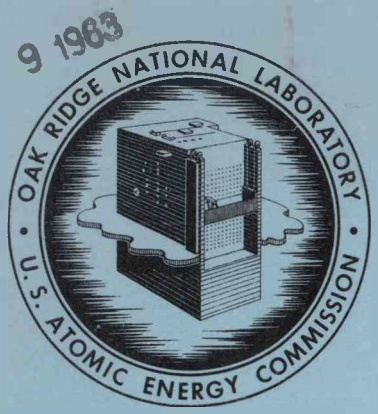

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PARTIALLY ENRICHED FUEL CYCLES

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ABSTRACT

Most of the power reactors built or proposed for construction in the United States use slightly enriched uranium for fuel of less than 5 w/o U^{235} initial concentration. The most common fuel type employs UO_2 in Zircaloy or stainless steel tubes. With a few notable exceptions--the experimental reactors such as the Sodium Graphite Reactor, the Experimental Gas-Cooled Reactor, the Peach Bottom Reactor which is now being built, and the Fermi Reactor--all reactors are pressurized, light water-cooled, and moderated.

The technology for fuel manufacture and reprocessing now exists, but because of the small discharge rate of spent fuel and the relatively small requirement for fresh fuel, a coordinated fuel recycle complex does not exist. Fuel fabrication plants are small batch plants which operate far below optimum economic size, frequently on a part-time basis. No reprocessing of power reactor fuels has been done either in government-owned or private facilities. One small reprocessing plant which will derive about 50 per cent of its operating cost from AEC-supplied base load is now under construction in the State of New York. Designed for a nominal production capacity of approximately one metric tonne per day of less than five per cent enriched uranium, it, too, is below optimum economic size but may, depending upon its actual performance and costs, provide reprocessing at a cost of $32 to $35/kg of uranium.

In spite of these important economic handicaps, the emerging small-scale, somewhat-fragmented nuclear fuel cycle industry in the United States probably will provide fuel for about 1.75 mills/kwhr to 2.0 mills/kwhr electricity for the more advanced stations, such as the proposed Bodega Bay Reactor, assuming: private purchase of $13/kg natural uranium; "toll" enrichment in government-owned diffusion plants at $30/kg separative work charge; and a plutonium buy-back price by the government maintained at $8/gram of plutonium, or above. Thus, fuel cost is about 16 to 20 cents

per million Btu. The Bodega Bay Reactor will produce power for an estimated cost of about 5.5 to 6.2 mills/kwhr, a cost which is competitive with conventional plants in those high power-cost regions of the United States which constitute possibly 15 per cent of the electrical power generating capacity in the U.S.A. Each 0.1 mill/kwhr electricity reduction in cost will add possibly 5 per cent to the competitive market for nuclear energy.

That much lower fuel cycle costs are possible with large cycle capacity is both a supportable fact and a heartening promise. For reactor systems which are basically fueled with uranium enriched in a diffusion plant, over-all cycle costs of about 0.8 mills/kwhr electricity are possible in an essentially single-purpose cycle at a capacity of ten tonnes of uranium per day.

To achieve a competitive status with the highest performance conventional power stations in the United States, nuclear energy plants must produce electricity for about 4.0 mills/kwhr. This appears to be possible with pressurized water reactors in sizes larger than the proposed Bodega Bay Reactor, as indicated in an as-yet-unpublished study of very large reactor systems (and high through-put fuel cycles) in the size range from 1200 to 8250 megawatt thermal. Further, fuel cycle studies using natural uranium as the fuel base for large heavy water-moderated reactors have reconfirmed the well known fuel cycle advantage of natural uranium.

The lowest fuel cycle costs for converter reactors can be achieved by use of natural uranium fuels in heavy water-moderated reactors of very large size. Natural uranium fuel cycles operated at cycle capacities of ten tons per day and larger offer over-all fuel costs that are disappearingly small, about 0.05 mill/kwhr thermal, or 0.18 mill/kwhr electricity at 28 per cent thermal efficiency. These costs assume 14 per cent per year charge on capital, a 5.5 per cent charge on consumable inventories, burn-up of 7000 Mwd/tonne of uranium and a plutonium credit of $6.70/gram. If plutonium is not recovered but discarded, the costs are approximately 0.9 mills/kwhr electricity. Plutonium can be recovered and recycled with depleted uranium to the same reactor system for approximately 20 per cent increase in fuel cycle costs over the natural uranium case. This natural uranium cycle is probably a factor of five less costly than the least

expensive partially enriched uranium case if plutonium (where initial U^{235} is 1.5 w/o or higher) is recovered and sold; a factor of two less than the least expensive enriched case if plutonium is not recovered (the throwaway case), particularly in fuel cycles of less than several tonnes per day capacity; and is attractive economically if the plutonium produced is recycled with depleted uranium.

1.0 CURRENT STATUS OF PARTIALLY ENRICHED THERMAL CONVERTERS

It is my purpose to discuss partially enriched uranium fuel cycles which I shall define to mean systems employing less than 5 w/o U^{235} in the uranium fuel and including natural uranium. Before discussing the fuel cycles and the net costs of nuclear fuel, it is important to establish the current technical and competitive economic status of such natural and partially enriched reactors and their longer-term role in a nuclear power economy which ultimately must be breeder-based.

1.1 Role of Converter Reactors

The best statement of the importance of converter reactors has been made in the recent report "Civilian Nuclear Power--A Report to the President – 1962."(1) Therein, the United States Atomic Energy Commission has clearly and objectively stated several conclusions and established certain programmatic objectives that will affect both the relatively short-term and the long-range direction of U.S. nuclear power. Since the wording of this report is precise, I have chosen to quote directly, with the addition of a few parenthetical remarks and in a sequence that is different from that of the original report.

These clear statements indicate both a logical role for converters as well as long-range direction for the development of the full potential of nuclear power:

> "...the development and exploitation of nuclear-electric power is clearly in the short- and the long-term national interest and should be vigorously pursued...."

because:

> "...nuclear energy can and should make an important and, eventually, vital contribution toward meeting our long-term energy requirements."
>
> (a) "...we will exhaust our readily available, low cost supplies of fossil fuel in 75-100 years and our total supplies in 150-200 years ...." (total estimated between 30 and 150 Q)
>
> (b) "...the fossil fuels have special advantages that are unmatched... by substitutes...for mobile power...essential metallurgical applications...(and) represent a priceless heritage of complex molecular substances, the possible uses for which are only beginning to be realized."

since:

> "Experience has shown that nuclear electric power is readily achieved technically..." (now in enriched uranium converters).

"Nuclear power is believed to be on or near competitiveness with conventional power for large plants in areas of the country where fossil fuel costs are high. Further cost reductions are definitely in sight....

"Certain classes of power reactors, notably water-cooled converters, are now on the threshold of economic competitiveness...in large installations in high fossil fuel cost areas of the country. In our opinion, economic power is so near at hand that only a modest efforst is required to initiate its appreciable use...."

For the long-term aspects, it is stated:

"...breeder reactors have not yet reached an economically useful stage of development. Even when they do, they will not initially, at least, make new material fast enough to provide the fuel required for new plants.... It will be necessary to fuel some portion of the installations with U^{235} until such time as improved breeding gains and reduction in relative rate of power growth enable breeders to be self-sufficient. ...in the transition stage (from converters to breeders), which will last for many decades, fast breeders...will probably be augmented by thermal converters burning U^{235} and producing plutonium at a slower rate. This need will enhance the desirability of advanced converters... because it is important that the combination of breeders and converters reaches an over-all net breeding capability... while relatively cheap (nuclear) fuel supplies are still available."

Thus, these program recommendations follow:

"1. The demonstration of economic nuclear power by assuring the construction of plants incorporating the presently most competitive reactor types." (These are all converters, most of which are fueled with partially enriched U^{235}.)

"2. The development of improved converter and, later, breeder reactors to convert the fertile isotopes to fissionable ones, thus making available the full potential of the nuclear fuels."

From the foregoing, it is apparent that opinion in the United States is that the converter reactor is likely to be the front-runner during the period of initial expansion into nuclear power, possibly the standard of comparison (because of anticipated improvements in performance and nuclear efficiency) during at least a portion of ramp period of nuclear power growth, and will play an indispensable role in the production of the necessary stocks of plutonium and uranium-233 for the initiation of a self-sufficient economy of fast and thermal breeders.

1.2 Present Status of Nuclear Power Reactor Systems

There are now six sizeable reactors in the United States operating with partially enriched uranium as fuel; six more reactors will be

completed in 1963. Most of these are water-cooled converters (0.5 to 0.7 conversion ratio) of different types operating with thermal neutron spectra, and most now produce saturated steam. Thermal reactors of higher performance, with respect to steam temperature and more efficient fuel utilization, are also being investigated; the Sodium Graphite Reactor, High Temperature Gas-Cooled Reactor, Experimental Gas-Cooled Reactor, Molten Salt Reactor, the Spectral Shift Reactor, and the proposed Westinghouse Seed-Blanket Reactor are examples. All are thermal reactors or near-thermal; at least three of these will use the thorium blanket and may ultimately convert to a uranium-233--thorium cycle after a period of operation using partially enriched U^{235} as fuel. The reactors built or proposed are listed in Table 1.

It has been estimated[2] that a 500 megawatt nuclear electric power plant could be placed in service by 1966 which would produce power at a cost of about 6.2 mills/kwhr initially, and which would compete with the best fossil fuel plant using coal costing 39 cents per million Btu. This cost of nuclear energy will be able to compete easily in the high cost fuel areas of the United States and could capture 15 to 20 per cent of the power market.

1.3 Economic Prospects for Nuclear Power and Growth

Table 2[3] projects the competitive position of nuclear power to be expected as the partially enriched converter technology improves. By 1980, nuclear power should be competitive with coal in most sections of the United States. By then, too, breeder technology should be sufficiently advanced to permit the gradual emergence of breeders as principal producers of nuclear energy. That they do ultimately become important contributors is inevitable and necessary for the conservation of fissionable material, but they will not take over completely from the natural or partially enriched converters until, in aggregate, they are capable of producing sufficient plutonium or uranium-233 to supply the expanding power economy.

Table 1. U.S. Nuclear Power Reactors over 10 Mw_e Operable, Being Built or Proposed as of 1963

		Nuclear Mw_e		% U Enrichment	Fuel -- Cladding
Metal Clad - Ceramic Core (4028 Mw_e)					
Shippingport Blanket	Operating	~30		0.7	UO_2 -- Zircaloy
Yankee	Operating	161		3.4	UO_2 -- Stainless Steel
Indian Point	Operating	151		93	ThO_2-UO_2 -- Stainless Steel[a]
Dresden	Operating	208		1.5	UO_2 -- Zircaloy[a]
Big Rock Point	Operating	48		3.2	UO_2 -- Stainless Steel
Humboldt Bay	Operating	48.5		2.6	UO_2 -- Stainless Steel
			647		
Carolinas-Virginia	In 1963	~15		1.5,2.0	UO_2 -- Zircaloy
Elk River	In 1963	15		93	ThO_2-UO_2 -- Stainless Steel
Pathfinder Boiler	In 1963	~44		2.2	UO_2 -- Zircaloy
Bonus	In 1963	16.3		0.7,2.4,3.25	UO_2 -- Zircaloy and Inconel
LaCrosse Boiling H_2O	Construction	50		3.4	UO_2 -- Stainless Steel
			140		
Niagara Mohawk	Proposed	500		~3.5	UO_2 -- Stainless Steel
Jersey Central	Proposed	500		~3.5	UO_2 -- Stainless Steel
Bodega Bay	Proposed	313		2.02	UO_2 -- Stainless Steel
Malibu	Proposed	450		3.8	UO_2 -- Stainless Steel
Camp Pendleton	Proposed	355		3.2,3.6,4.0	UO_2 -- Stainless Steel
Haddam Neck	Proposed	463		3.2,3.6,4.0	UO_2 -- Stainless Steel
Ravenswood	Proposed	660		3.5	UO_2 -- Stainless Steel
			3241		
Other Systems (1047 Mw_e)					
Shippingport Seed	Operating	~30		93	U-Zr -- Zircaloy
Hallam	Operating	75		3.6	U-Mo -- Stainless Steel (Na cooled)
			105		
Enrico Fermi	Loading	60.9		25.6,0.4	U-Mo core (Fast Breeder)
Piqua	In 1963	11.4		1.94	U-Mo-Al -- Aluminum (Organic cooled)
Pathfinder Superheater	In 1963	~15		93	UO_2-SS -- Stainless Steel
EBR-2	In 1963	16.5		46,0.2	U-Fs core (Fast Breeder)
Experimental Gas Cooled	Construction	21.9		2.46	UO_2 -- Stainless Steel (He cooled)
Peach Bottom	Construction	40		93	$(Th,U)C_2$ in graphite (He cooled)
New Production Reactor (Hanford)	Construction	776		0.94	U -- Zircaloy[b]
			942		
Total			5075		

[a] To convert to UO_2 clad in Stainless Steel for Core 2.

[b] To convert to UO_2 clad in Zircaloy in future.

Table 2. Projected Competitive Position of a 500 Mw electricity Nuclear Power Plant

Year in Service	Energy Cost mills/kwhr electricity Initial	After 5 years	Nuclear Plant Competitive with Coal Costing, ¢/10^6 Btu Initial	After 5 years
1966	6.2	5.6	39	33
1970	5.3	4.8	31	26
1975	4.5	4.2	24	20
1980	3.8	3.6	19	17

Assumptions:

(1) Charge rate on plant investment 14%/year.

(2) AEC-use charges on fuel (4-3/4% per year); or private ownership of fuel with \$5/lb U_3O_8, toll enrichment at \$30/kg uranium separative work, \$8/gram plutonium credit, 10% annual charge on fuel inventory.

(3) Fuel cycle charges based on small plants of less than optimum economic size.

(4) Capital cost of coal plant in 1966, \$129/kw; in 1980, \$110/kw. Heat rates are: 1966, 8500 Btu/kwhr; 1980, 7500 Btu/kwhr.

(5) 80% plant capacity factors.

These projected costs should provide the incentive to create a growth pattern for the U.S. nuclear power industry which approximates that given in Table 3.[4]

Table 3. Projected United States Electrical Generating Capacity

Year	Nuclear Capacity Megawatts	Total Capacity Megawatts	% Nuclear
1965	1,350	239,850	0.6
1970	5,000	329,100	1.5
1975	16,000	438,700	3.6
1980*	40,000	565,500	7.0
2000	734,000	1,651,800	45.0

*Breeders achieve competitive position

From the standpoint of fuel cycle capacity, the requirements for fuel reprocessing and fuel fabrication are below what appears to be economic plant size and, as a consequence, fuel costs will remain unnecessarily high until about 1980, or until essentially a single unit fuel cycle can be supported at a capacity of six to ten tonnes of uranium per day with fuel exposures in the range of 16 to 25 Mwd/kg uranium. If the growth rate predicted in Table 3 does occur, then the following fuel cycle capacity requirements will evolve (Table 4).

Table 4. Fuel Cycle Capacity Requirements for the Period 1965 to 2000

Year	Nuclear Electric Megawatts	Thermal Efficiency %	Assumed Fuel Exposure[5] Mwd/kg U	Required Fuel Cycle Capacity Tonnes U/day*
1966	1,350	32	17	0.3
1970	5,000	32	20	0.9
1975	16,000	32	23.5	2.5
1980	40,000	32	25	5.9
2000	734,000	37**	40**	58

*Assumed 85 per cent on-stream time for fuel cycle plants

**Guess of average performance in a mixed converter--breeder economy

1.4 The Effect of Scale on Power Costs in Slightly Enriched Reactor Systems

The effect of increasing nuclear reactor and fuel cycle facility size is to decrease the cost of power. This is illustrated for light water, partially enriched reactors by data from two sources, the Report to the President – 1962, which provides estimates for individual reactors to 500 Mw electricity in size; and the Bechtel report, "Large Reactor Study for Sea Water Distillation,"[6] which provides a summary of rough estimates for individual reactors to 3000 Mw electricity (net), and of reactor stations to 9000 Mw electricity (net) in size, producing power only. With near-current technology, using capital and inventory annual charge rates that are representative of private ownership, reactors of 1000 Mw electric can be built that would produce power for less than

5 mills/kwhr. If municipal or government financing can be justified, and there is a case for this even in the United States, power can be produced from partially enriched systems by 1000 Mw electricity reactors for less than 3.5 mills/kwhr. The over-all effects of increasing reactor (and reactor station) and fuel cycle size are shown in Table 5.

Table 5. Effect of Scale on Cost of Power for Partially Enriched Converters, Power Only

Plant Size Mw electricity (net)	Cost of Electricity, mills/kwhr Current Technology 14% capital	1980 Technology 14% capital	
From the Presidential Report Appendices:			
200	6.9		
300	6.1		
400	5.8		
500	5.6	3.6	
From the Bechtel Study:	14% capital	7% capital	
470 (1 reactor)	5.9	4.0	
940 (2 reactors)	5.6	3.8	
1410 (3 reactors)	5.4	3.7	
1002 (1 reactor)	5.0	3.5	
1140 (1 reactor)	4.8	3.0	graphite moderated
9000 (3 reactors)	3.4	2.2	advanced converter

Notes: (1) In the Presidential Report all reactors are light water-cooled and moderated. Private ownership of fuel at 10% annual inventory charge; plutonium credit $8/gram.

(2) In Bechtel Study all reactors referred to are light water-cooled, except as noted, and are essentially near-current technology except for the advanced converters. Private ownership of fuel at 7.7% annual inventory charge for 14% capital charge rate, at 5% inventory charge rate for 7% capital charge rate. Plutonium value varies from $8.50 to $9.10 per gram.

1.5 Natural Uranium-Heavy Water Converters and Fuel Cycles as Competitors for Partially Enriched Converters

The Report to the President gave only scant attention to converter reactor systems using natural uranium and heavy water. In spite of low burn-up and inventory costs and other recycle advantages, natural uranium systems have not yet become an important part of the developing nuclear power program in the United States. Outside of the United States, the case for natural uranium--heavy water reactors has been ably and consistently championed by Dr. W. B. Lewis within the framework of the Canadian nuclear energy program.

In the United States, the du Pont Company has studied natural uranium--heavy water power reactor systems, using their extensive experience with the large production reactors at Savannah River as a basis.(7) Early in the study, du Pont observed that lease charges on heavy water inventory, plutonium buy-back price, and reprocessing charges set by the government resulted in an inadvertent but unfair bias against natural uranium--heavy water systems.

Other factors than government-established pricing policies have influenced our thinking about the relative economic attractiveness of natural uranium reactors. Probably the most important is that the size of the reactors studied was too small to uncover the economic advantage of the natural uranium--heavy water systems in large sizes. The capital cost of partially enriched--light water cooled reactors is lower for reactor sizes of less than about 750 Mw electricity. However, in sizes where it is impossible to build pressure vessels for pressurized light water reactors and pressure tubes must be used, the heavy water moderated reactor competes with respect to capital costs.

The cost advantage of the natural uranium fuel cycle, although evident from production experience, has never really been brought effectively into the power cost arguments. In large capacity cycles (between 10 to 30 tonnes of uranium per day) with reasonable values for plutonium, such as $6 to $7 per gram, the net fuel costs approach zero. This fact, together with the knowledge that large heavy water moderated, natural uranium reactors are competitive in capital and operating cost with partially enriched reactors of the same size, can have a profound effect on the cost of both power and plutonium required to initiate a

breeder-based economy. Very large natural uranium reactors and recycles probably can produce power for 1.5 to 3.0 mills/kwhr electricity, depending upon the method of financing.

There are two factors affecting this argument for natural uranium cycles that have not yet been adequately evaluated. First, the cost of enriching uranium to low-enhancement of the U^{235} concentration has not been optimized. The toll enrichment charges of $38/kg uranium for separative work can be reduced. It is quite possible that optimized cascades, using the best possible operating characteristics, will produce very slightly enriched uranium at a cost low enough to give lower net fuel costs. The incremental cost of enrichment to less than 1 per cent U^{235} possibly can be off-set by the extension of nuclear fuel lifetime and by increasing plutonium yield.

The other important variable is the value of plutonium. Very large reactors, such as those proposed by Hammond[8][9] for desalination would produce large quantitites of plutonium, probably sufficient to glut the market. If there were no customers, plutonium could be recycled to a heavy water reactor, using either depleted or natural uranium as the make-up fertile material. Preliminary calculations indicate that the net fuel cost may be lower with recycled plutonium than natural uranium purchased at $20/kg. Thus plutonium probably can be used economically to enrich heavy water reactors if a market cannot be found which will yield more than about $3/gram.

Hammond's observations on the effect of scale on reactor costs have sparked a very recent re-evaluation of both natural uranium--heavy water and partially enriched light water reactors in sizes in excess of 500 Mw electricity. Factored into these evaluations have been fuel cycle costs that are obtainable from large capacity recycle plants.

The first of these studies was for a D_2O moderated with boiling light water cooled reactor, producing steam at 600 psi. The natural uranium fuel elements were of UO_2 clad in Zircaloy. A reactor station of 25,000 thermal megawatts consisting of three reactors of 8330 Mw each was estimated. The net electrical generating capacity was about 7500 Mw and fuel exposure was assumed to be 7000 Mwd/tonne uranium. It was further assumed that fuel from this complex would be fabricated and reprocessed in fuel cycle facilities of 10 tonnes/day capacity, sufficient to process

three 25,000 Mw thermal reactor stations. At an over-all thermal efficiency of 31 per cent, power costs were between 1.6 and 2.6 mills per kwhr electricity, as shown in Table 6.(10)

Table 6. Cost of Electricity from 25,000 Mw Thermal Natural U-D_2O Stations

Fixed Charges on Reactor and Fuel Plant	Cycle	Cost mills/kwhr electricity
7.7%	Pu credit (6.70/gram)	1.6
14.0%	Pu credit (6.70/gram)	2.6
7.7%	Throwaway	2.0
14.0%	Throwaway	3.0

The fuel cycle costs are given in Table 7; the capital charge rates used were 7.7 per cent (14%); inventory charge rate, 5.5 per cent.

Table 7. Fuel Cycle Cost at 10 tonnes/day Natural Uranium

	With Plutonium at $6.70/gram	Throwaway Cycle
Heat cost, cents/10^6 Btu	0.9 (1.40)	4.3 (4.5)
Mills/kwhr	0.09 (0.15)	0.47 (0.52)

This study has been followed by the Large Reactor Study by Bechtel, mentioned previously. The heavy water moderated reactors studied in this report, based on reactor design by du Pont and fuel cycle costs from the Oak Ridge National Laboratory, were organic cooled. For fuel, natural uranium as UC, clad in Zircaloy, was used. Three cases were studied. The results of this study are shown in Table 8.

Table 8. Results of Bechtel Large Reactor Study for Natural U-D_2O Systems (for 7% and 14% capital depreciation)

Reactor Net Mw electricity	Fabrication Capacity Tonnes/day	Reprocessing Capacity Tonnes/day	Power Cost mills/kwhr electricity 7%	14%
1110 (1 reactor)	less than one	1	3.01	4.84
2770 (1 reactor)	1	2	2.18	3.50
8300 (3 reactors)	10	30	1.67	2.96

Thus the natural uranium systems, using D_2O as a moderator with either water or organic cooling, appear surely to be competitive with the most advanced partially enriched uranium converter (and I limit the meaning of "partially enriched" to mean above 1.5 per cent U^{235} and exclude plutonium recycle, pending further investigation). Taking costs for partially enriched systems from Table 5 (the Bechtel Study), the following comparison can be made between natural and partially enriched systems:

Table 9. Comparison of Power Costs from Natural and Partially Enriched Uranium Converters at Large Scale mills/kwhr electricity at 7% capital charge rates

Approximate Net Mw electricity	Partially Enriched	Natural U-D_2O Water Cooled	Organic Cooled
1000-1100 (1 reactor)	3.5		3.0
8300-9000 (3 reactors)	2.1	1.6	1.7

All reactors receive plutonium credit.

2.0 Costs of Recycle of Natural and Partially Enriched Reactors

The cost elements which make up fuel cycle costs are: (1) the net cost of feed; (2) inventory costs in reactor and fuel cycle; (3) conversion costs of feed to desired fuel form; (4) fuel element fabrication; (5) fuel shipping charges; and (6) reprocessing.

The most important factor in determining the cost of recycle is the

scale at which the recycle is operated. At a capacity representative of a mature nuclear economy (recycle plants above six tonnes per day in size) over-all fuel costs, exclusive of net burn-up and inventory costs, can be insignificant. Or in other words, conversion costs of raw feed, fuel element fabrication, and reprocessing costs (now costed at a small scale) can be reduced by factors of four to ten by increasing capacity. Net burn-up costs for partially enriched uranium already have the advantage of large scale enriched UF_6 production, since costs for enrichment are now supported by large production for other purposes. For natural uranium costs, obviously the same comments are valid; the well known effect of mass production in lowering costs is already discounted by the size of the ore industry required to support the diffusion cascades and plutonium production from natural uranium.

2.1 Current or Near-current Costs for Partially Enriched Reactor Cycles

The cost of fuel recycle with current or near-current technology for partially enriched converters is sufficiently low to produce 5.5 to 6.0 mills/kwhr electricity. Of this total cost, the fuel cycle costs will contribute 1.75 to 2.0 mills/kwhr, assuming that fuel element fabrication is done in plants of capacity less than one ton per day of uranium in fabricated fuel and that reprocessing is done in a plant with a nominal capacity of one tonne per day.

For a light water-cooled partially enriched reactor of characteristics, see Table 10; current or near-current fuel cycle costs are estimated in Table 11.

Table 10. Characteristics of 1000 Mw (thermal) Partially Enriched Converter

Net megawatts, electric	303.5
Thermal efficiency, %	30.2
Fuel loading, kg U	70,800
Initial enrichment, w/o U^{235}	2.02
Discharge enrichment, w/o U^{235}	0.82
Pu content, spent fuel, grams/kg U	6.6
Average fuel exposure, Mwd/kg U	16.65
Time in reactor at 80% LF (discharged in three batches)	4 years
Cladding	Stainless Steel

Table 11. Fuel Cycle Costs for 1000 Mw thermal Partially Enriched Converter at 1 ton U (or less)/day

	$/kg U	mills/kwhr electricity
Gross burn-up charge ($5/lb U_3O_8 and "toll" enrichment)	96.17	0.80
Less $8/gram Pu credit	52.80	0.44
Net Burn-up Cost	43.37	0.36
Use charges at 4.75% for 4.8-yr cycle	17.42	0.15
FEED COSTS	60.79	0.51
Conversion of $UO_2(NO_3)_2$ to UF_6	5.60	0.05
Fuel fabrication, including UF_6 to UO_2	102.00	0.85
FABRICATION	107.60	0.90
Reprocessing and waste disposal to tanks	32.01	0.27
Fuel shipment	10.00	0.08
PROCESSING	42.01	0.35
TOTAL	210.40	1.75

If this reactor fuel could be processed in fuel cycle facilities with a capacity of ten tonnes of uranium per day, the over-all fuel cost would be approximately 1.05 mills/kwhr electricity if our method of scaling is about correct. The break-down of these costs is shown in Table 12.

Table 12. Possible Current Light Water Slightly Enriched Fuel Cycle If Done in 10 ton/day Cycle

	$/kg U	
Burnup and Inventory (53%)		
Burnup without Pu credit assuming "toll" enrichment U_3O_8 at $5.00/lb (2.02% U^{235} charged, 0.82% U^{235} discharged)	96.20	
Inventory charges	17.40	
	113.60	
Less Pu credit at $8.00/gm, 6.6 gm/kg U	52.80	
		60.80
Fabrication (33%)		
Conversion UF_6 to UO_2	0.95	
Fabrication	34.50	
Rejects at 5%	1.80	
		37.25
Reprocessing, Shipment, Waste Disposal (14%)		
Reprocessing and waste disposal	5.95	
Losses, 0.25% Pu, 1% U	0.35	
Fuel and Product Shipment	10.00	
		16.30
		114.35
10% Contingency		11.45
Total		125.80
Cost of heat at 16,550 Mwd/tonne, thermal	0.317 mills/kwh	
Cost of electricity at 30.1% efficiency	1.05 mills/kwh	
Cost of heat, cents/10^6 BTU	9.3	

We have ventured to prepare over-all curves relating plant capacity to the three capacity-sensitive steps in the fuel cycle--conversion of UF_6 to UO_2, fuel fabrication, and reprocessing. The estimates were prepared by J. W. Ullmann, F. E. Harrington, A. L. Lotts, and D. A. Douglas of the Oak Ridge National Laboratory as a part of the Bechtel Large Reactor Study. These curves are applicable to partially enriched uranium fuel, using pelletized UO_2 in stainless steel or Zircaloy tubes about 0.4 inch in diameter. The reprocessing plant is based on solvent extraction separations, using a shear--leach head-end feed preparation.

Figure 1 gives cost of the conversion of UF_6 to UO_2. Figure 2 shows the reprocessing costs, including waste disposal to tanks with 20-year capacity for high-level wastes. Figure 3 provides an approximate capacity reduction factor for enrichments higher than 3 per cent, to be used with the reprocessing plant. Figure 4 gives the cost of fabrication of fuel elements fabricated with UO_2 pellets clad with either stainless steel or Zircaloy.

2.2 Burn-up Cost of Partially Enriched Uranium Cycles of Advanced Converter Reactors

An estimate of the performance of advanced converter reactors using partially enriched uranium was offered in the Report to the President. No data were given on the type or design of fuel elements so that a complete cost estimate of fuel cycles cannot be made. However, burn-up and inventory charges, which are about 50 per cent of the fuel cycle cost at a capacity of 10 tons per day, can be calculated. Assuming "toll enrichment" at \$30/kg separative work, \$5/lb U_3O_8 feed, \$2.70 conversion charges, \$6.70/gram plutonium credit, and an inventory charge rate of 5.5%/year, and reactor performance as shown in Table 13, and burn-up and inventory costs as shown in Table 14, were calculated.

For this estimate we have used \$6.70 rather than \$8 per gram for the value of plutonium to make a consisten comparison with the natural uranium--heavy water case in the next section. The actual value of plutonium will be dependent upon the type of reactor in which it is used. It could be anywhere from \$3/gram for recycle to a heavy water moderated reactor to about \$10/gram for fast breeder fuel.

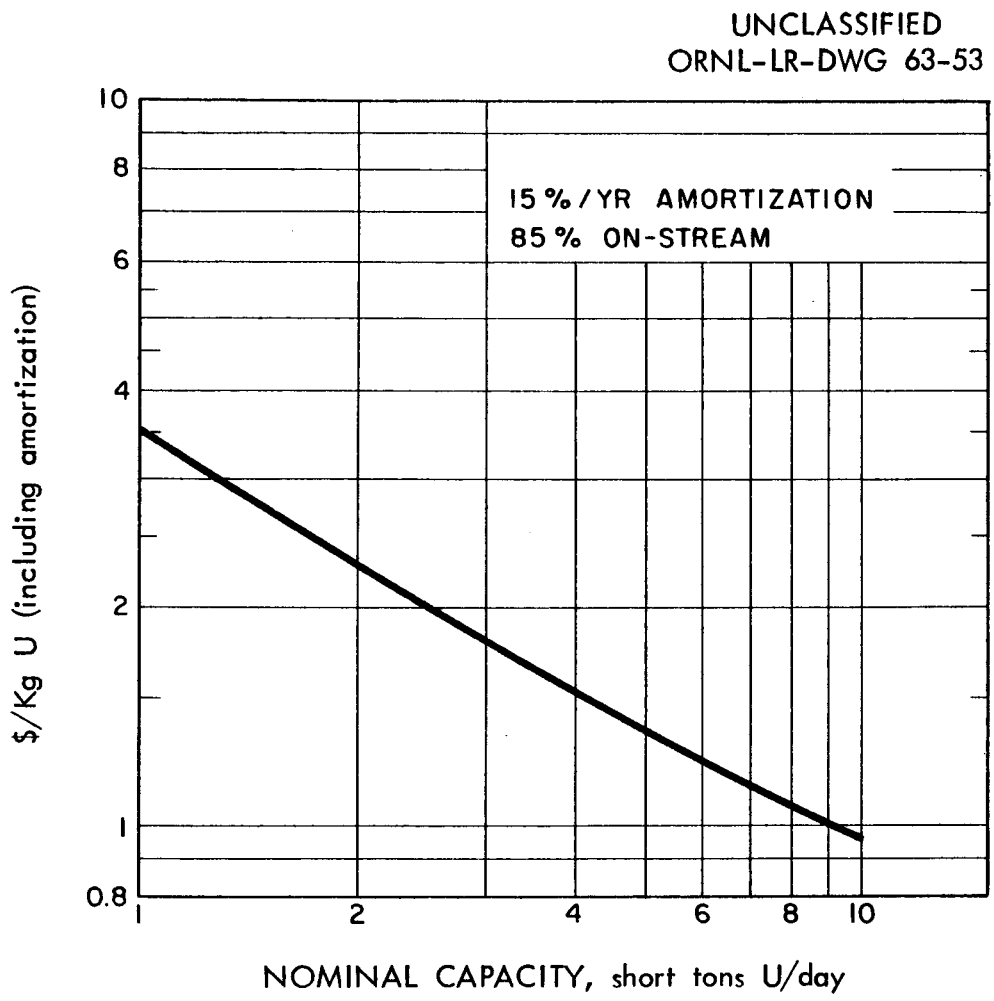


Fig. 1. Unit Conversion Cost, UF_6 to UO_2 Base Case as a Function of Capacity.

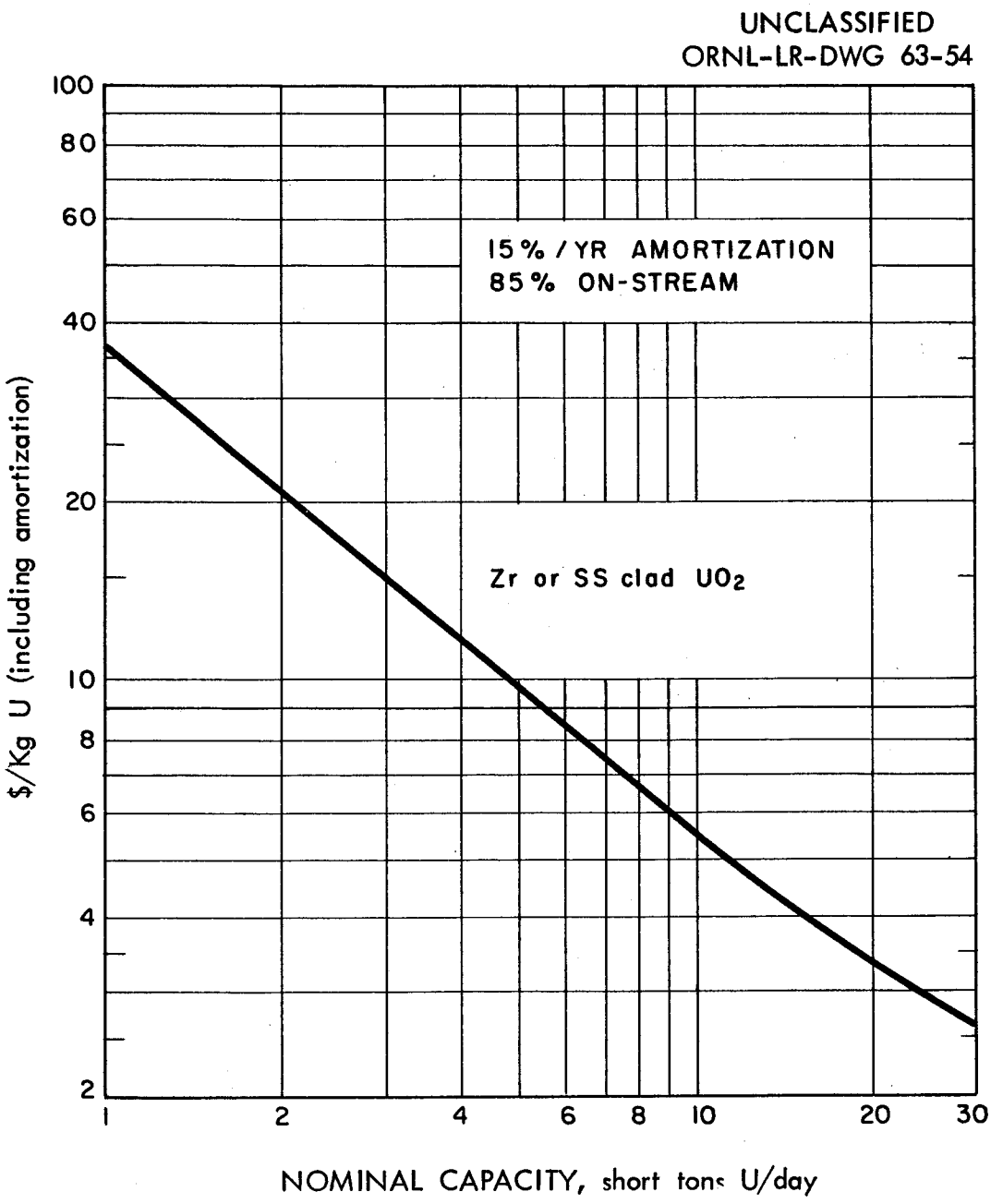


Fig. 2. Unit Spent Fuel Processing Cost Base Case as a Function of Capacity.

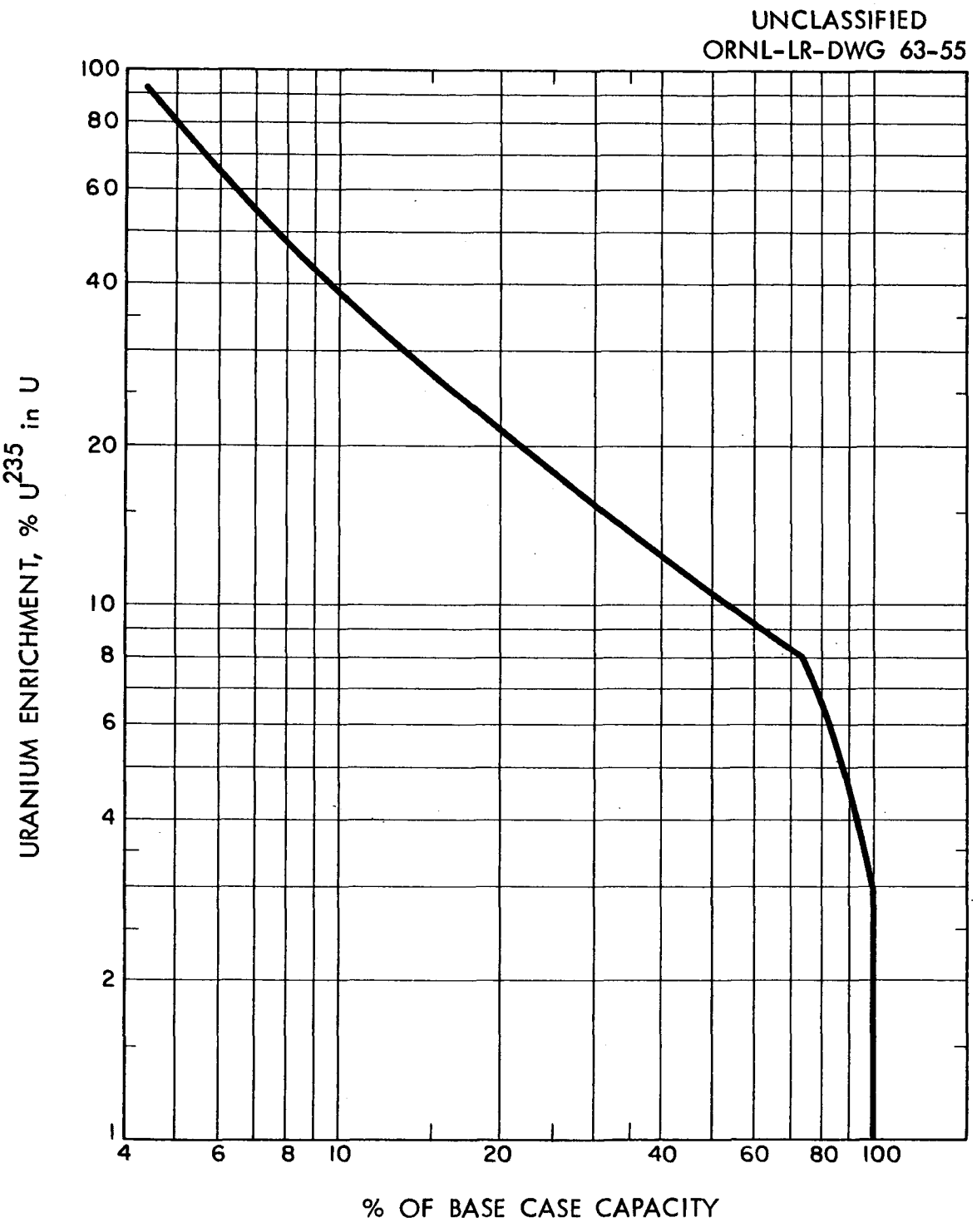


Fig. 3. Percentage of Base Case Capacity as a Function of Enrichment.

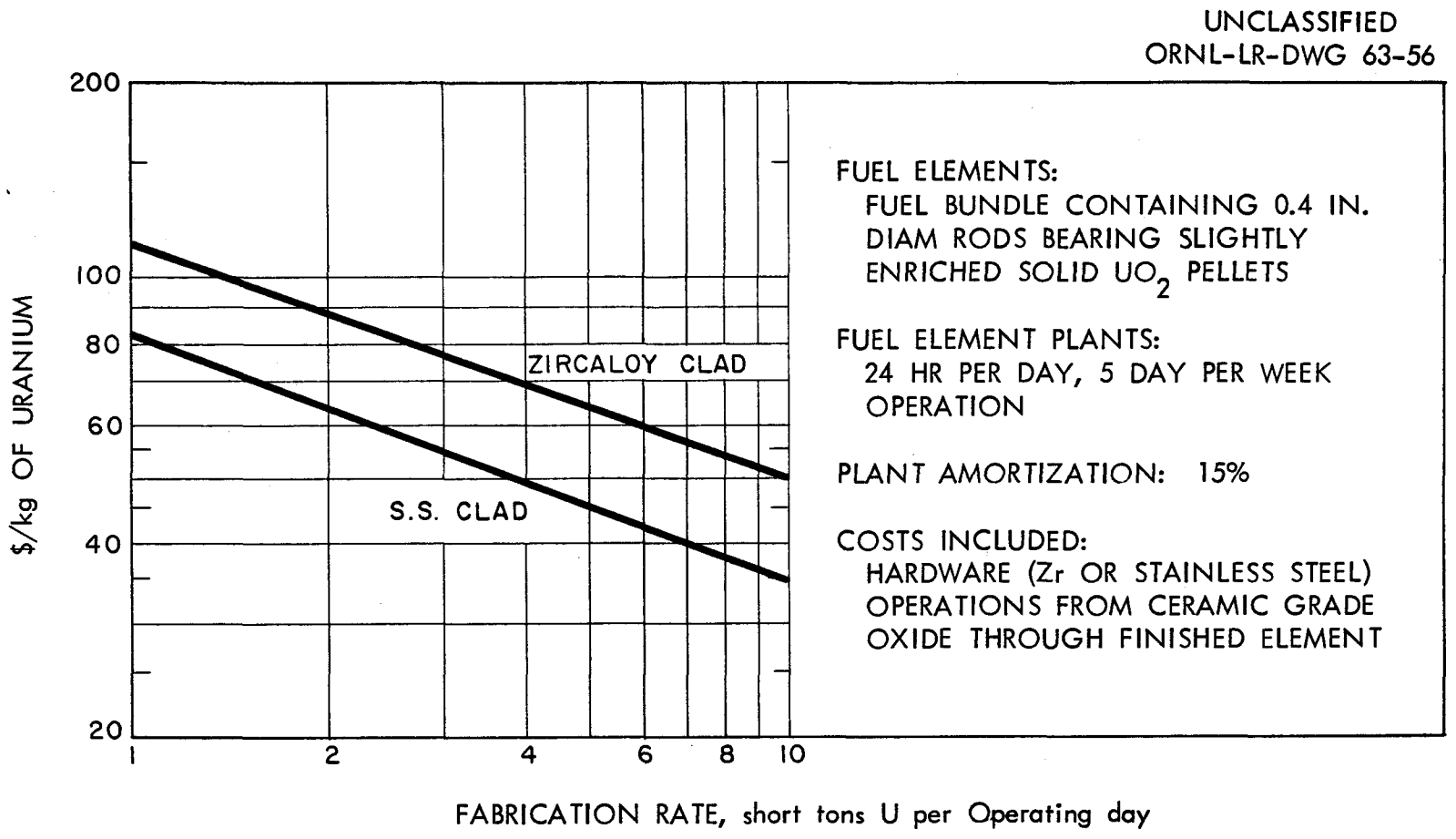


Fig. 4 Effect of Plant Size on Cost of Fabrication of Fuel Bundles Bearing Slightly Enriched UO_2

Table 13. Predicted Performance of Partially Enriched Uranium Converters of About 500 Mw electricity, 1980

	1980
Cycle time in reactor at 80% load factor, years	3.1
Assumed recycle time, years	0.9
Over-all thermal efficiency, %	40.

Table 14. Estimated Burn-up and Inventory Costs for Advanced Partially Enriched Uranium Converters, 1980

	$/kg uranium
Initial cost of uranium	207.82
Discharge value of uranium	65.97
	147.55
Pu value at $6.70/gram, credit	50.10
NET BURN-UP COST	97.45*
Average value of uranium in reactor (assumed arithmatic average)	136.90
Average value of plutonium in reactor	25.13
Average value of uranium in fabrication	207.82
Average value of U and Pu in reprocessing and cooling	116.07
INVENTORY CHARGES AT 5.5%/YR	34.26*
TOTAL BURN-UP AND INVENTORY	131.71**
Mills/kwhr electricity at 40% efficiency	0.55

2.3 Fuel Cycle Costs for Natural Uranium Cycles

For natural uranium, the over-all fuel cycle cost can approach zero if there is a market for plutonium selling for at least six dollars per gram.

At any scale, from the standpoint of recycle costs only, the natural uranium fuel system will be less costly than all others and can be used as the standard of comparison for other cycles.

The natural uranium cycle estimates which follow are based on the 8330 Mw thermal heavy water moderated, light water cooled reactor estimated by ORNL and Sargent and Lundy for the large desalination reactor study. The fuel element is patterned after a concentric tube being developed by du Pont, containing UO_2 powder brought to density by vibratory compaction. The cladding is Zircaloy tubing. The fuel element consists of two annular concentric rings with intervening cooling channel and a center hole; the outside diameter is about 4 inches. The uranium to Zircaloy ratio is 10, much higher than the current more segmented cores, using bundles of fueled rods of 0.5 inches in diameter or less (first Dresden core U/Zr = 3.4).

For capacities ranging from 1 to 30 tonnes per day, the fuel cycle costs for natural uranium are shown in Table 15.[11] The burn-up assumed for the natural uranium fuel is 7000 Mwd/tonne. The plutonium yield is 4 grams/kg uranium and is valued at $6.70/gram. These data are plotted in Figure 5.

2.4 Comparison of Cost of Partially Enriched and Natural Uranium Fuel Cycle Costs

In the foregoing sections, I have given estimates of fuel cycle costs for both partially enriched and natural uranium cycles. That a significant difference in costs exists between systems is evident, as developed in Table 16.

Because data were taken from a number of sources in preparing Table 16, there are some internal inconsistencies but the errors so introduced are not sufficient to change the rather conclusive evidence in favor of the natural uranium cycle.

Therefore, since it appears that natural uranium--D_2O moderated reactors of large size compete with partially enriched light water cooled reactors in capital and operating costs, the advantage offered by the natural uranium cycle is very significant.

From recycle plants of 10 tons per day capacity or larger, natural uranium fuel can be supplied at a net cost that approaches zero if

Table 15.

Cost vs. Capacity for Natural Uranium Fuel Cycle
(All costs $/kg U unless noted otherwise)

Capital charges: 7.7%/yr
Inventory charges: 5.5%/yr
Burnup: 7000 Mwd/tonne
Pu produced: 4 gm Pu/kg U
Concentric tube Zircaloy clad UO_2, U/Zr = 10

Capacity: Short tons U/day
On stream factor: 85%
All components single purpose

Cost Component	1 ton/day (2.82×10^5 kg U/yr)	2 tons/day (5.64×10^5 kg U/yr)	4 tons/day (1.14×10^6 kg U/yr)	6 tons/day (1.69×10^6 kg U/yr)	10 tons/day (2.82×10^6 kg U/yr)		20 tons/day (5.64×10^6 kg U/yr)	30 tons/day (8.46×10^6 kg U/yr)
					Throwaway			
U_3O_8 concentrate	13.00	13.00	13.00	13.00	13.00	13.00	13.00	13.00
Refinery U_3O_8 to UO_3	5.91	2.75	1.82	1.34	0.79	0.79	0.62	0.48
Zircaloy costs	4.50	4.19	3.58	3.58	3.58	3.58	3.54	3.40
Fabrication & shipping	8.09	4.49	4.42	3.56	2.55	2.55	1.50	1.09
Reject 5% (excl. U)	0.90	0.69	0.49	0.42	0.34	0.34	0.28	0.25
Subtotal, fuel fabrication	32.40	27.51	23.31	21.90	20.26	20.26	18.94	18.22
Reprocessing & Pu loss at 0.25%	25.34	13.52	7.62	5.91	1.00	3.93	2.56	1.91
Shipping 1200 mi RT	1.32	1.31	1.30	1.29	-	1.27	1.27	1.27
Inventory, reactor included	4.17	3.91	3.69	3.60	1.43	3.50	3.44	3.14
Subtotal, reprocessing	63.23	46.25	35.92	32.70	22.69	28.96	26.21	24.65
10% Contingency	6.32	4.63	3.59	3.27	2.26	2.90	2.62	2.46
GROSS, TOTAL	69.55	50.88	39.51	35.97	24.95	31.86	28.83	27.11
Pu credit @6.70/gm	26.80	26.80	26.80	26.80	-	26.80	26.80	26.80
	43.55	24.08	12.71	9.09	24.95	5.06	2.03	0.31
Mills/kwh, thermal	0.26	0.14	0.08	0.05	0.15	0.03	0.01	-
Cents/10^6 BTU, thermal	7.6	4.1	2.3	1.5	4.35	0.9	0.3	-
Gross cost at 14%/yr capital, 7.7%/yr inventory						35.18		
Net cost at 14% and 7.7%						8.38		

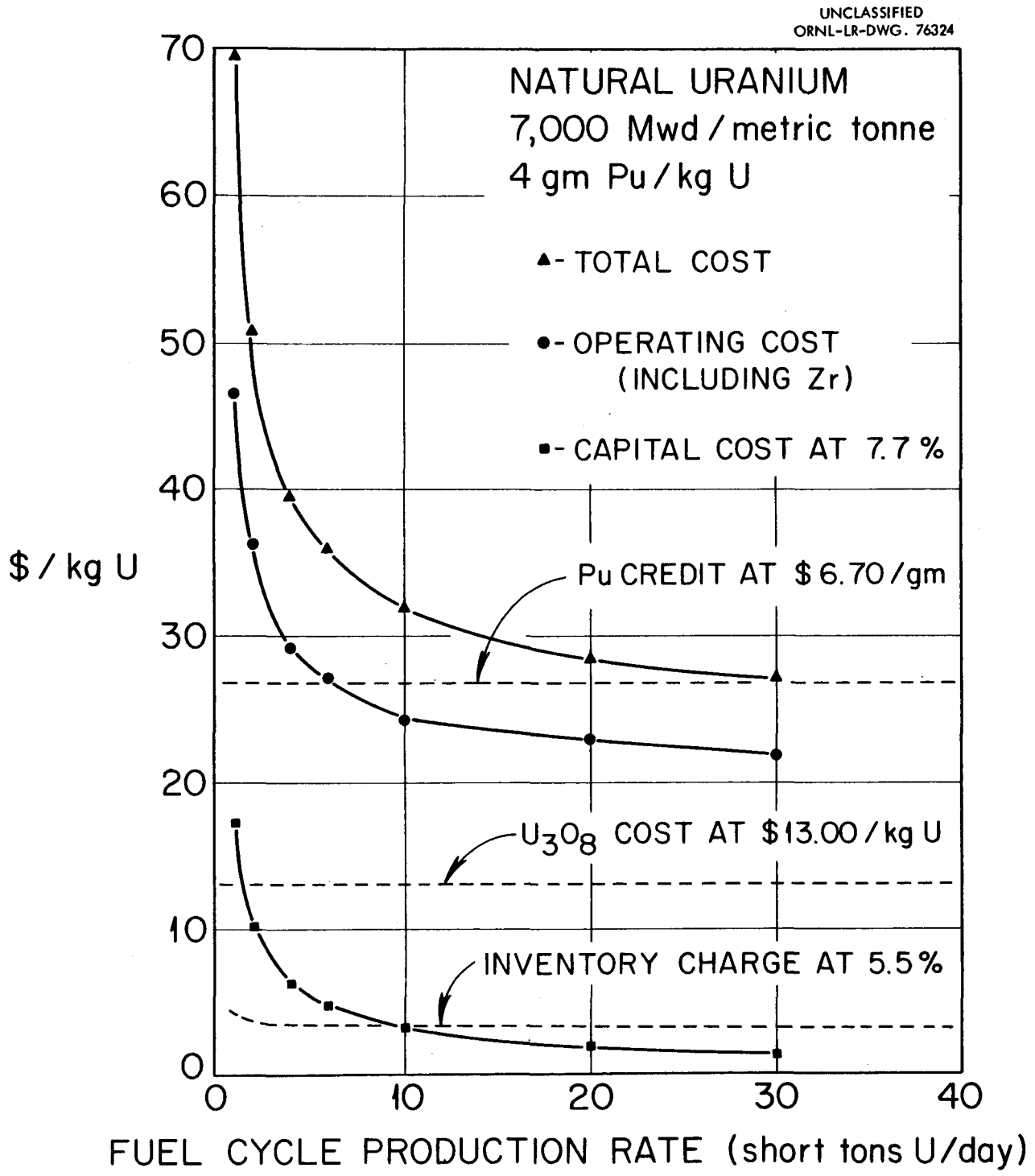

UNCLASSIFIED
ORNL-LR-DWG. 76324
NATURAL URANIUM
7,000 Mwd / metric tonne
4 gm Pu / kg U
▲- TOTAL COST
●- OPERATING COST (INCLUDING Zr)
■- CAPITAL COST AT 7.7 %
Pu CREDIT AT $6.70/gm
U_3O_8 COST AT $13.00 / kg U
INVENTORY CHARGE AT 5.5%
$ / kg U
70
60
50
40
30
20
10
0
0
10
20
30
40
FUEL CYCLE PRODUCTION RATE (short tons U/day)

Fig. 5.

plutonium can be sold for six to seven dollars per gram.

If plutonium cannot be sold because of temporary over-supply, then it can be stored (using the Thowaway case, Table 16) at a cost for fuel which is less than any of the enriched fuel cycle cases. It can also be recycled to the heavy water moderated system at a net cost that is not greater than that of natural uranium fuel.

Table 16. Comparison of Fuel Cycle Costs for Natural and Partially Enriched Fuels

	$/kg uranium Natural			$/kg uranium Partially Enriched	
	1 ton/day	10 ton/day	10 ton Throwaway	1 ton/day	10 tons/day
Feed costs	13.00	13.00	13.00	96.17	96.17
Inventory	4.17	3.50	1.43	17.42	17.42
Conversion and Fabrication	19.40	7.26	7.26	107.60	37.25
Reprocessing	25.34	3.93	1.00	32.01	6.30
Fuel Shipment	1.32	1.27	-	10.00	10.00
10% contingency	6.32	2.90	2.26	-	11.45
	69.55	31.86	25.05		
Adjustment[a]	17.16	3.32	1.48		
TOTAL	86.71	35.18	26.53	263.20	178.59
Pu[b] credit ($6.70/gram)	26.80	26.80	-	43.30	43.30
	59.91	8.38	26.53	219.90	135.29
Mills/kwhr electricity[b]	1.07	0.16	0.51	1.78	1.1
Advanced converter[c]					0.8

[a] Natural uranium case adjusted to raise capital charge rate from 7.7%/year to 14%.

[b] For natural uranium reactor, Pu yield: 4 grams/kg U at 7000 Mwd/tonne; For partially enriched reactor, Pu yield: 7.5 grams/kg U at 16,550 Mwd/tonne.

[c] Advanced converter performance from Table 13, burn-up cost and inventory from Table 14.

3.0 FUEL CYCLE TECHNOLOGY FOR PARTIALLY ENRICHED REACTORS

By far the most widely used fuel element materials for currently operating or proposed pressurized water, boiling water, and gas-cooled reactors are pelletized uranium dioxide, or a mixture of partially enriched uranium dioxide and thorium oxide clad in the protective metals, stainless steel or Zircaloy-2, as shown earlier in Table 1.

There are other important fuel systems. Metallic fuel cores of uranium-molybdenum alloys or uranium metal are to be utilized in the first cores of the Fermi Fast Breeder Reactor, the Hallam Reactor, the EBR-II, the Sodium Graphite Reactor, but will be replaced with cores of UO_2, UC, UC_2 or cermets of UO_2--stainless steel because of the limited metallurgical life under the operating conditions of radiation and temperature for these reactors.(12) Uranium--zirconium alloy seed cores will be replaced in the later cores of the Shippingport Pressurized Water Reactor with UO_2-ZrO_2 ceramics to achieve higher burn-up.

For gas-cooled reactors, advanced fuel designs incorporate UC or UC_2 in graphite without metal cladding, or BeO-UO_2 ceramics possibly clad with beryllium metal or refractory oxides; thorium may be included as the fertile material. The first such reactor to be constructed will be the Peach Bottom, utilizing a graphite container.(13)

For the next decade or two, it appears certain, however, that the fuel cycle for major power reactors in the United States will be based on UO_2, PuO_2, and ThO_2 ceramic cores clad in metal, either stainless steel or Zircaloy. For this reason, I shall describe briefly this most important recycle and will mention only the other methods that are under development or are of limited use.

For reactors fueled with partially enriched uranium, the over-all fuel cycle is shown in Figure 6. The fuel cycle employing natural uranium fuel is the same except that diffusion cascade is not required and the depleted uranium from the reprocessing plant is converted to possibly UO_3 or U_3O_8 for storage.

3.1 Uranium Ore Concentration

The fuel cycle starts with the recovery and concentration of natural uranium from ore. In the United States, uranium is recovered from ores and concentrated by hydrometallurgical operation, i.e., leaching with

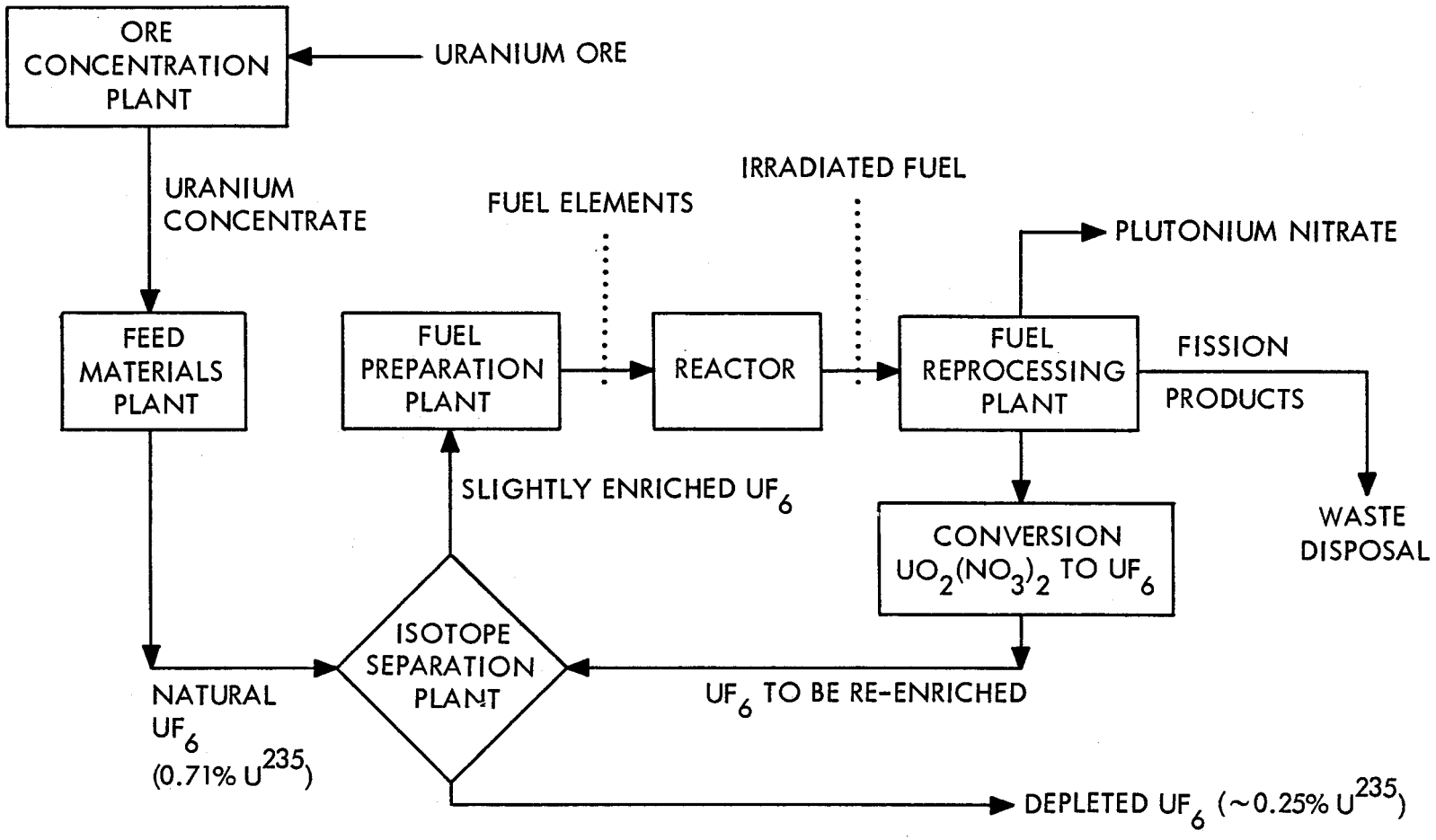


Fig. 6. Fuel Flowsheet for Thermal Reactors Fueled with Slightly Enriched Uranium.

sulphuric acid or sodium carbonate solutions.[14] Physical concentration of ores in the United States does not result in an acceptable yield of uranium. Uranium milling produces a uranium concentrate usually containing 70 to 90 per cent U_3O_8 from ores generally assaying from 0.08 to 0.5 per cent U_3O_8. Approximately 75 per cent of the United States milling capacity employs acid leaching and 25 per cent carbonate leaching. Between 15- and 20,000 tons/year of U_3O_8 are produced in the U.S.A.

3.2 Production of Feed Materials

The uranium ore concentrates are shipped from the mills to refineries in which the uranium is purified and converted into uranium metal or uranium dioxide, for use as natural uranium reactor fuels, or into uranium hexafluoride for feed to the isotope separation process.[15] Figure 7 illustrates the conventional process flowsheet used at the feed materials centers of the U. S. Atomic Energy Commission.

Although the chemical reactions underlying the various operations in the conventional process are practically the same as those used in the 1940's, the various operations have been the subject of intensive process and equipment research and development. The early batch operations have, for the most part, been replaced by continuous methods, and the equipment used in contacting reacting phases has been improved greatly. Large reductions in cost have been made.

In addition to the many considerable improvements already incorporated in the conventional process for uranium refining and conversion, new processes designed to replace part or all of the conventional process have received intensive study. Two U. S. processes which have been carried as far as the pilot plant stage with encouraging results involve:
(1) the production of pure UF_4 more directly from raw ore; and
(2) the refining of concentrates directly to UF_6 by fluorination. The first method involves the recovery of uranium for leach liquor and conversion to uranyl chloride by anion exchange, further purification by cation exchange, electrolytic reduction of the uranium to the tetravalent state, and finally aqueous precipitation of uranium tetrafluoride. This process permits the integration of uranium milling, refining and conversion (to UF_4) operations in one plant. Although it is not presently in use in the United States, the process has been adapted

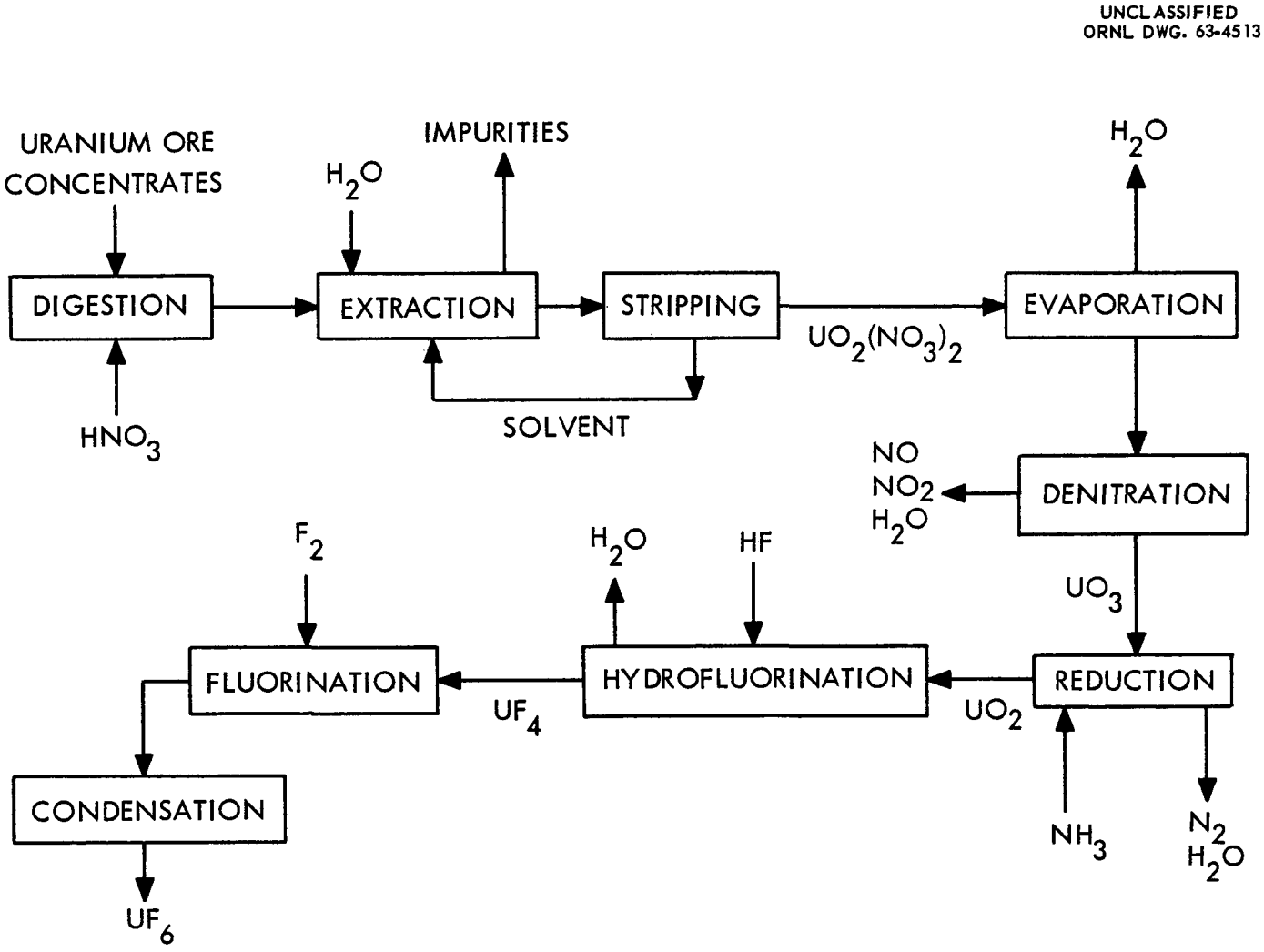


Fig. 7. Conventional Process for Uranium Refining and Conversion.

for use in Japan.[16]

The second process[17] has been utilized in the design of the newest uranium refinery in the United States, which was started up in 1959 and has a design capacity of 5,000 tons of U_3O_8 per year. The plant is privately owned and operated by the Allied Chemical Corporation which has a contract to supply UF_6 to the U. S. Atomic Energy Commission. It is the only plant-scale refinery operating in the United States which does not use solvent extraction for purification. Instead, differences in the volatility of the fluorides of uranium and its impurities are utilized in effecting purification. Fluidized-solid bed reactors are employed for all the chemical conversion steps.

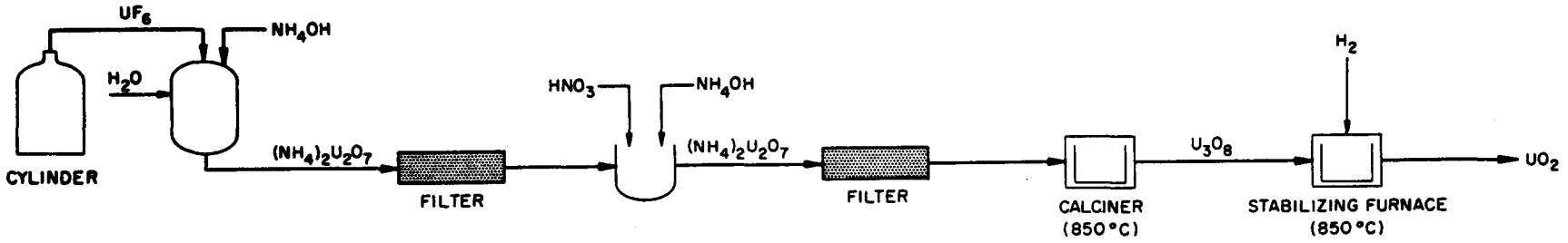
3.3 Uranium Isotope Separation

At least part of the fuel for all U. S. power reactors consists of uranium richer in U^{235} than 0.7115 wt %, the content of natural uranium. This enriched uranium is produced in the gaseous diffusion plants of the U. S. Atomic Energy Commission, from which UF_6 containing anywhere from 93.5 per cent to less than 0.3 per cent U^{235} can be withdrawn.

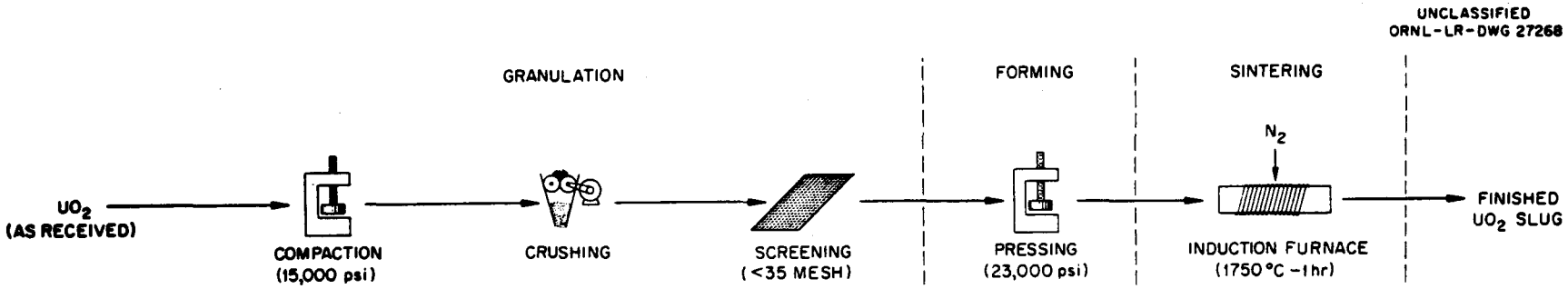
3.4 Preparation of Oxides and Fabrication of Fuel Elements

The UO_2 preparation cycle starts with 1.5 to 5.0 per cent enriched UF_6 from the gaseous diffusion cascades for partially enriched reactors. There are various methods of UO_2 preparation[18] but the one most successfully used employs alkaline diuranate precipitation and is shown in two variations in the schematic flowsheet, Figure 8.

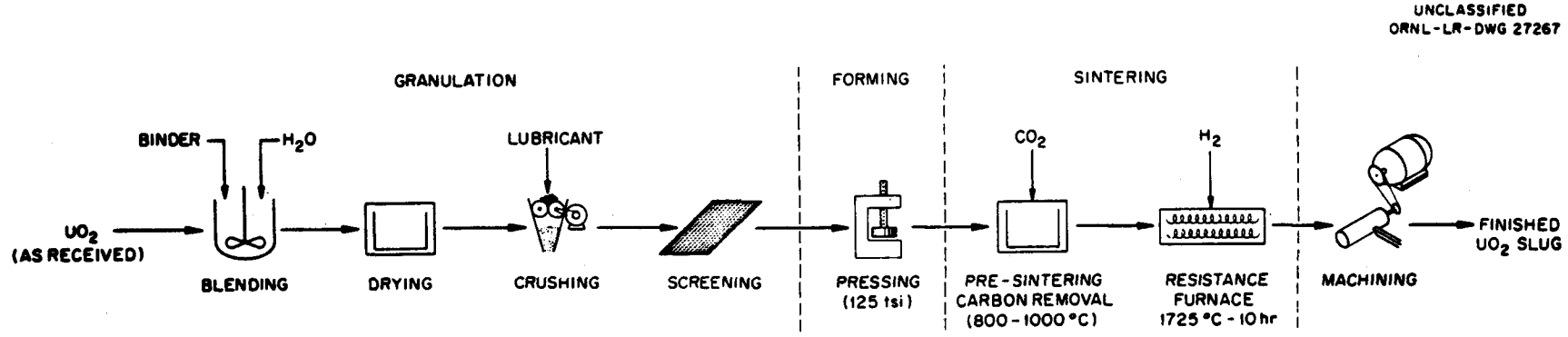
The most widely used process for the production of enriched UO_2 involves the hydrolysis of UF_6 in a dilute ammonia solution to give a precipitate of ammonium diuranate. After filtration and drying, this $(NH_4)_2U_2O_7$ is converted first to U_3O_8 by pyrohydrolysis with steam at 800°C and then to UO_2 by reduction with hydrogen at 800°C. The resulting UO_2 is ground to a fine powder in a cone mill to improve its sinterability. The UO_2 pellets are then manually or semi-automatically assembled into tubular cladding, inspected, and tested. The assembly and inspection steps for most tubular fuels are similar to those given in Figure 9, which



FLOWSHEET FOR PRODUCING UO_2 FROM UF_6



FABRICATION PROCESS FOR UO_2 PELLETS



ALTERNATE PELLET FABRICATION PROCESS

Fig. 8. Methods of Preparing UO_2.

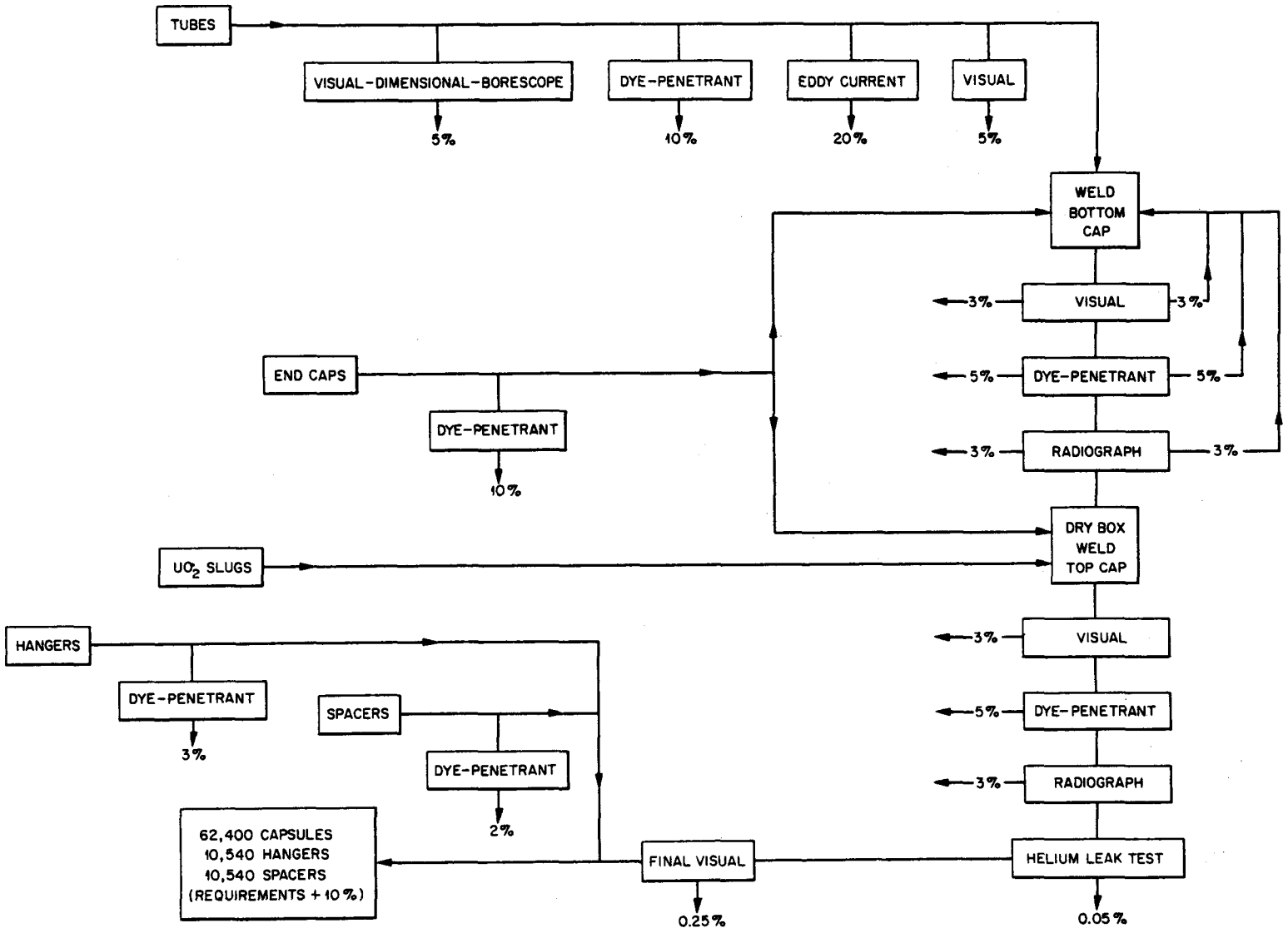


Fig. 9. Fuel Element Inspection Flowsheet.

also gives the rejection rate for stainless steel tubes and assembled fuel elements of the Experimental Gas-Cooled Reactor fuel loading.[19] This EGCR fuel assembly, shown in Figure 10, differs from all others only in the number, length, diameter of tubes, cladding material, and in that the oxide pellets have a center hole. It has been purchased at the lowest cost per kilogram of contained uranium of any fuel element fabricated to date, $62/kg uranium.

There are other methods of UO_2 preparation under study and development. UO_2 has been prepared by hydrogen reduction of UO_3 prepared by direct calcination of uranium nitrate, known as UNH or PWR type UO_2; it was used as the first loading of the Shippingport Pressurized Water Reactor. This material can be sintered to 92 per cent theoretical density at 1750°C after compaction at a pressure of 100 tsi.[20] In theoretical density it compares unfavorably with diuranate prepared oxide since the diuranate can be sintered to 97 per cent theoretical density at 1650°C after compacting to 20 tsi. Hanford has used fused UO_2, crushed and reformed. Hanford also has developed the Dynapak process which gives powders that can be compacted to 97 to 99 per cent theoretical density. Both Hanford and ORNL have used high density powders as feed for the manufacture of fueled tubes by vibratory compaction; compacted powder-filled tubes of 92 to 95 per cent theoretical density have been made. Hanford and ORNL have increased powder filled-rod UO_2 packing densities by swaging.

The Oak Ridge National Laboratory has a new process based on preparing UO_2 (or UO_2-PuO_2) via a sol-gel state which has produced essentially 100 per cent theoretically dense particles suitable for fabrication by vibratory compaction. Hanford has made UO_2 single crystals by electrodeposition from a fused potassium-sodium chloride melt[21] to be used possibly as a method for fuel recycle for the Plutonium Recycle Reactor. Argonne has produced UO_2 from UF_6 by steam hydrolysis in fluidized beds.

3.5 Reprocessing Irradiated Fuel Elements

Fuel elements and fertile blankets must be removed from the power reactor because of loss or mechanical deterioration of the fuel due to radiation or corrosion. The residual value of the fssionable and fertile material is always great enough to economically justify reprocessing

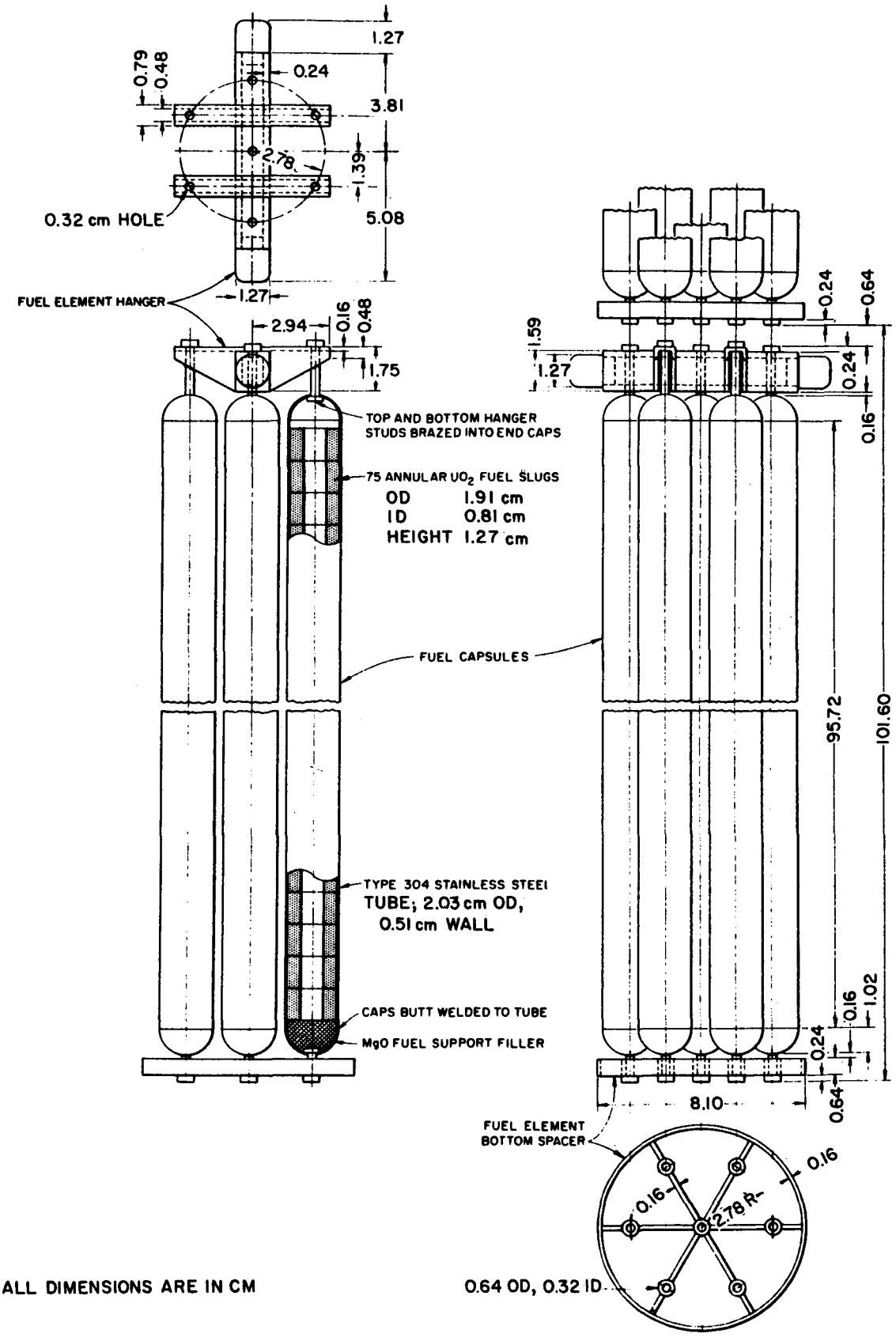


Fig. 10. EGCR Fuel Assembly.

partially enriched fuels. For natural uranium fuels, the value of plutonium that can be recovered may not justify reprocessing if the capacity of the reprocessing plant is small and, as a consequence, the unit costs of reprocessing is high.

3.51 Separations Processes

The basic chemical separations processes that have been studied for the separation of fissionable and fertile materials from spent fuel that are applicable to partially enriched uranium recycle are:

(1) Solvent extraction in which uranium, plutonium, and thorium are selectively removed from a nitric acid aqueous solution of the fuel by an immiscible organic solvent, tri-n-butyl phosphate in a purified kerosene diluent. This process and its modifications are highly developed; extensive production experience exists. By relatively simple changes in conditions solvent extraction can be used for all required power reactor fuel separations. It has been used for all radiochemical fuel processing to date, will most probably be used for power reactor fuels for some years to come, and will serve as the cost standard to which all other processes must be compared.

(2) Fluoride volatility in which the volatile of hexafluoride uranium, and possibly plutonium, is separated from relatively non-volatile fission product fluorides. At present, there are two process and process engineering approaches to fluoride volatility, fused salt and inert fluid beds. One variant of this basic technique is being tested with irradiated reactor fuel elements of the PWR and core type. Fully-enriched uranium alloyed with zirconium and clad with Zircaloy-2 is dissolved in a fused salt at 600°C by HF; UF_4 in the fused salt is converted to UF_6, which is volatized from the salt leaving behind impurities and fission products. Operability and high decontamination factors on a pilot plant scale have been demonstrated for fully enriched uranium; plutonium work is now in progress.

PuF_6 has been prepared and tracer experiments indicate that plutonium can be removed from active and inactive contaminants. Thorium fluorides are not volatile. The applicability of fluoride volatility processes to

the variety of partially enriched power reactor fuel elements containing plutonium has not been demonstrated, although current research offers very real promise that a process can be developed. Until data are available, the fluoride volatility processes cannot be compared cost-wise with solvent extraction for partially enriched fuels. I do not think that they will be cheaper than the well established aqueous methods for fuels that can be easily converted to aqueous solutions. Chloride volatility processes are not as promising as those using the fluoride system.

For the immediate future and almost certainly for the next decade, most power reactor fuels will be processed by the aqueous solution-organic extraction methods because of the vast production experience and great versatility of the method. The solvent extraction techniques that will be employed probably will be based on the use of tri-n-butyl phosphate as an extractant (although many others can be used) and will probably be modifications of the basic Purex process. I will not describe this well known and widely used technique.

3.52 "Head-end" or Feed Preparation Processes for Power Reactor Fuels

Once in solution, all fuels can be processed essentially in the same separation contactors used in the solvent extraction process. One controlling requirement is that the fuel elements or, better still, the core materials be placed in a mineral acid solution that is or can be converted to a predominantly nitrate base.

Therefore, during the past eight years effort has been concentrated on devising dissolution or "head-end" processes for power reactor fuels. For each of the present generation of power reactor fuels at least one, and in many cases more than one, acceptable dissolution and feed preparation method exists.

The "head-end" processing method that now appears to be capable of handling the greatest variety of fuel elements is the shear leach process. A chopper or shear has been developed which can cut bundles of fueled pins into short segments of tubing in which the core is exposed. The shear has been designed to contain all radioactive dust, to be remotely operable, to be capable of decontamination, and to discharge cut segments of fueled tubes and dust under sealed conditions to either a batch or continuous dissolver. By cutting Zircaloy and stainless steel jacketed pins to expose

the nitric acid soluble fuel cores, only one dissolver is required for a variety of fuels; further, the jackets can be discarded from the first step in processing. They are not dissolved in the solution feed stream, and as a consequence, the large quantity of cladding material need not be discharged with the high-level fission product wastes. Because of the potential for handling a variety of fuels, the more concentrated feed solutions (with respect to uranium) which can be prepared for the solvent extraction cascade, and the low-solids content high-level waste which will result from processing, the shear-batch leach process will be used by the Nuclear Fuel Services, Incorporated, the first small reprocessing company in the United States.

For stainless steel-clad fuels, a chemical dejacketing method using sulfuric acid (the Sulfex Process) has been developed. Stainless steel jackets can be dissolved selectively, leaving the fuel oxide core behind. After careful washing of the exposed oxide to remove sulfate ions, the core can be dissolved in nitric acid, A concentrated feed for solvent extraction is produced. To resist the corrosive attack of both sulfuric acid and nitric acid, a Nionel vessel and system is required. Nionel is an alloy of Ni(40%), Cr(21%), Fe(31%), Mo(3%), Cu(1.75%), with traces of manganese and silicon. This head-end will be used in the Eurochemic Plant for stainless steel fuels. The process produces a large volume of slightly active sulfate waste which must be neutralized and stored.

For Zircaloy-clad fuels, a similar chemical dejacketing process exists (the Zirflex Process) in which a solution of ammonium fluoride and ammonium nitrate is used to remove the metal. HNO_3 is used to dissolve the core. This process can be performed in stainless steel or Nionel. It produces a relatively large volume of slightly active waste which must be neutralized and stored. It will be used by Eurochemic. By adding hydrogen peroxide in controlled amounts, the Zirflex Process can be used to dissolve completely Zircaloy-clad, zirconium--uranium alloys.

Complete dissolution processes for stainless steel fuels are electrolytic dissolution (being perfected by du Pont and Phillips) and dissolution with aqua regia (the Darex Process). The latter must be performed in titanium equipment to a point in processing at which chloride ion is removed by distillation.

The electrolytic process may be usable for zirconium-containing fuels, also. Zirconium (and the tin in Zircaloy) dissolves but immediately precipitates as an oxide which must be removed. For complete dissolution of zirconium-containing fuels a mixture of nitric and hydrofluoric acid can be employed in a stainless steel dissolver, if the rate of addition of hydrofluoric acid is controlled (the Niflex Process). Work on this method has been done at Brookhaven National Laboratory, the Savannah River Laboratory, Hanford Atomic Products Operations, Phillips Petroleum Company, and the Oak Ridge National Laboratory.

My selection of processes for partially enriched fuels is shown in Figure 11, along with some of the other methods mentioned. Possibly the most compact summary of aqueous process possibilities is given in Nucleonics, (August, 1962).[22]

3.6 Waste Disposal

The technology for safely removing or isolating waste radioactive materials either exists or will be developed within the next ten years. Further, the selection of permanent disposal sites is being studied; several geologic formations are promising.

Radioactive gaseous fission products xenon, krypton, and iodine can be removed from the exhaust gases of the reprocessing plant. Processes exist that have been used under plant conditions.

Aerosols, both solids and liquids, have been removed by efficient scrubbing and filter systems. The filter systems can be improved in efficiency, versatility, ruggedness, and can be better engineered; but they work and are economically acceptable now.

Twenty years of experience with the burial of slightly contaminated solids has shown that this can be done safely and economically. Slightly contaminated combustible solids have been burned safely to reduce their storage volume, but the engineering evolution of incinerators and their gas cleaning systems has not kept pace with other waste disposal research; this is particularly true for combustion of slightly contaminated organic solvents.

Disposal of the high-level fission product wastes has received recently a large measure of research and development attention. It is very

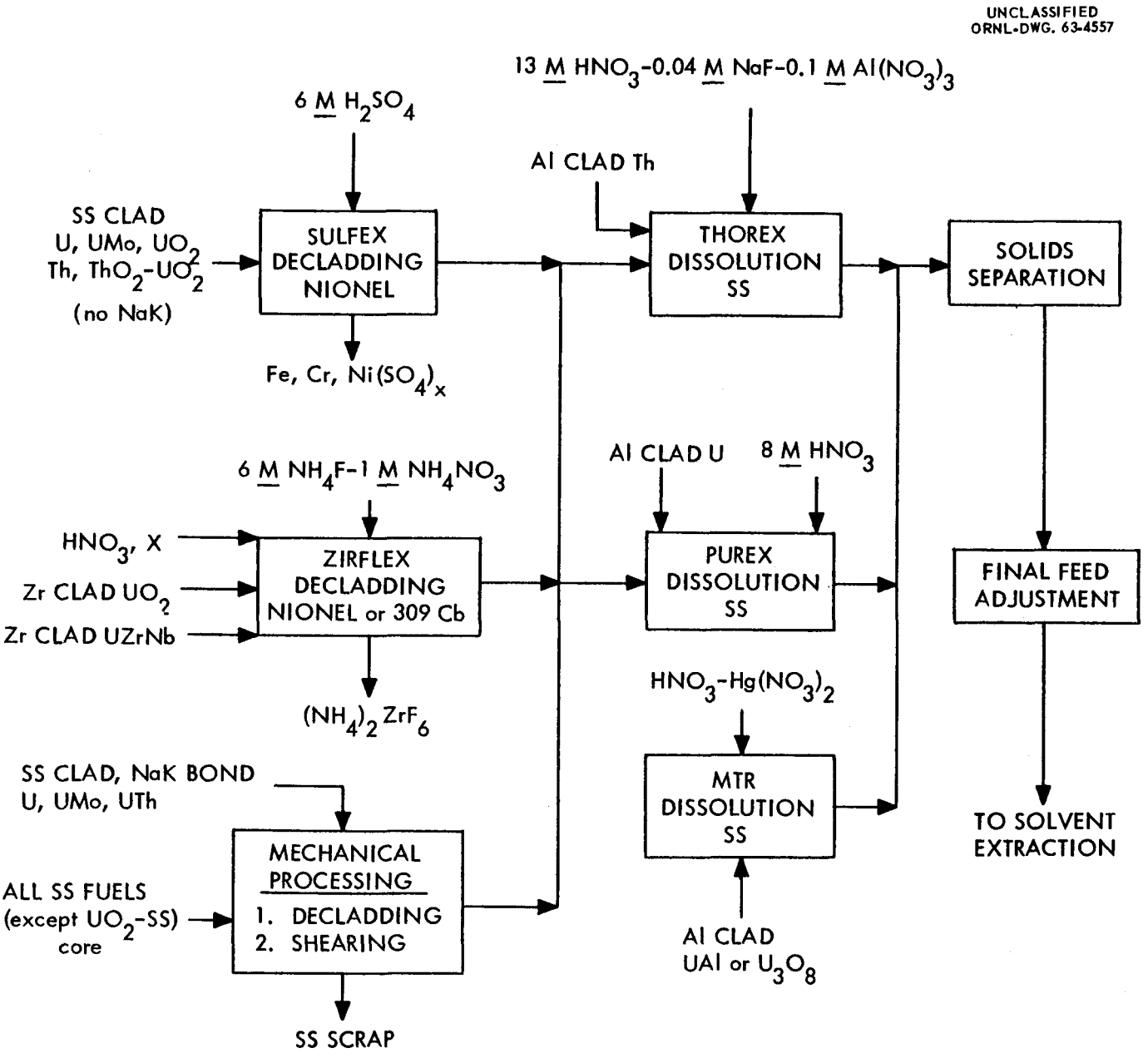


Fig. 11. Schematic Head-End Flowsheets.

probably that the fission products can be immobilized in very high fired insoluble oxides or glasses by any one or a combination of the four or five processes currently under development. In the United States, pilot plant demonstrations of processes for converting high-level wastes to refractory solids should be completed before 1970. At Idaho, a fluidized bed calciner is now ready for active runs. At Hanford, a versatile pilot plant facility is being built to test the Hanford spray calciner, the ORNL pot calciner, the Brookhaven, ORNL, and probably the Hanford glass-making process, and other innovations that will evolve during this proof testing period. Similar work in France and England is moving at about the same pace. That these waste fixation schemes are economically acceptable has been shown by a three year economic analysis at ORNL.[23][24][25]

Low-level, large volume liquid wastes can be discharged to the environment after treatment by evaporation, precipitation, ion exchange, foam separation, soil exchange, and several other methods under development. Complete demineralization and recycle of water is possible. Most methods are economically acceptable, some much more so than others. Development now in progress will make it possible to select the most efficient and economical methods.

Some promise of having revenue from the fission products exists, particularly for small heat sources.[26][27] Processes exist or are being developed for almost quantitative removal of strontium, cesium, cerium, promethium, and other isotopes. However, a rather cursory study of the cost of recovery versus the sale of these isotope would indicate that no significant reduction in the cost of waste disposal would result.[28]

Study of the formations that are best for permanent waste disposal sites in the earth's crust is now going forward. For high-level wastes, the extensive salt deposits in the United States are promising. For intermediate-level wastes, an adaptation of the hydrofracturing technique used by the oil industry is being studied. Waste solutions are mixed with cement and sand, infected under high pressure into a rock formation.

REFERENCES

1. United States Atomic Energy Commission, Civilian Nuclear Power ... A Report to the President - 1962, USAEC (Nov. 20, 1962)

2. Appendices to Civilian Nuclear Power ... A Report to the President - 1962, USAEC, Table 2, p. 56 (Nov. 20, 1962)

3. Ibid., Table 4, p. 58

4. Ibid., pp. 66-67

5. Ibid., Table 14, p. 62

6. Bechtel Corporation (et al), Large Reactor Study for Sea Water Desalination, Appendix A., pp. A-1 through A-39 (July 1963)

7. St. John, D. S., Wade, J. W., "Economics of Heavy Water Reactors," Nucleonics 20, p. 52 (Dec. 1962)

8. Hammond, R. P., "Large Reactors May Distill Sea Water Economically," Nucleonics 20, p. 45 (Dec. 1962)

9. Young, Gale, Hammond, R. P., Spiewak., I., Prospects for Sea Water Desalination with Nuclear Energy, ORNL-TM-465 (Jan. 1963)

10. Weinberg, A. M., Young, Gale, "Scale, Nuclear Economics, and Salt Water," Nuclear News, ANS (May 1963)

11. Culler, F. L., The Effect of Scale on Fuel Cycle Costs for Enriched and Natural Uranium Fuel Systems, ORNL-TM-564 (April 1963)

12. "Fast Breeders," Power Reactor Technology, USAEC, pp. 85-96 (Sept. 1961)

13. de Hoffman, F. D., and Fortesque, P., "Application of High Temperature Gas Cooling to Nuclear Power Plants, the HTGR," General Atomics 1515 (1960)

14. Clegg, J. W., and Foley, D. D., Uranium Ore Processing, Addison-Wesley Publishing Co., Inc., Reading, Mass. (1958)

15. Harrington, C. D., and Ruehle, A. E., Uranium Production Technology, D. Van Nostrand Co., Inc., New York (1959)

16. Chemical Week 85: 35 (July 11, 1959)

17. Lawroski, S., et al, "Production of Refined UF_6 from Ore Concentrates by Fluidization and Fractional Distillation Techniques," PICG(2), 4: 44, United Nations, Geneva (1958)

18. Uranium Dioxide; Properties and Nuclear Applications, Chapter 2, USAEC (1958)

REFERENCES (cont.)

19. Patriarca, P., and Hiestand, R. L., "A Bulk of UO_2-Stainless Steel Fuel Element for a Gas-Cooled Reactor," Paper 17, TID-7559 (Part 1) USAEC, Report of Fuel Elements Conference (August 1959)

20. Harrington, C. D., and Ruehle, A. E., "Preparation and Properties of UO_2 Powder," Chemical Engineering Progress 54, No. 3 (1958)

21. Lyon, W. L., Voiland, E. E., The Preparation of UO_2 from a Molten Salt Solution of Uranyl Chloride, HW-62431, Hanford Atomic Products Operation, General Electric Company

22. Culler, F. L., Blanco, R. E., Ferris, L. M., Nicholson, E. L., Rainey, R. H., Ullmann, J. W., "Proposed Aqueous Processes for Power Reactor Fuels," Nucleonics, pp. 124-127 (Aug. 1962)

23. Bradshaw, R. L., Perona, J. J., Roberts, J. T., Blomeke, J. O., Evaluation of Ultimate Disposal Methods for Liquid and Solid Wastes. I. Interim Liquid Storage, ORNL-3128 (Aug. 7, 1961)

24. Perona, J. J., Bradshaw, R. L., Roberts, J. T., Blomeke, J. O., Evaluation of Ultimate Disposal Methods for Liquid and Solid Radioactive Wastes. II. Conversion to Solid by Pot Calcination, ORNL-3192 (Sept. 27, 1961)

25. Chemical Technology Division Annual Progress Report, Period Ending June 30, 1962, ORNL-3314, pp. 90-95 (Sept. 13, 1962)

26. Davis, H. L., "Radionuclide Power for Space. I. Isotope Costs and Availability," Nucleonics, pp. 61-65 (March 1963)

27. Rohrman, C. A., Radioisotope Heat Sources, HW-76323, Hanford Atomic Products Operation, General Electric Company (Feb. 1, 1963)

28. Perona, J. J., Blomeke, J. O., Bradshaw, R. L., Roberts, J. T., Evaluation of Ultimate Disposal Methods for Liquid and Solid Radioactive Wastes. V. Effects of Fission Product Removal on Costs of Waste Management, ORNL-3357 (June 1963)

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