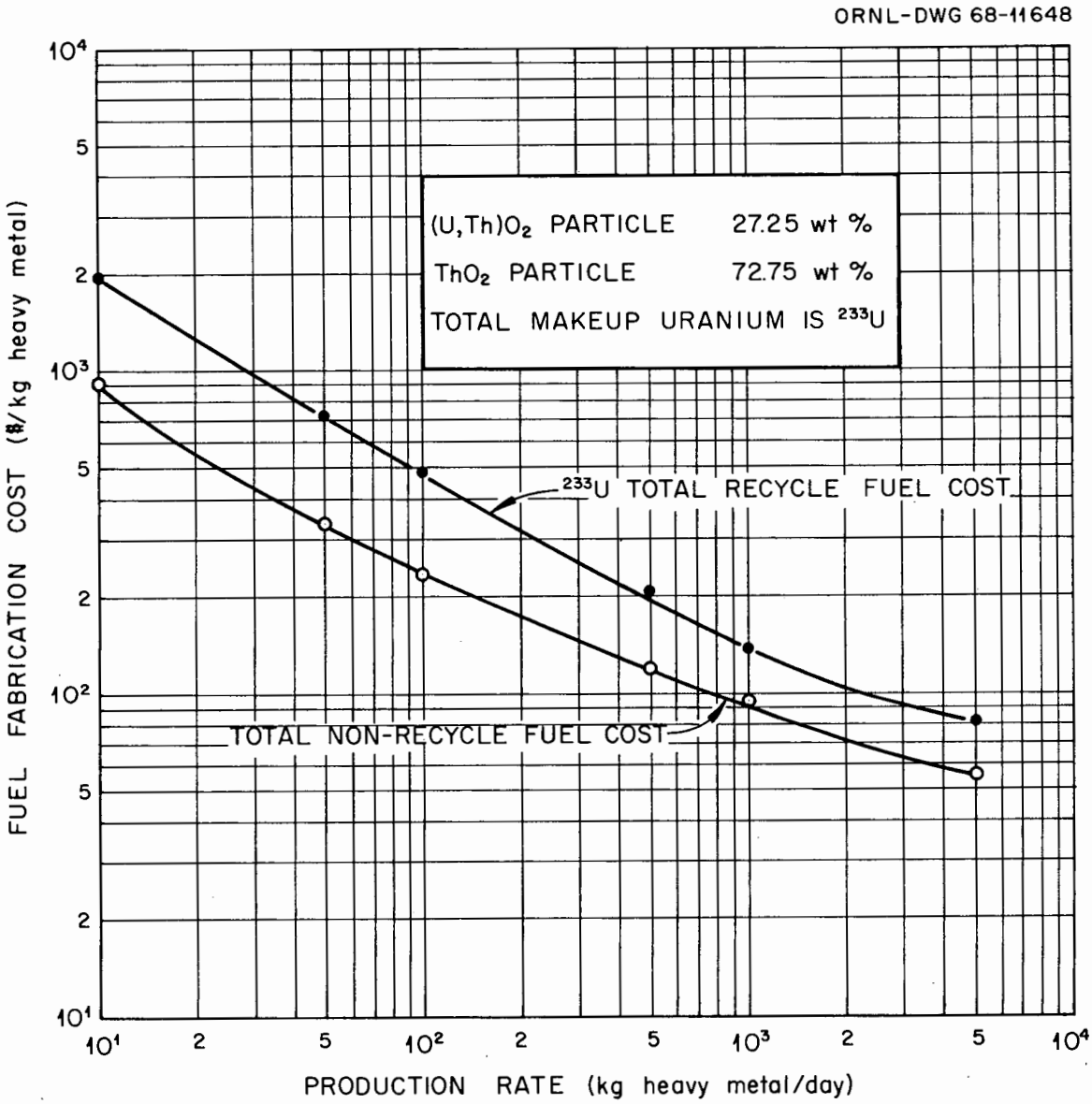

Fig. 18. Comparison of Non-Recycle and Recycle Processing Costs.

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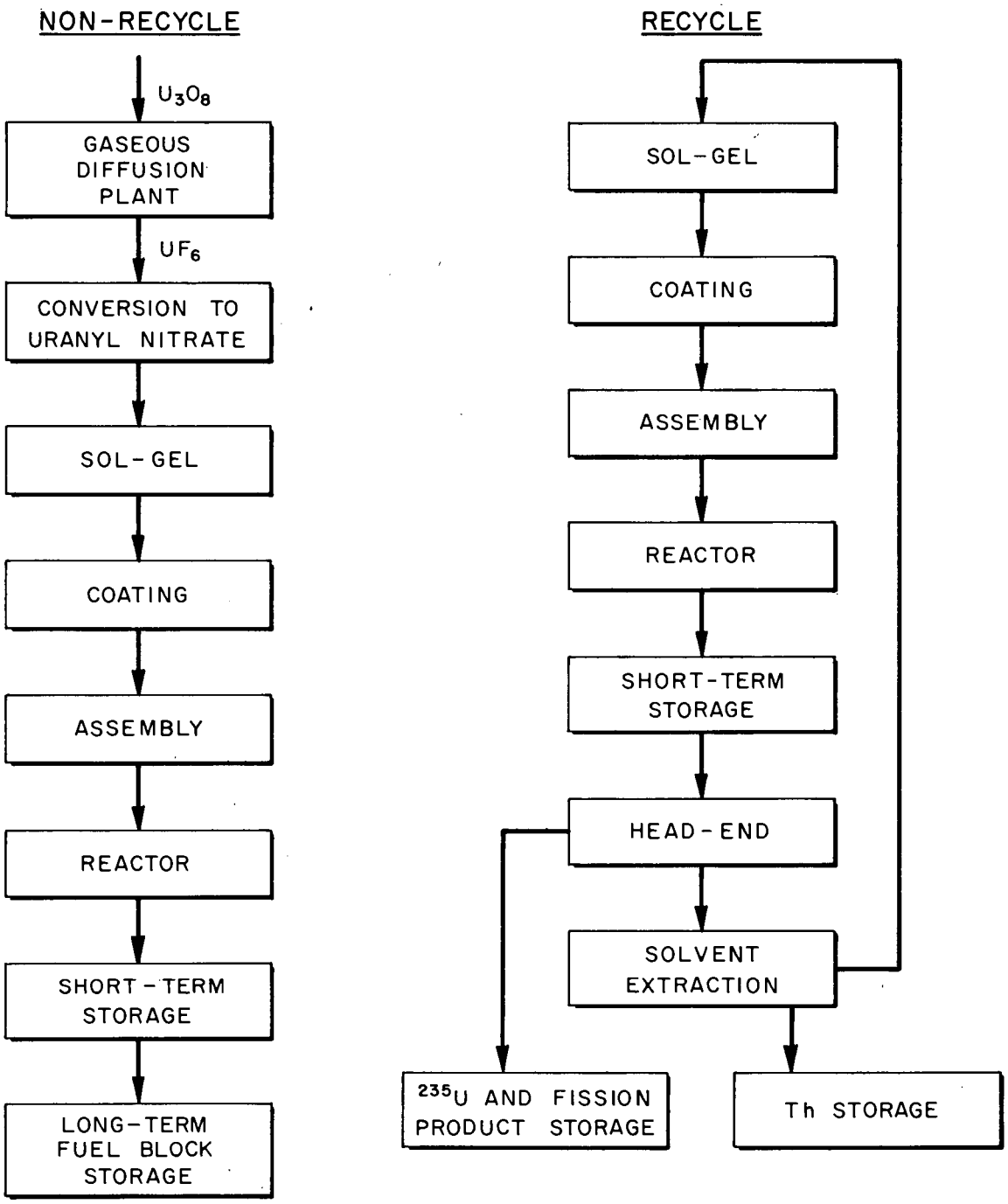

Fig. 19. Non-Recycle and Recycle Fuel Cycle Processes for the HTGR.

The first category, ^{235}U fuel block fabrication, includes all operations from the purchasing of ^{235}U from the gaseous diffusion plant to the final assembly of the fuel block. (See Non-Recycle Process in Fig. 19.) This series of processes is used for all the fuel for non-recycle operations and is also used for makeup fuel in recycle operations. The second category, ^{233}U reprocessing and refabrication, is concerned with the steps in receiving ^{233}U from the spent fuel blocks and refabricating this material into fuel blocks for recycle operations. (See Recycle Process in Fig. 19.) The third category, irradiated fuel block storage, consists only of storing spent fuel blocks which are not processed in a recycle facility, as would be the case when the reactor industry is operating before recycle facilities are completed. The fourth category, reprocessing by-product storage, is a combination of all storage connected with the reprocessing industry. The main items in the storage are the ^{235}U, which is contaminated with ^{236}U and fission products, and thorium, which is contaminated to a lesser degree with fission products.

Once the costs of the four categories mentioned above are obtained for different plant sizes, they can be combined with the mass balances for non-recycle and recycle operation of a 1000-Mw HTGR [40] and the assumed growth rate of the HTGR industry to give the economically optimum time for startup of recycle operations.

A simplified outline of the calculational procedure is shown in Fig. 20. The mass balances, industry growth, and assumed start of recycle operations are combined to give the total mass balance for the HTGR industry. From this mass balance, demand curves for the four categories of the HTGR fuel industry are calculated. Next, plant construction schedules for each of the four categories are selected by an optimizing procedure, [41] and plant sizes and costs are calculated.

The plant costs, ^{235}U fuel block fabrication costs, ^{233}U reprocessing and refabrication costs, fuel block storage costs, reprocessing by-product storage costs, and demand curves are combined to give yearly costs. These yearly costs are then converted to present worth and added to give the cost of all the items considered for a particular time of recycle startup. The entire calculation is repeated for other times of recycle startup, and the optimum time can be selected and the relative additional costs of other startup dates can be found.

The assumptions used in the delay analysis were as follows. Enriched ^{235}U was assumed to cost $11,175 per kg. It was assumed that 1 kg of ^{235}U can be replaced by 0.833 kg ^{233}U. There were no restrictions on the amount of ^{235}U that can be replaced. One other simplifying assumption made was that the ^{233}U and ^{235}U would be fabricated in separate fuel blocks, so the ^{235}U could be fabricated in a hooded plant and the ^{233}U-bearing fuel elements in a remote plant. This is just a cursory description of the method used in this analysis. Further details are given in other reports. [41–43]

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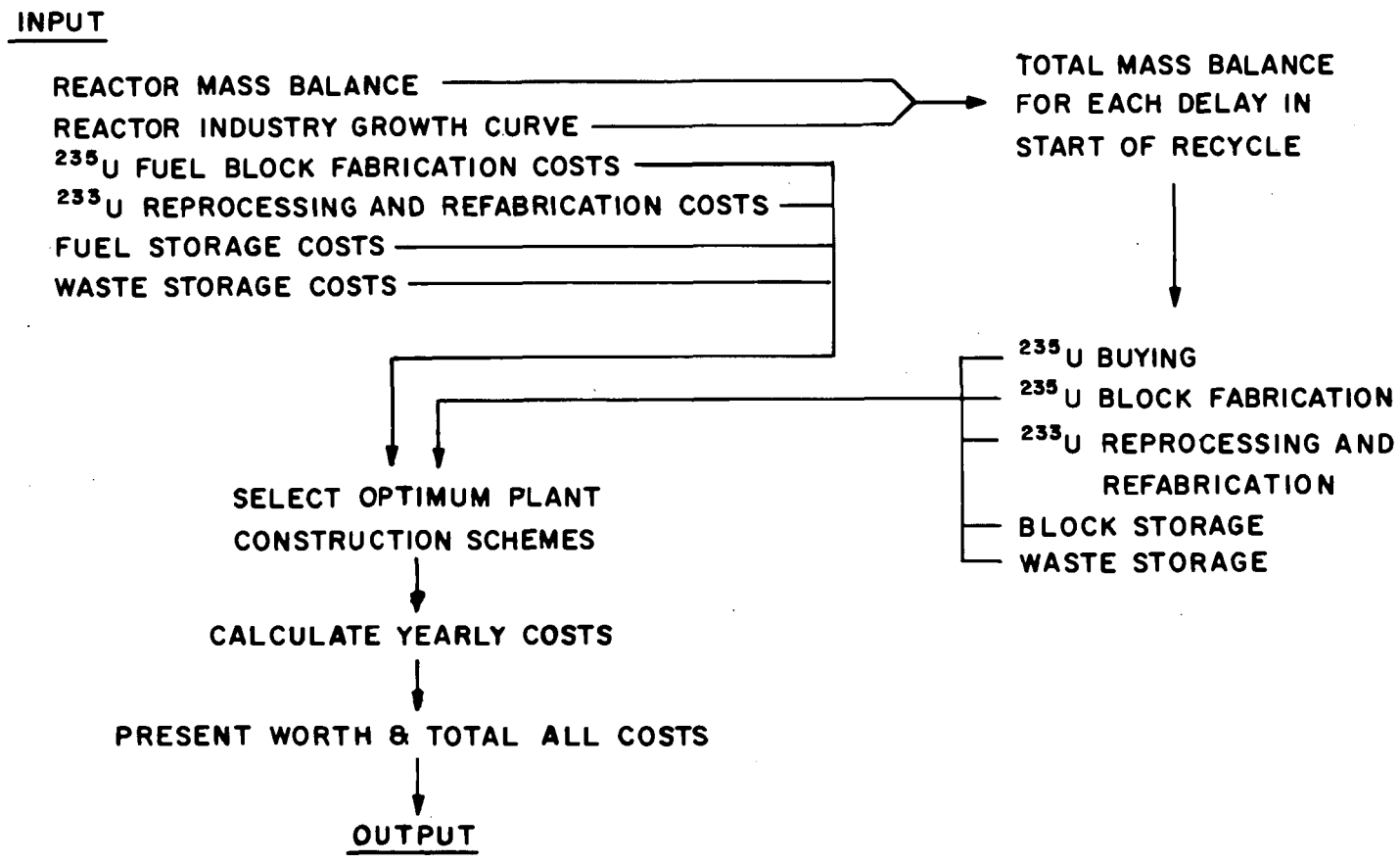

Fig. 20. HTGR Recycle Cost Calculation Method.

To carry out the analysis, the growth rate of Fig. 21 was used in the study. Using this growth curve, the cost adjusted with a present worth factor of 7% to 1974 for the various items of HTGR recycle are shown in Fig. 22. The total curve exhibits a minimum, which is the optimum time for start of recycle. A more explicit method of presentation of the minimum time for delay is shown in Fig. 23, which shows the difference in costs between recycle operations starting in 1981 and recycle operations starting at economically unfavorable dates. Inclusion of the development cost does not shift the economically optimum time for start of recycle significantly.

For the assumed HTGR economy, Table II shows the optimum plant startup dates, plant sizes, and capital costs for the ^{235}U fuel block fabrication plants and the ^{233}U reprocessing and refabrication plants. The plant sequences were optimized with a code that uses capital cost and operating cost as a function of plant size and also accounts for the cost of unused overcapacity. [41] The first non-recycle fabrication

Table II. Plants Selected in Optimum Case

Type Plant	Startup Date	Size (kg heavy metal/day)	Cost ($ million)
^{235}U fuel element fabrication (non-recycle plant)	1974	540	35
	1977	1230	49
	1992	630	37
^{233}U Reprocessing and refabrication (recycle plant)	1981	1190	141
	1996	1080	136

plant starts in 1974. The second plant starts in 1977 and runs concurrently with the first plant until the first plant is obsolete in 1989. The third plant starts in 1992 when the second plant is declared obsolete. The first recycle plant starts in 1981. The second plant starts in 1996 when the first plant is declared obsolete.

Other growth rates were used as input to the program. The total cost curves, with scales modified for curve comparison, are presented in Fig. 24. The curves included are the growth curve previously discussed (39 reactors), a higher level, longer growth period curve (105 reactors), and a growth curve that reaches a maximum growth rate and continues growing at that level. No minimum illustrated in Fig. 24 was displaced more than one year from the minimum exhibited by the original case, regardless of the greatly differing growth curves.

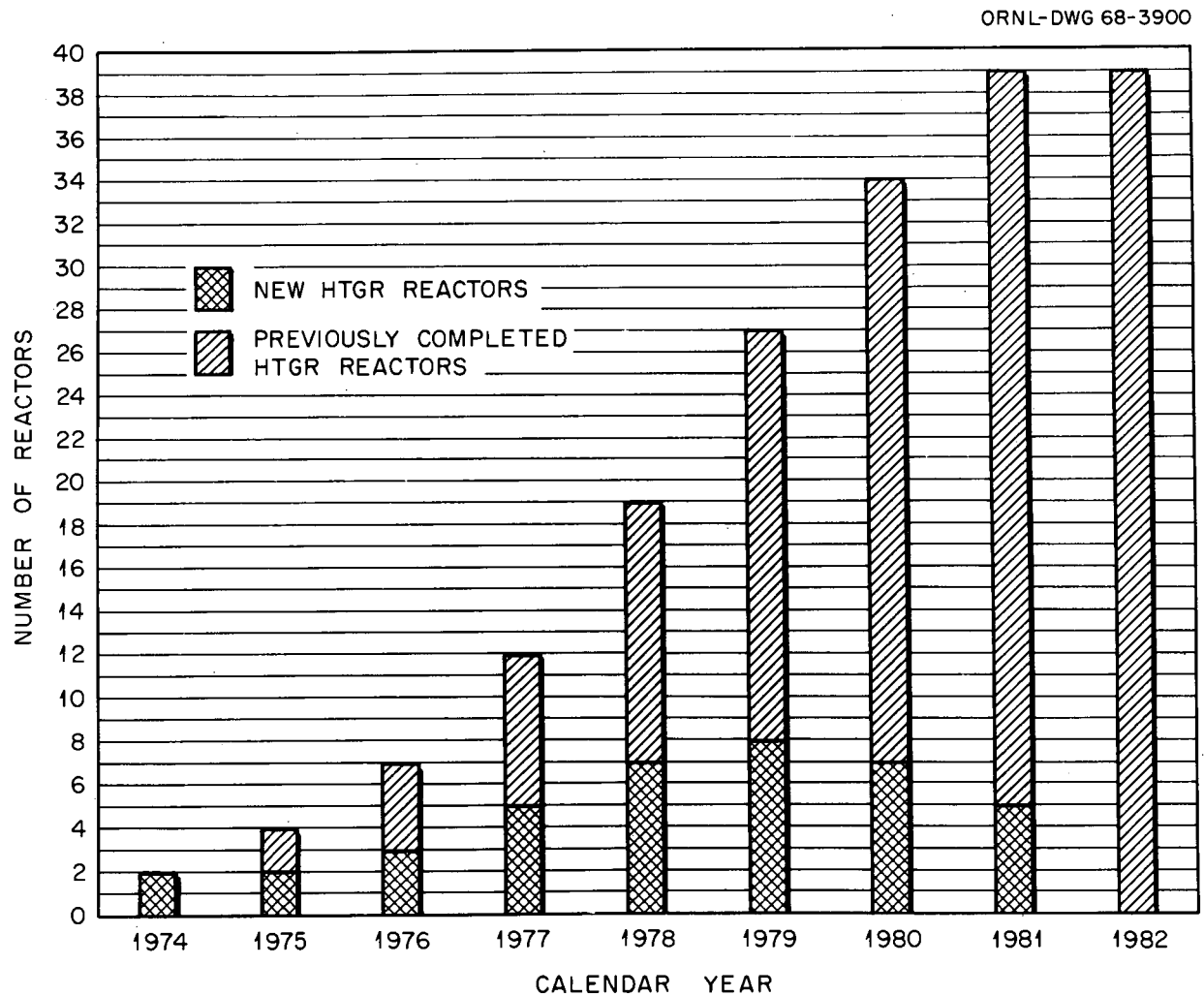

Fig. 21. 1000-Mw HTGR Growth Rates.

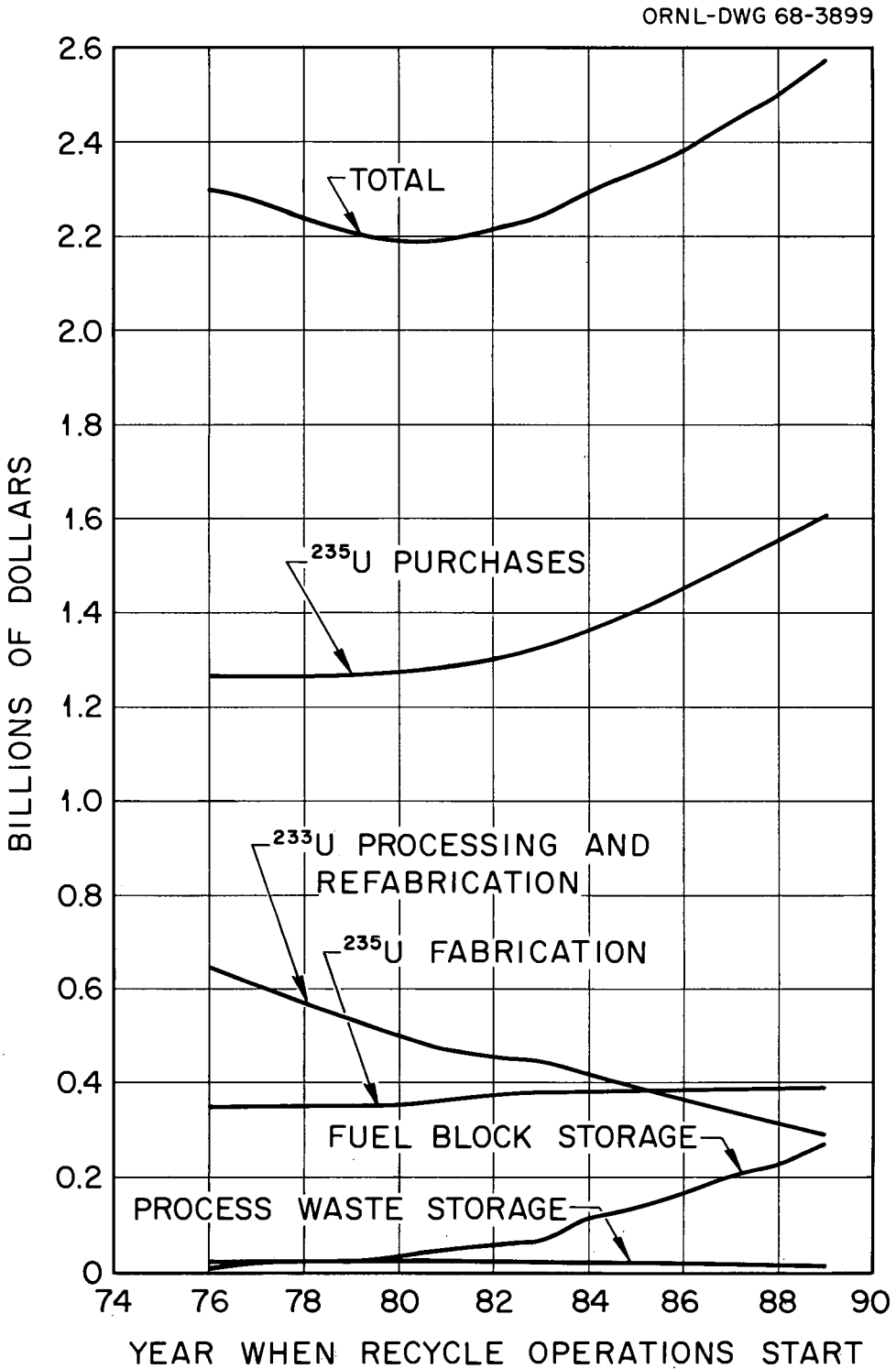

Fig. 22. Costs of HTGR Recycle Economy as a Function of Time When Recycle is Started.

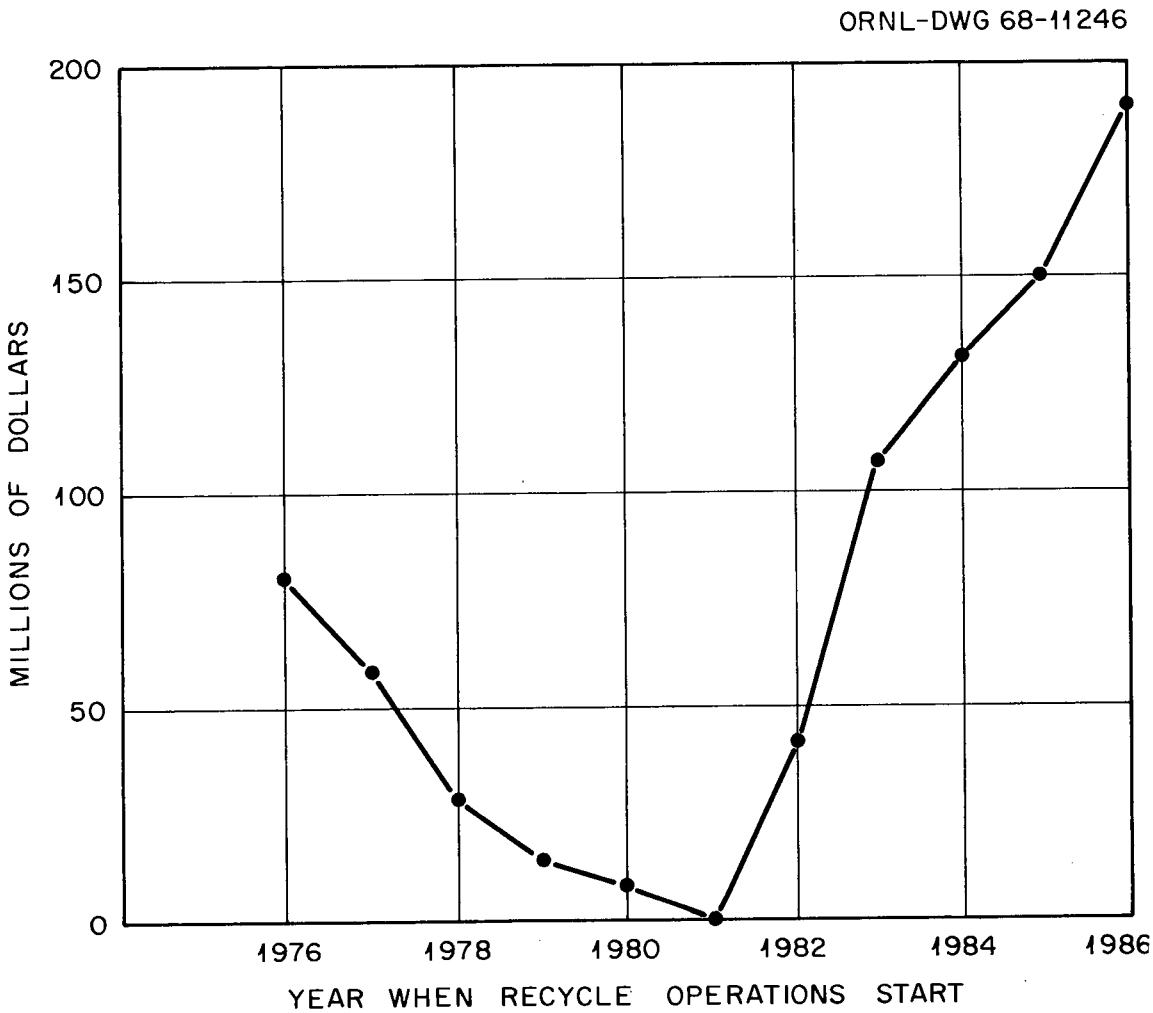

Fig. 23. Cost Penalty for Starting Recycle Operations at Economically Unfavorable Time.

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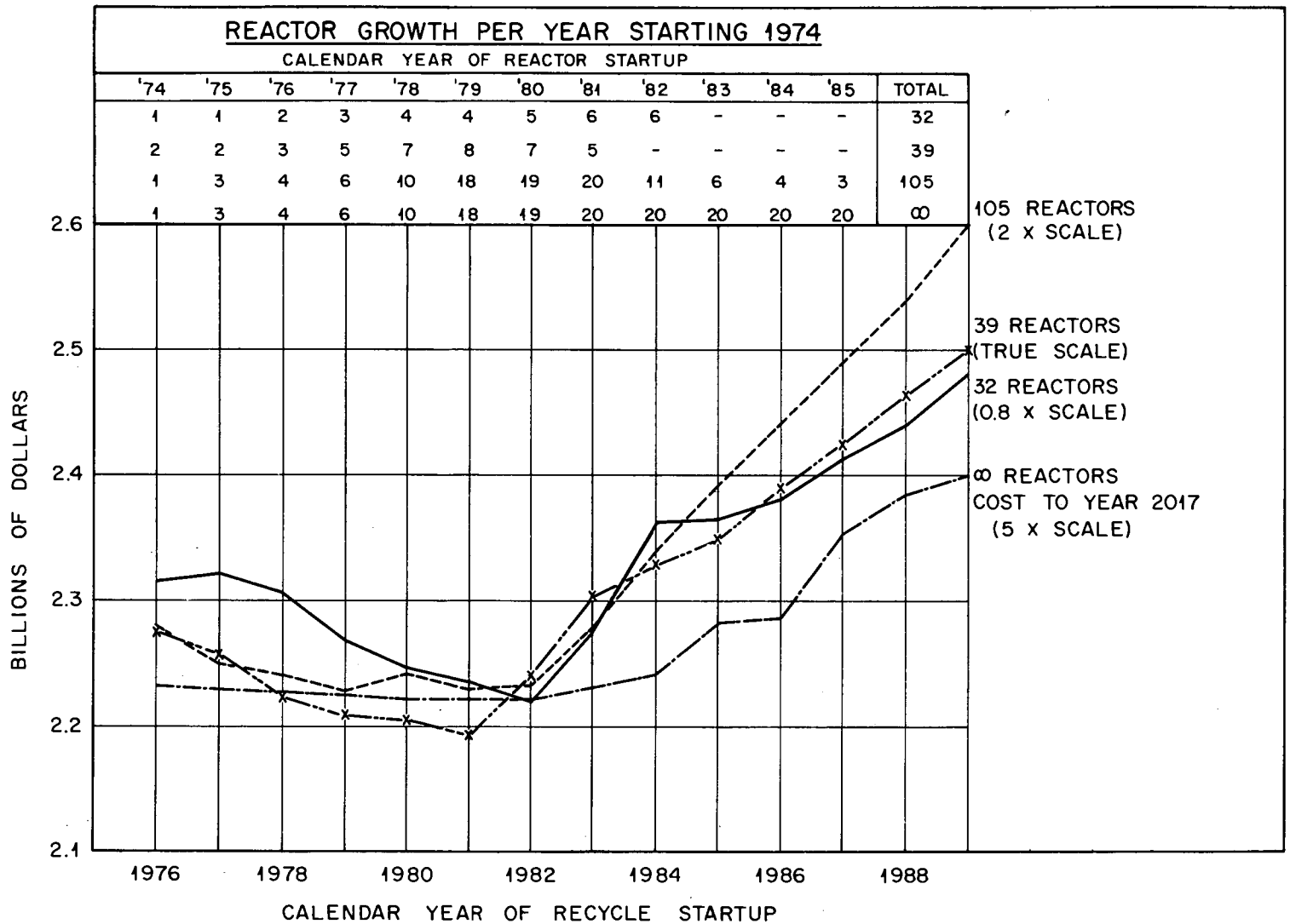

Fig. 24. Variation of Total Cost (at 1974 worth) with Time of Recycle Startup.

An important factor concerning the HTGR recycle cost was the interest rate (7% in this study) used in calculating the present worth. Figure 25 shows the total costs on a present worth basis for interest rates varying from 0 to 14%. The minimum is observed to occur at the same time for all curves, thus revealing the insensitivity of the optimum delay period to the interest rate. The reprocessing and refabrication cost estimates were based on the best available data. The source of greatest error would be the scaling factor used in estimating the savings in capital and operating costs as the plant size is increased. To test the effect of such an error, the estimated scaling factors of 0.51 for capital and 0.62 for operation costs were increased to the highly conservative values of 0.75 for capital and 0.9 for operation costs. The effect of this shift is shown in Fig. 26; the resulting minimum point shifted only one year.

From the study conducted, we conclude that the economically optimum time for start of recycle operations is 7 ± 1 years after startup of the initial 1000-Mw HTGR.

SUMMARY

High-temperature gas-cooled reactors using the thorium-^{233}U fuel cycle are being developed because of their potential for achieving low power costs and high fuel utilization. Attainment of this potential depends on successfully developing HTGR fuel recycle. Accordingly, a comprehensive fuel recycle program is required having as its objective construction and operation of a commercial central reprocessing and refabrication plant.

The work is focused on a reference recycle fuel element for a near-term 1000-Mw (electrical) HTGR. The reference element is similar to the fuel element currently designed by Gulf General Atomic for the Public Service of Colorado Fort St. Vrain Reactor. The reference recycle fuel particle is a sol-gel-prepared $(^{233}U,Th)O_2$ particle with pyrolytic carbon and SiC coatings. Non-recycle fuel particles are mixed with recycle fuel particles and molded into sticks with a carbonaceous binder and inserted into carbon fuel blocks during the fabrication process.

The development work includes processes for removal of intact particles from spent fuel blocks, separation of ^{235}U particles from the others, dissolution of the fissile and fertile materials, separation and purification of these products by aqueous processing, manufacture of mixed oxide particles by the sol-gel process, and refabrication of the prismatic fuel elements. The present status of the recycle development is: (1) laboratory and small-scale hot cell development work has been done on head-end processing, (2) aqueous processing methods have been demonstrated on an engineering scale for purification and separation of the fissile and fertile components, and (3) the sol-gel process and the refabrication processes have, in general, reached the stage where

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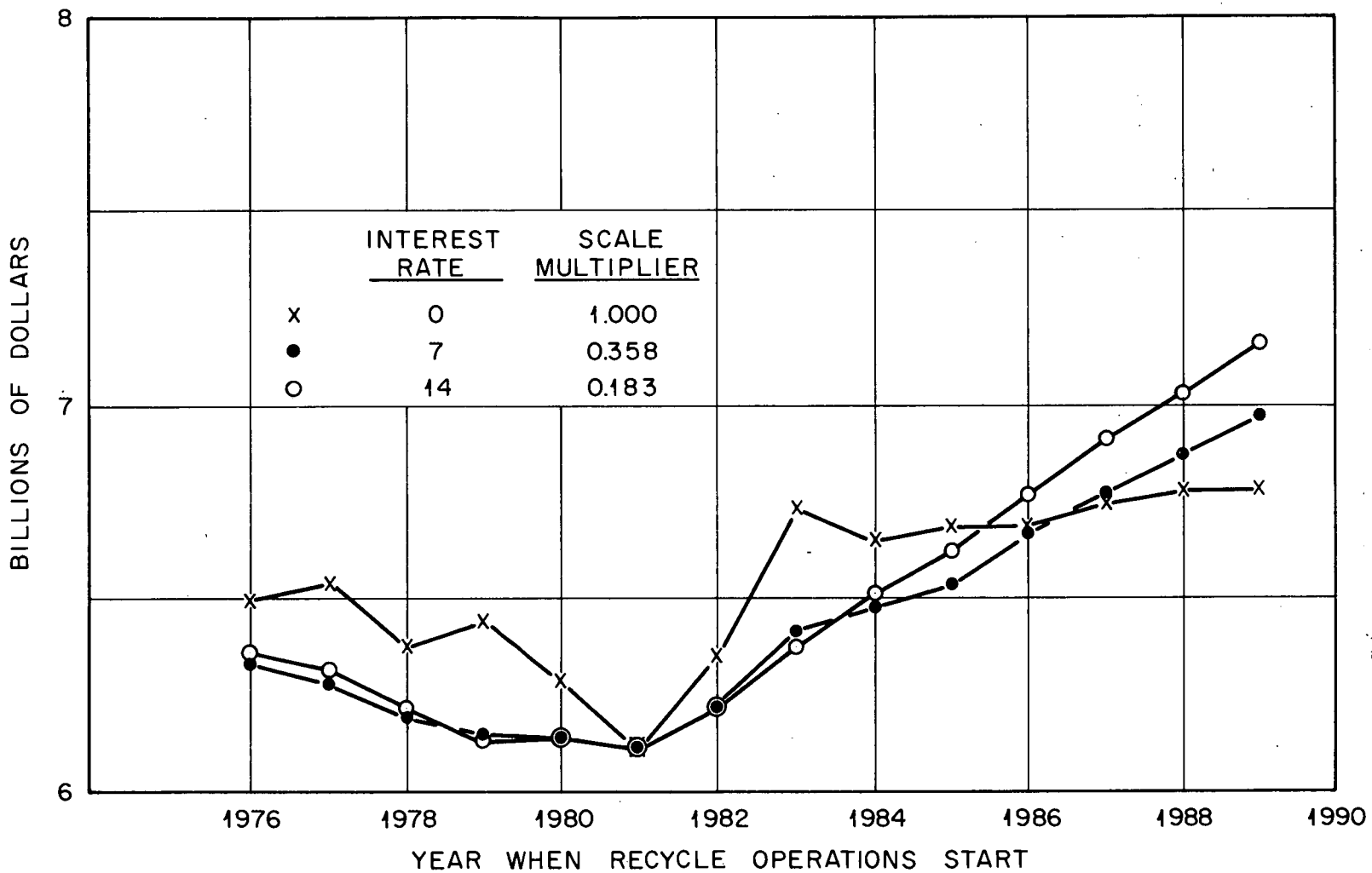

Fig. 25. Total Cost (at 1974 worth) vs Time for Recycle Startup for Various Interest Rates.

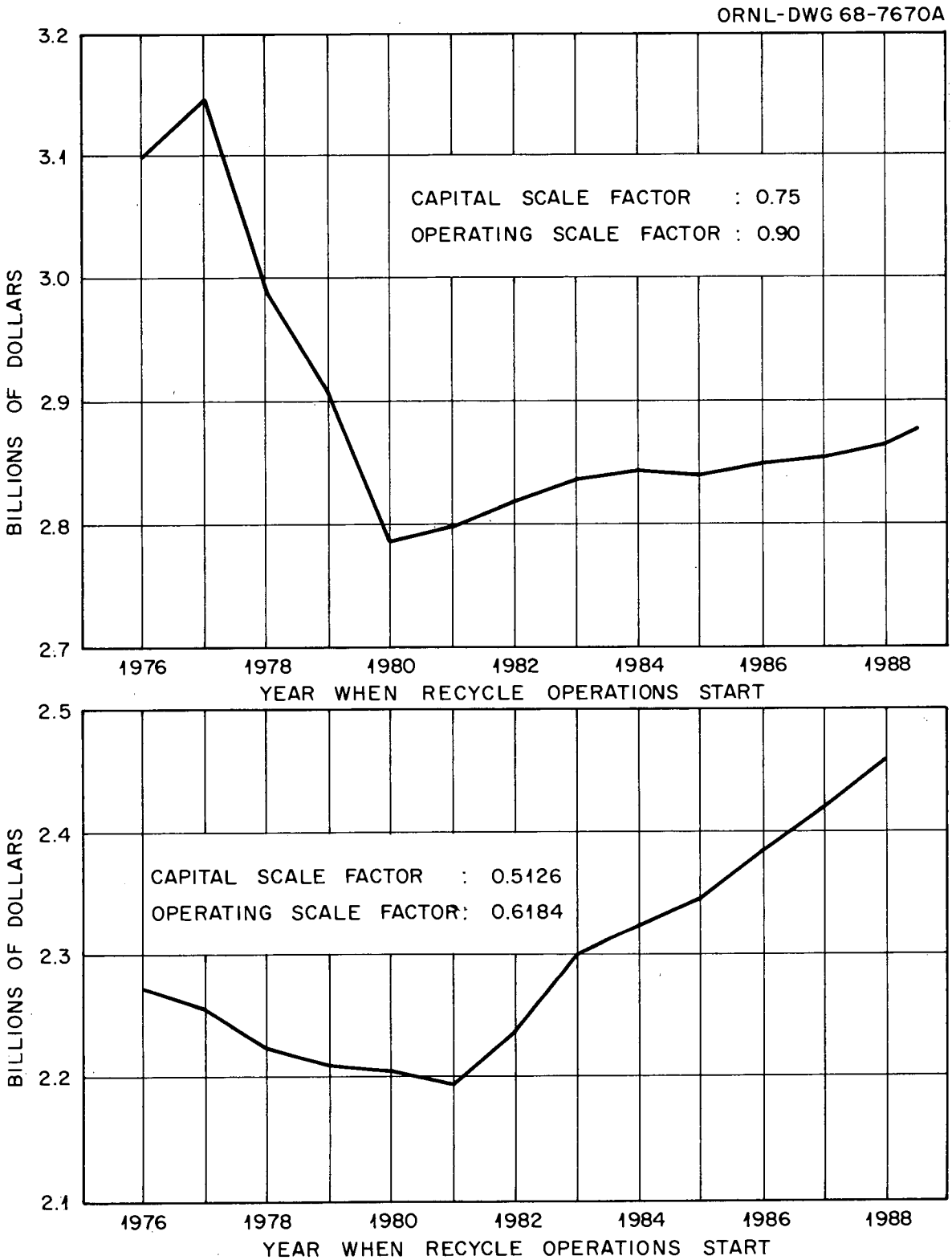

Fig. 26. Effect of Increased Scale Factor.

engineering-scale equipment can be designed. The technology for the manufacture of particles that meet the required burnups and conditions of HTGR's is well known. However, requirements for bonded sticks and methods for formation of fuel sticks are not yet fully developed.

What is required now is a design and engineering effort aimed at scaleup of the known processes, some additional research to develop satisfactory process steps in several areas, and construction of equipment and systems for demonstration of HTGR recycle technology. Shielded facilities, although they are generally not equipped, exist for carrying out the necessary pilot-scale development.

Through economic analyses of growing HTGR economies, we have shown that a central recycle plant is optimally scheduled if it is brought on line in 1981. This conclusion was reached through the analysis of a number of different HTGR growth rates; assuming in each case that the first 1000-Mw (electrical) reactor would be placed on line to produce power in 1974. Another conclusion of the economic study is that both reprocessing steps and refabrication steps benefit substantially from the effect of increasing plant size.

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