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STATUS OF ELECTROLYTIC

DISSOLUTION FOR

LMFBR FUELS

AUGUST 1971

HANFORD ENGINEERING DEVELOPMENT LABORATORY
UNITED STATES ATOMIC ENERGY COMMISSION

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C.R. Cooley

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STATUS OF ELECTROLYTIC DISSOLUTION FOR LMFBR FUELS

ABSTRACT

Electrolytic dissolution technology was reviewed and found sufficiently developed to reprocess LMFBR fuels. Capacity is limited by design configuration, heat generation and current density. New designs of multibarrel electrolytic dissolvers were estimated to process 0.5 to 1 MT/day of LMFBR fuel depending on the amount of stainless steel cladding processed with the fuel. Although estimates of the capital costs for new electrolytic dissolvers are reasonable, the incremental cost for installation of additional plant capacity for waste systems to handle additional liquid waste from the process is excessive as compared to current shearing and acid leaching.

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STATUS OF ELECTROLYTIC DISSOLUTION FOR LMFBR FUELS

1.0 INTRODUCTION

Reprocessing of irradiated fuel from Liquid Metal Fast Breeder Reactors (LMFBR's) will be required in order to recycle large quantities of fissionable plutonium for use as fuel. As a first step in reprocessing spent Light Water Reactor fuels, shearing and leaching is used at commercial plants for dissolution prior to solvent extraction. Alternatively at USAEC production sites electrolytic dissolution of specialty fuels is used as a head end step in which cladding and fuel are totally dissolved rather than leaving hulls from shear leach operations for disposal. To gain perspective on the applicability of electrolytic dissolution for future LMFBR fuels, this overview study was conducted.

In order to more fully assess the performance of electrolytic dissolution on LMFBR fuels, existing technical information was reviewed and applied to the special situations introduced by LMFBR fuels. In particular, attention was given to the uncertainties concerning sodium with the fuel, overall capacity, criticality, subsequent solvent extraction, waste disposal and capital costs. The influence of electrolytic dissolution on each of these areas is important, particularly, if it becomes necessary to process LMFBR fuels in any of the existing AEC production facilities until commercial companies can economically justify capital investments for processing campaigns on LMFBR fuels.

If USAEC production facilities process any LMFBR fuels, electrolytic dissolution could be used. An electrolytic dissolver for LWR fuels already exists at Savannah River^(1,2) and one is being constructed at ICPP.^(3,4) Electrolytic dissolution was chosen for both locations because considerable development work was already done, because electrolytic dissolution offered a low initial cost for equipment to process low throughputs of specialty fuels and because shearing facilities did not already exist.

2.0 SUMMARY

Electrolytic dissolution is an attractive processing method for low throughput of specialty fuels (<1/4 MT/day per dissolver). The technology

is well developed and considerable practical experience exists. However, the capacity throughput (MT/day of fuel) is limited by heat loads from electrical resistance of electrodes and dissolver solution. Consequently, the dissolution rate of stainless steel, which is directly related to the amount of direct current, is slow; as a result, equipment for high capacity units is expected to be expensive.

For LMFBR fuels, geometrically controlled criticality in dissolvers is preferable to nuclear poisons because of uncertainties about formation of sludge which may include PuO_2 . This introduces a requirement for multiple barrels and multiple electrodes. Within the geometric restraints of criticality, the available volume for electrolytic dissolution is small. Further, the electrical current which can be transmitted does not permit high dissolution rates. Although nuclear poison and multiple electrodes can be used to increase capacity, the capital investment for scale up is projected to be excessive as compared with a shear leach system. If only the capital investment for a dissolver were compared to a shear leach, electrolytic dissolution would be competitive with shear leach. However, increases in the capacity of waste handling equipment including that required for waste solidification (~70% of total costs), escalates the capital costs; e.g., incremental capital costs for a 1 MT/day capacity in a new plant for the AI Reference fuel are \$3290K for electrolytic dissolution as compared to \$1250 to \$2400K for shear leach.

Electrolytic dissolution can be considered reasonably competitive to shear leach for overall plant capacities less than 0.5 to 1 MT/day. Beyond this capacity shear leach is expected to be less costly primarily because a single multipin shear is expected to have a capacity of at least 5 MT/day.

3.0 STATE OF DEVELOPMENT FOR LWR FUELS

At two AEC sites, NRTS, Idaho and Savannah River, South Carolina, extensive development work has been conducted on electrolytic dissolution. Savannah River has operated electrolytic dissolution with radioactive fuel⁽²⁾ and Idaho Nuclear is currently installing a dissolver for use on EBR-II fuels. Developments have been extensively documented. Significant generalized facts on electrolytic dissolution include:

1. The largest operating dissolver is 10,000 amperes at Savannah

River with approximately 5 ft² of platinum anode. Nearly 55 kg of SS tubular fuel have been dissolved per day.

2. Dissolution is conducted at 40 volts or less and with a current density of about 2 amps/cm² of electrode area. This dissolves about 0.45 g SS/amp-hr (about 75% of theoretical).
3. From 3 to 8 weight percent of the stainless steel remains in the dissolver solution as a fine metallic particulate which readily settles as a sludge, some 40% is -325 mesh (44 micron). For zirconium cladding, nearly 85% occurs as ZrO₂ flakes, about 50% is -40 +80 mesh.
4. Nitric acid use through evolution of NO_x and formation of soluble salts is 20 moles HNO₃/1000 amp-hr or 44 g-moles/kg SS dissolved.
5. The cathode can be constructed of niobium, tantalum or titanium. However, the process acidity must be kept >6M and the total nitrate concentration >7M in order to keep quantities of hydrogen liberated at the anode minimal and thereby prevent embrittlement. At ICPP the dissolver design uses the dissolver vessel walls as the cathode. At Savannah River electrodes are used inside a stainless steel dissolver. Localized anodic corrosion of the dissolver is prevented by shielding the cathode.
6. The anode must be constructed of platinum since it is the only known material which is not severely attacked as an anode in nitric acid. In order to reduce costs, an inert base metal is used to provide strength for the anode. Niobium and tantalum are suitable base metals as long as their individual breakdown voltage is not exceeded, i.e., 45 volts for niobium and 140 volts for tantalum. The corrosion of platinum anodes decreases as the nitric acid concentration increases. However, the corrosion of platinum increases as the temperature is increased. The corrosion rate is about 2 to 3 mils/year up to 185°F. At 195°F the rate is >6 and at 200°F >11 mils/year. Thus, the operating temperature is usually kept <160°F.
7. The dissolver basket, which serves to electrically insulate the fuel from contacting the cathode or anode, can be constructed of niobium

or aluminum oxide. If niobium is used, then aluminum oxide is used to insulate the basket from the electrodes. A niobium basket is expected to last at least a year, but enough electrical arcing occurs between the basket and the fuel that over a prolonged period arcing is expected to deteriorate the basket.

8. One of the most important material problems associated with electrolytic dissolvers is not the dissolver material but materials for equipment which must handle the waste containing the dissolved stainless steel. Chromium⁺⁶ ions are formed in nitric acid solutions which in turn react with Cr⁺³ oxide protective layer on stainless steel to produce continuing corrosion. In the dissolver, Cr⁺⁶ is produced at the anode, but is reduced by the oxides of nitrogen at the cathode. Because the dissolver is never operated above 85°C and no boiling occurs to remove oxides of nitrogen, corrosion in the dissolver is not considered severe enough to be a problem. However, boiling solutions containing stainless steel in nitric acid produce serious corrosion problems, i.e., 100 to 200 mils/month, and boiling of such waste solutions has been avoided. Generally the corrosion of stainless steel equipment is increased by the following conditions:⁽⁵⁻⁹⁾
 - a. Increases in the concentrations of Fe⁺³ and Cr⁺⁶ which are predominant at higher nitric acid concentrations.
 - b. Increases in the total nitrate content of the solution.
 - c. Increases in temperature.
 - d. Decreases in the nitrite concentration.
9. Nearly all of the electrical energy is dissipated as heat in the electrolyte. Sufficient cooling must be provided to keep the temperature at optimum process conditions (120 to 175°F). Boiling is undesirable because gas "blinds" the electrode.
10. Uranium loss to the solids has been low, i.e., <0.1%.
11. Hydrogen concentration in the dissolver off-gas without dilution is usually between 1 and 2% which is well below the dry explosive limit of 4% H₂.

12. Sodium bonded uranium fuels have been dissolved with a sodium content of 0.01 kg Na/kg U and 0.5 kg SS/kg U or ~ 0.02 kg Na/kg SS.
13. Some electrode sludge, mostly silica, occurs and cleaning of electrodes with NaOH may be required to achieve extended operating campaigns. Little testing has been done with simulated fission products and the effect of long term accumulation on electrodes is not well known.
14. Nearly all materials have been dissolved electrolytically except vanadium and vanadium alloys (a future cladding candidate). No record was found on their dissolution electrolytically.
15. Use of soluble poisons in the dissolver is planned by Idaho Nuclear. The effects of soluble poisons on dissolver operation are being determined but are not expected to be detrimental.

4.0 APPLICATION TO LMFBR FUELS

4.1 Projected Capacity

Considerable scale up beyond current experience is required if the capacity of a single electrolytic dissolver is designed for 1 MT/day of spent LMFBR fuel. The logical approach to scale up involves using multi-barrel dissolvers and increasing the effective amperage through the electrodes, e.g.: use of a 6-barrel dissolver and increasing the amperage from the normal 2 up to 4 amps/cm² of anode area (if proven practical) would provide sufficient capacity for 0.5 MT/day of LMFBR fuels. Increasing the anode current to 8 amps/cm² of effective anode area (if proven practical) would provide sufficient capacity for 1 MT/day of spent LMFBR fuel. The maximum current density needs further definition particularly if the effective surface of an electrode is increased until the solution becomes current density limiting.

Although a limit of about 1 MT/day was selected as the maximum processing capacity of an electrolytic dissolver, this is little different from a shear-leach six basket dissolver if it is designed geometrically safe. If the volume of hulls is 17 ft³/1000 kg SS, e.g. 20 ft³/MT for AI reference fuel with 1189 kg SS/MT, and the basket is limited to 5 inches in diameter and a 9 foot fill (1.2 ft³/basket) then a 12 hour

leaching cycle would give 0.7 MT/day as compared to 0.4 MT/day for electrolytic dissolution. As the amount of stainless steel increases and the volume of hulls increases, e.g. for FTR, the capacities of a geometrically safe shear leach dissolver and an electrolytic dissolver are within 0.1 MT/day of each other with the shear leach having the slightly greater capacity. Of course the use of poisons in a shear leach approach permits larger baskets and correspondingly higher capacities, while the use of poisons in the electrolytic dissolver does not directly provide a means to significantly increase the throughput of the system for LMFBR fuels because the undissolved fuel particles restrict the design to one critically safe by geometry.

4.1.1 Dissolution Rate

The overall capacity of an electrolytic dissolver can be affected by the rate of dissolution of stainless steel and the rate of dissolution of UO_2 and PuO_2 . The rate of dissolution of UO_2 and PuO_2 is important because the oxide must dissolve sooner than or at the same time as the cladding or a separate operating step for dissolution of the residual oxide will be required. On the basis of present sparse data on LMFBR fuels, a separate UO_2 - PuO_2 dissolution step may be required. Subsequent developments may improve the dissolution characteristics and eliminate the need for a separate step since it is generally not required for LWR fuels. The high acid concentration ($>6M$) for electrolytic dissolution is favorable for complete dissolution.

A number of factors⁽⁸⁻¹¹⁾ affect the rate of dissolution of ceramic oxides in nitric acid.

1. The dissolution rate of the oxide can be increased by a factor of 3 or greater by the presence of nitrite in the solution.
2. Increases in the nitrate concentration generally increase the dissolution rate in proportion to the nitrate concentration raised to the 2.8 power.
3. The presence of ferric ion increases the dissolution rate by a factor of 2 or more. Its effect is about the same as the combined effect of dissolved stainless steel.

4. Dissolution rate increases with temperature up to the boiling point with approximately a doubling of the rate for every 10°C increase in temperature. However, boiling reduces the dissolution rate from one half to one fifth of the rate without boiling because of the reduction in nitrite concentration.
5. Similarly, agitation at temperatures below boiling can decrease the dissolution rate by a factor of four.
6. Normally the dissolution rate of UO_2 is about 4 times higher than metallic uranium. However, the introduction of PuO_2 into the UO_2 reduces the overall dissolution rate of UO_2 by at least 40%. However, the increase in the surface area inherent in dissolution normally increases the rate based on the original nominal surface by a factor of 2. On spent fuel from a reactor, the dissolution rates of UO_2 are generally about a factor of 5 higher than on a new ceramic fuel.

Since an electrolytic dissolver will be operated between 50 and 85°C, a simple scouting experiment using 50°C and 6M HNO_3 was conducted on the dissolution rate of FTR* pellets. After 6 hours, 43% of the plutonium originally in the pellet had dissolved to give an overall penetration rate of 3 mils per hour based on plutonium dissolution. The U/Pu weight ratio in the solution after 6 hours was 5.2 as compared with the initial pellet of 3.0. In another test a pellet was exposed to 3M HNO_3 at 25°C for 6 hours after which 2.4% of the plutonium had dissolved and the U/Pu ratio in the final solution was 15.2 as compared to the initial 3.0. In other tests** on FTR pellets from the same batch using boiling 12M HNO_3 for 6 hours, 98% of the uranium had dissolved and 85% of the plutonium had dissolved. Thus, the dissolution rate of unirradiated pellets under electrolytic dissolver conditions is about one half the rate in concentrated acid.

If these sparse data are used to project the probable dissolution cycle in an electrolytic dissolver for an LMFBF fuel which typically

* Fast Test Reactor being constructed at Richland, Washington.

** Work to be published by R. E. Lerch, WADCO.

may be 200 mils in diameter, the dissolution of 100 mils of oxide at 50°C would require some 30 hours (about the time required for batch dissolution of the stainless steel). Although raising the temperature improves the rate there is considerable doubt that residual particles will be dissolved. Therefore, the basic assumption for analyzing the overall capacity of an electrolytic dissolver was to use the electrolytic dissolver for the dissolution of stainless steel without attempts to assure complete dissolution of the ceramic oxide during dissolution of the stainless steel. Either a subsequent digestion step in the same dissolver will be required or the undissolved particles of $\text{UO}_2\text{-PuO}_2$ must be transferred to a separate vessel for digestion in order to assure complete dissolution.

Since the amount of stainless steel which dissolves in an electrolytic dissolver is directly proportional to the amount of current passing through the stainless steel, prediction of the overall capacity involves simply knowledge of probable efficiency of the dissolver, i.e., total current utilization, and the amount of stainless steel between the electrodes. Although the overall capacity of throughput is somewhat complicated by the fact that there are different amounts of stainless steel along the profile of a fuel, the effect of variations is minimized by designing the electrodes closer together at the bottom. The combined effect of less solution resistance and a higher bulk density of stainless steel increases current flow at the bottom which in turn increases the dissolution rate to the point that the dissolution rate of bulky portions of the fuel (e.g., the Inconel 600 reflector rods at each end of the pellet stack of the FTR driver fuel) should approach uniform throughput as long as the residual rods from previous subassemblies do not physically hold the next fuel subassembly above the electrode level. Therefore, as long as the design properly accounts for the amount of residue from previous subassemblies, it will permit operation of the dissolver at maximum efficiency and capacity.

The rate of dissolution of stainless steel directly depends upon the total current which is limited by current density, amps/cm^2 . Current density is limited by anode configuration, depth and operating conditions. It is a function of voltage, solution composition, solution

temperature and the rate of heat transfer from the anode surface. Normally, 2.0 to 2.6 amps/cm² are used for platinum electrodes although values as high as 3.5 amps/cm² have been used. Platinum normally limits the current to 10 to 20% below the other materials under the same conditions. Therefore, dissolving rate is normally not limited by cladding or cathode materials in the system but by the surface area of platinum adjacent to the fuel. Increasing the temperature lowers current density, but current density increases as the electrolyte is agitated. The sole objective of maximizing current density is to provide the maximum dissolution rate of stainless steel between electrodes. Thus, the maximum bulk density of stainless steel pieces between the electrodes is also desirable.

On the basis of present design and experience, the electrical efficiency with full electrode coverage (i.e., fuel between all portions of electrode) has been about 75% to dissolve 0.42 to 0.46 g SS/amp-hr.⁽²⁾ If the electrode is not well covered, its efficiency is reduced to near 60%. Overall dissolution rates vary from 55 kg/day with tubular fuel to 85 kg/day for plate type assemblies if the current flows 86% of the time.

On the above basis, for an FTR subassembly (shown in Figures 1 and 2) the dissolution rate can be estimated as shown in Table 1 if the time to digest residual UO₂-PuO₂ fines is not included as part of the throughput cycle. About 75 subassemblies (SA's) per year (~2.5 MT/year) may be discharged from FTR. If these are handled whole, 136 days of operating time would be required for stainless steel dissolution without the oxide digestion step. If the fuel is dismantled by only 2 cross section cuts (leaves 1325 kg SS/MT), the time is reduced to 62 days. If the fuel is completely dismantled down to individual pins containing only the core length, 41 days will be required to dissolve the stainless steel cladding.

The capacity relationship for a single barrel was calculated as shown in Figure 3 for the FTR fuel. (Here the anode area is set to match the projected area of the fuel; therefore, rates are slightly different from Table 1.) The normal situation is shown by a dotted

FIGURE 1: Fast Test Reactor (FTR) Driver Fuel

FIGURE 2: FTR Driver Fuel Pin

*SURFACE DENSITY IS THE WEIGHT OF S.S. BETWEEN ELECTRODES: 1.8 = Maximum in FTR fuel 0.7 = Overall average of fuel with plenum end and bottom end removed
 0.6 = Average of fuel 0.5 = Ends, plenum and duct removed
 with only ends removed
 0.4 = Only ends and duct removed.

FIGURE 3: Capacity of a Single Barrel Electrolytic Dissolver for FTR Fuel

line where 2 amp/cm² of anode area operates at 80% efficiency dissolving 9.6×10^{-4} kg SS/hr-cm² of anode area. The calculated dissolution of FTR fuel is shown for various surface densities* of FTR fuel obtained by various degrees of dismantling of the fuel. Principally, Figure 3 shows the differences in overall dissolving rates for different sections of the fuel under the same conditions. The 1.77 kg/cm² surface density is for the portion of the subassembly which contains Inconel 600 rods as reflectors. Other surface densities are averages for the section of the subassembly indicated. Thus, for a typical operating condition (1×10^{-3} kg SS/hr-cm²) the rates vary about a factor of 5 from .35 to 1.8 SA's/day. Dismantling normalizes operations to give a variation of 1 to 1.8 SA's/day. Removal of the duct (about 40% of the SS in a SA) gives rates of 1.5 to 1.8 SA's/day while retaining the duct reduces the rate to 1 to 1.2 SA's/day.

TABLE 1. Typical Estimated Throughput of FTR Fuel for Existing Designs of Electrolytic Dissolvers Not Critically Safe by Geometry^(a)

<u>Dismantling</u>	<u>kg SS/MT</u>	<u>FTR SA's/Day</u>	<u>MT/Day</u> ^(b)
None, Maximum SS/U-Pu Ratio	2910	.55	.019
Ends Removed	1990	.81	.028
Bottom End and Plenum End Removed	1325	1.21	.042
Ends Removed and Duct Removed	1250	1.28	.044
Ends, Duct and Plenum Section Removed, Minimum SS/Pu-U Ratio	870	1.83	.063

- a. i.e., only the throughput is estimated without consideration of whether poisons or other means are used to maintain nuclear safety.
- b. MT's of U and Pu based on a vertical dissolver with 4-3/4 ft² of anode area, an electrode height of 35" and a rate of 55 kg/day with a nominal 29 FTR SA's/MT and with current flow 86% of the time.

*

Surface density as used here is the weight of stainless steel in the fuel projected onto a plane surface bounded by the outside dimensions of the fuel.

A good approach to increasing capacity, as shown by the slopes in Figure 3, appears to be increasing the current density. The principal limitation to increasing current is the rate of heat transfer from the anode and possible corrosion effects. If we assume that the current density can be increased from a nominal 2 amps/cm² to 8 amps/cm² either by improved heat transfer or by increasing the effective area of the electrode per unit of projected area, the increase in capacity for a typical situation in a 6-barrel dissolver is shown in Table 2.

TABLE 2. Effect of Increase in Amperage on Throughput,
FTR Subassemblies/Day^(a)

	2 amps/cm ²		8 amps/cm ² (b)	
	1 barrel	6 barrels	1 barrel	6 barrels
2-X Section of Core (1325 kg SS/SA) 54" Length 2000 cm ² Anode Area 75% Efficiency	0.95	5.7	3.8	22.8
Ends Only Cut Off (1990 kg SS/SA) ~100" Length 3500 cm ² Anode Area 75% Efficiency	1.11	6.7	4.4	26.6

- The rates do not include time for charging or time for residual UO₂-PuO₂ digestion.
- Effective current densities of 8 amps/cm² have not been tested or proven.

The maximum rate of 26.6 SA's/day approaches 1 MT/day (29 SA's/day). If 4 charges are made/day which take 30 min each, the rate corresponds to 24 SA's per day or about 0.8 MT/day.

The FTR driver fuel design is somewhat different from other proposed LMFBR's; an estimate of the throughput of other LMFBR fuels is shown in Table 3. Here, the area of each of 6 individual electrodes is 5000 cm². Charging and cleanout is assumed to take 20% of the operating time. Because of the high SS content, the FTR fuel has the lowest throughput rate. Other follow-on LMFBR fuels have a higher throughput than FTR by a factor of 3 to 4. Again, it was assumed that any fuel digestion period would take place in a separate system.

TABLE 3. Estimated Scale Up Capacities for A Six-Barrel Electrolytic Dissolver^(a)

Fuel	kg SS/MT (U+Pu)	MT (U+Pu)/Day	
		2 amps/cm ²	8 amps/cm ²
FTR Driver	1990	0.3	1.2
AI Reference (Core + Blanket)	1189	0.4	1.8
AI Follow-on (Core + Blankets)	813	0.6	2.6
GE Follow-on	502	1.0	4.2

- a. Note, 6 barrels are indicated only because of criticality; otherwise a single dissolver chamber would also handle the capacity as long as the anode area is equivalent. Anode area is 5000 cm²/barrel for a total of 30,000 cm² (32 ft²). Time efficiency of 80% is included in these numbers but the limit of heat generation is not included.

Since normal operation of an electrolytic dissolver has been at 2 amps/cm², a 6-barrel dissolver under normal operation could handle close to 1/4 MT/day of fuel. A 2-barrel dissolver at the higher amperage (if proven practical) would also handle the same dissolution rate for 1/4 MT/day. These numbers hold true as long as charging time does not become excessive and as long as the digestion of residual UO₂-PuO₂ particles is done in another system. If it is done in the electrolytic dissolver, the overall rates will be probably about 1/2 those indicated since about 1/2 of the time can be devoted to SS dissolution and the remainder to dissolution of the oxide.

4.1.2 Heat Limitations

Although there is no precise limitation which the heat load imposes on the capacity of an electrolytic dissolver, overall practical judgments indicate that about the maximum design capacity of a single dissolver should be 1 MT of LMFBR fuel per day. Indications which lead to this conclusion are:

1. Electrolytic dissolvers with a current of 10,000 amperes have been successfully operated on a production scale using current densities of about 2 amperes per square centimeter.

2. Geometric considerations involving criticality and ratios of electrode area to the projected area of a fuel indicate that about 6 dissolving barrels could be used in one dissolver to give a capacity up to 1 MT/day depending on the amount of stainless steel with the fuel.
3. The amount of heat produced in each barrel will be approximately 0.87×10^6 Btu/hour (255 kW assuming 25 volts including approximately 5 kW of fission decay heat in each barrel, i.e., essentially all energy is converted to heat). This quantity of heat, much of which is generated by electrical resistance of the solution, is easily transferred out of the barrel by using a 30°F temperature rise in the solution which is pumped through the barrel at 61 gpm. The circulation rate of 61 gpm could be induced in the tube by an air lift leg having a diameter of 4 inches or larger and by using from 2 to 10 scfm⁽¹²⁾ of gas or alternatively the solution could be pumped. (However, an air lift will strip nitrite from the solution as well as iodine.) If 50% of the cross section in a 6" barrel is available for flow, this would give an acceptable velocity of 1.4 ft/sec. At this velocity (if upward flow) particulates of oxide in the range of 10 to 100 mesh begin to become fluidized. The maximum pressure drop is partially indicated by data on 1/2 inch pellets which exhibit about 1.2 psi/ft of bed if the barrel were completely packed with pellets.⁽¹³⁾ Thus, the 61 gpm of flow or a smaller flow and a higher ΔT appears reasonable.
4. The exchange of heat from the solution establishes the practical limit on the scale-up of electrolytic dissolvers. A heat exchanger, which is cooling liquids with the temperature at 50°C, can transfer about 5000 Btu/hr-ft² to the coolant. If each barrel were attached to an individual exchanger, 0.87×10^6 Btu/hr would require about 175 ft² for each barrel. Customarily heat exchangers have about 15 to 20 ft² of heat exchange area per square foot of exchange cross section and per foot of length. Thus, to keep the exchanger diameter critically safe, nearly a 50 foot long exchanger would be required. Otherwise, a 10 foot

exchanger would need to be 13 inches in diameter which would pose criticality problems. Instead more likely, this would require special designs for criticality such as slab exchangers (e.g., 4" x 36" x 120") or the use of fixed or soluble poisons.

A considerable fraction of the heat could be dissipated by flashing the solution under vacuum. This would be favorable since the particulate plugging or erosion problem would not exist in the vapor condenser as compared with a liquid-liquid exchanger. However, low temperatures (120°F for efficient operations of the dissolver) require a high vacuum and flashing would only remove part of the heat from the system for a practical size unit. Therefore, a dual system would probably be required for removal of heat.

Commercially, vacuum crystallizers and vacuum coolers are used in the range needed but generally are quite large. They operate on a temperature drop of 95°F to 50°F by circulating 8 gpm/ton of refrigeration and use about 28 lbs of steam for evacuation per hour per ton of refrigeration. If this type of unit were attached to an electrolytic dissolver barrel which generates 0.87×10^6 Btu/hr, then the needed circulation rate through the barrel and the vacuum flash chamber would be about 73 gpm.

If some efficiency could be sacrificed or a way is found to operate electrolytic dissolution more efficiently at higher temperatures, e.g., 160 to 185°F then flashing would be a promising method of removing heat.

To put the heat limitation in perspective, a comparison can be made with some designs already in use. For example, the condensers⁽¹⁴⁾ on existing dissolvers range from 0.67×10^6 Btu/hour or about the same as the heat exchange requirement of a single barrel of an electrolytic dissolver to units up to 20×10^6 Btu/hour with about 1300 ft² of exchange surface. This Btu range can be compared with the values listed in Table 4 which would be required for an electrolytic dissolver.

The distribution of heat in an electrolytic dissolver is not uniform. About 15% of the electrical energy is dissipated within the region of the fuel, some 25% is dissipated in the region of the cathode and about 60% is dissipated in the region of the anode. The 15% heat load in the

fuel region along with the fission product decay heat requires at least 15% of the circulation through the region of the fuel particularly if the temperature is to be kept below the melting point of sodium for safe dissolution. This type of circulation is normally less than 2% of the design flow through the fuel during reactor operation. In addition, a major portion of the heat exists at the external portion of the fuel next to the cathode where circulation is good. Circulation here is somewhat independent of particulate plugging of the fuel bundle.

TABLE 4. Electrolytic Dissolver Heat Loads ^(a)

Fuel Characteristic kg SS/MT (U+Pu)	Electrical Load kWH/MT	Heat Load for Processing Rate Indicated (Millions of Btu's/MT)		
		0.5 MT/day	1.0 MT/day	2.0 MT/day
500 ^(b)	27,750	2.0	4.0	8.0
800	44,400	3.2	6.3	12.6
1,200	66,720	4.8	9.6	19.2
2,000 ^(c)	111,200	7.9	15.8	31.6

- a. Based on 25 volts and 0.45 g SS dissolved/amp-hr.
 b. Typical for GE follow-on LMFBR fuel.
 c. Typical for FTR fuel.

Without the aid of a series of detailed case studies, only a general judgment of the limitation of the dissolver system can be made. Near 1 MT/day appears to be about the maximum practical size. It should be recognized that the cooling water requirements of at least 230 gpm for the heat exchange cooling of the system are large because of the low ΔT . However, the cooling water could be used for subsequent condensing service after it leaves the heat exchangers on the dissolver.

The heat load from fission product decay is a relatively small percentage of the total heat load in an electrolytic dissolver and was

generally omitted in Table 4. For example at 30 day cooling, the fission decay heat in fuel @ 0.5 MT/day would be 0.25×10^6 Btu/hr while the electrical heat loss is 2.0×10^6 Btu/hr, or about 12%. Decay heat in the cladding for 30 day cooling is about 5.2 kW/MT or 0.009×10^6 Btu/hr.

The amount of heat generated by a fuel in an electrolytic dissolver as well as the dissipation of electrical energy must be known since the temperature of the solution would normally be kept below 85°C in order to (a) minimize corrosion, (b) keep the sodium solid and (c) to maintain efficiency by avoiding excessive gassing around the fuel pins or the electrodes. The amount of heat was calculated using the RIBD program for the FTR subassembly.⁽¹⁵⁾ Typical values which can be used as a basis for FTR fuels in an electrolytic dissolver are listed in Table 5.

TABLE 5. Decay Power of FTR Driver Fuel Core

<u>Decay Time, days</u>	<u>Decay Power as Percent of Operating Power</u>	<u>Actual Decay Power, kW^(a)</u>	<u>kW/MT^(b)</u>
Shutdown	7.057	383.0	
10	0.1645	9.0	261
20	0.1162	6.36	184
30	0.09345	5.11	148 ^(c)
40	0.07915	4.33	126
50	0.06926	3.79	110
60	0.06198	3.39	98
90	0.04791	2.62	76
120	0.03913	2.14	62
180	0.02807	1.53	44

- a. Based on an average exposure of 45,000 MWd/MT (received in 4 periods of 70.9 days with shutdown of 23.6 days between) and based on an average value of 34.5 kg (U+Pu) per subassembly with a specific power of 5472 kW/subassembly (158.6 kW/kg).
- b. Based on 29 subassemblies per metric ton.
- c. Additional decay heat in cladding is ~5.2 kW/MT at 30 days after shutdown.

4.2 Solvent Extraction Considerations

Information on the relative processing capacity of dissolver solutions originating from a shear leach and an electrolytic dissolver was developed by using the ORNL reference flowsheets⁽¹⁶⁾ as a basis. Rather than develop entire flowsheets here, a comparison of the flows in the first extraction cycle was made by using the feed solution listed in Table 6.

TABLE 6. Electrolytic Dissolver Solution Composition^(a)

	Reactor Source ^(b)			
	FTR	AI Reference	GE Follow-on	AI Follow-on
g SS/MT	1,990	1,140	480	780
Concentrated U	25	64	132	89
Pu	6.7	7.4	11.2	10.0
SS	66	84	72	80
FP's	1.5	2.7	6.3	4.2
HNO ₃	6	6	6	6
Total NO ₃	9.6	10.8	10.8	10.8
Volume, l/MT	30,000	13,500	6,700	9,700

- a. Basis: (1) Final concentration of HNO₃ >6M; (2) Total NO₃ ≥8M; (3) U + Pu + SS <300 g/l Hot and <150 g/l Cold @ 6M HNO₃; and (4) Based on 1 MT of U + Pu charged to reactor (ORNL-TM-3108).
- b. AI Reference, WASH 1099, GE Followon, GEAP 5678, AI Followon, AI-AEC.12792.

For calculations, the dissolver solution was diluted to meet the following criteria:

1. The 1AF feed to first cycle solvent extraction should be no more than 6M NO₃⁻, including HNO₃. (Higher nitrate concentrations may result in excessive Zr extraction.)
2. The 1AF Pu concentration should be less than or equal to 5 g/l.
3. The 1A A/O flow ratio should be no more than 1.0 in the extraction section.
4. All other flows in the 1A, 1BX, 1BS, and 1C columns should be the same as the ORNL reference flowsheet (ORNL-TM-3108) relative to the 1AX flow.

5. The Pu + U concentration in the LAP should not exceed 0.16M (58% solvent saturation, about the same as in the ORNL and Purex flowsheets).
6. Column diameters of 10" for the 1A, 1BX, and 1C columns and 4" for the 1BS column were chosen to operate at maximum volume velocities of 1200, 800, 1200, and 1500 gph/ft², sum of flows, respectively. These capacities are typical for 30% TBP flowsheets but may be on the low side for the ORNL 15% TBP flowsheet. The assumed 1A capacity was deliberately chosen low (about 2/3 normal) to allow for capacity loss with SS impurities such as silica.

Once the above criteria were established for the feed solution, the ratios to the reference flowsheets presented in ORNL-TM-3108 were calculated and shown in Table 7 to get a measure of the limitation imposed by processing the stainless steel along with the uranium and plutonium.

The comparison indicates several significant points:

1. Solvent extraction equipment does not impose any severe limitations on processing solution from an electrolytic dissolver.
2. The liquid waste handling system of a plant does restrict capacity and could reduce the throughput to from 55 to 90% of the shear leach capacity even if no solidification of waste is assumed.
3. The solidification of wastes will be the "bottleneck" for capacity if the solidification equipment is originally sized to match the nominal shear leach processing capacity. If direct coupled waste solidification is used (i.e., without interim acid storage) then the capacity of the plant could be from 30 to 60% of the nominal shear leach capacity.

The columns used as a basis should process 1.3 to 1.5 MT/day on the ORNL shear leach flowsheets and will process from 1 MT/day up to 1.5 MT/day on electrolytically derived feed. The columns are nominally rated at 1 MT/day for LWR fuels and therefore from the viewpoint of solvent extraction, there should be no penalty for processing LMFBR fuels. However, the head end requirements, the waste disposal requirement and

TABLE 7. Calculated Effects of Electrolytic Dissolution on Capacity
(Percent of Shear Leach Capacity)^(a)

	<u>FTR</u>	<u>AI Reference</u>	<u>AI Follow-on</u>	<u>GE Follow-on</u>
1. Solvent Extraction System (based on column throughput limits on first A and B column).	40	80	100	100
2. Volume of 1AW (used as a measure of Waste Evaporation Capacity). ^(b)	30	60	80	90
3. Moles of HNO ₃ /MT (as a measure of Acid Fractionator or system without solidification).	25	55	80	110
4. Total NO ₃ handled/MT (as a measure of Waste Solidification Capacity).	15	30	45	65

a. All numbers were compared to values in ORNL-TM-3108 and rounded to nearest 5.

b. Cr⁺⁶ in electrolytic dissolver solution precludes evaporation in SS equipment.

plutonium requirement are more restrictive than the mainline solvent extraction system. The limitation, i.e., ~30% of shear leach imposed by basing the LMFBR waste capacity on total nitrate, is less restrictive than basing capacity comparison on a volume of solids basis where the capacity could be 10% of the shear leach waste solidification capacity.

4.3 Waste Considerations

Although many factors are involved in the evaluation of electrolytic dissolution, one important consideration is a comparison of the high level waste generated by electrolytic dissolution with the waste from other processing options. Study data on the FTR fuel were developed as one example of an LMFBR. Generally values for fuels are proportional to the value shown here for FTR fuels.

Since the stainless steel dissolved by electrolytic dissolution eventually becomes part of the solidified waste, the volume of solidified waste provides one basis for determining the amount of mechanical cutting operations, if any, which may be desirable before dissolution.

Normally, processing of an LWR fuel by a chop-leach operation produces a volume of the total hardware for disposal nearly equal ($\pm 10\%$) to the overall volume occupied by the original fuel. Typically, the volume of LWR hulls and hardware after a chop-leach operation averages from 12 to 15 ft³ per metric ton of fuel.

Since LMFBR fuels contain much more hardware than LWR fuels, use of a final hardware volume equal to the initial fuel volume is probably optimistic unless a compaction operation is conducted. By using an equal volume basis as an estimate for FTR fuel, nearly 50 ft³ of hardware will be generated per metric ton of U-Pu or approximately 3 times the volume of an LWR fuel for an equal tonnage (metal content basis).

Table 8 shows the waste volumes possible from FTR fuel and the changes in total waste volume for disposal which would occur by various choices on the amount of fuel disassembly prior to electrolytic dissolution. Normally electrolytic dissolution of a complete FTR sub-assembly may generate about 3 times the original volume of the

subassembly in wastes as calcined stainless steel oxides. This volume can be reduced to below 2 times the volume of the original subassembly by cutting off the plenum section along with the flow duct around the plenum section and the bottom end of the subassembly (i.e., 2 cross section cuts/subassembly).

TABLE 8. Volume of Solid Waste Expected from One Metric Ton of FTR Fuel, Cubic Feet (Rounded to Nearest 5)

Operation with:	kg SS/MT	Chop Leach ^(a)	Calcine ^(b)	Calcine plus Hardware ^(c)
1 Complete Subassembly	2900	50	125	125
2 Ends Removed without penetration of plenum)	1787	35	85	100
3 Ends and Total Duct Removed	1250	30	55	75
4 Ends, Plenum and Associated Duct Removed	1325	20	55	85
5 Ends, Complete Duct, and Plenum Removed	870	15	35	70

- a. Assumes hardware from chop leach occupies same volume as original fuel. Number indicates volume left after the indicated operation is complete.
- b. Assumes conversion of all metal to oxides with a bulk density of 1.2 g/cc. Note, this does not include fission products from fuel.
- c. Combined volume of hardware removed and hardware dissolved and calcined.

If it is assumed that the cladding hulls from a shear-leach process for FTR fuels must be shipped to a burial site in the same manner as other high level waste, the number of containers of waste provides one basis for comparison of the shear-leach process with electrolytic dissolution. Since two cross section cuts of the FTR subassembly appear to be the most likely mechanical operation, they are assumed as a reference basis for comparing the solid waste from processing. This permits disposal of the bottom and the plenum section separate from other wastes.

From the viewpoint of solid waste management, processing spent FTR fuels (45,000 MWD/MT @ 158 MW/MT) may require one of the following process routes for the cut fuel section if processing is to be conducted about 30 days after discharge of the fuel from the reactor.

1. One metric ton of fuel (U,Pu) can be dissolved electrolytically and processed by solvent extraction, but the aqueous waste must be aged about 6 months to reduce the decay heat before the waste can be solidified into two 24-inch diameter containers (9-foot fill height and cooled in water). A diameter of 24 inches is about the largest practical size for shipping. Also, this size is required to hold the volume of ceramic which will contain the stainless steel and fission products. If the aqueous waste is aged >1 year, a single larger container could hold the waste if a larger container could be shipped.
2. The stainless steel cladding could be dissolved electrolytically while possibly less than 25% of the fission products and fuel dissolve if the dissolver solution is kept below 50°C during dissolution. The aqueous waste from solvent extraction of this "cut" could be solidified directly as 2 containers of ceramic at the time that one metric ton is processed. The remaining 75% of the fuel could be dissolved and processed separately. Solidification of the resulting waste will produce slightly less than one additional container of ceramic about 12 inches in diameter (9-foot fill) if the age of the waste is about one year.
3. The fuel could be sheared, leached with acid and processed by solvent extraction. Slightly less than one container (24-inch diameter and 9-foot fill) will be required for the hulls. Solidification of the waste from solvent extraction will require slightly over one container per metric ton (<12 inch diameter) if the waste is aged about one year before solidifying.

Solidifying the waste from either electrolytic dissolution or shear leach processing after 30 days from fuel discharge will require about

6 containers of waste per metric ton of fuel in order to dissipate decay heat. The diameter of the containers used for electrolytically derived waste must be larger than those from shear-leach derived waste. Since about one container is required to handle the cladding hulls from a chop leach process, the number of containers for shear leach is always one more than for electrolytic dissolution unless the hulls are deliberately placed in the waste container at the time of solidification. This holds true as long as the dissipation of heat is limiting. As soon as the waste is aged longer than one year, the practical diameter of 24 inches becomes limiting. After one year the same number of containers is required for either process. The only difference being that electrolytic dissolution requires larger containers.

In order to determine the above effects of different process routes and the associated fission product heat, the following options were considered to determine the relationships between waste and the fuel processing method.

1. Electrolytic dissolution of the minimum amount of stainless steel after complete dismantling of the FTR fuel to remove the duct, ends and plenum section.
2. Mechanical removal of the bottom end and the plenum section of the FTR fuel by making two cross section cuts which leaves the flow duct intact around the core.
3. Electrolytic dissolution of the complete FTR subassembly without any mechanical preparation.
4. Shear leach without dissolution of stainless steel.

The corresponding volumes of waste for each option are given in Table 9. Note that, in general, the total volume of waste including ceramic and hardware, is about a factor of 2 higher for electrolytic dissolution as compared to a shear leach process. Although boron or other nuclear poisons may be used in LMFBR flowsheets, quantities have not been included at this time because flowsheets are not firm and because boron can be used as a substitute for a major portion of the sodium additive needed to make a ceramic. Similarly, cadmium can be considered as a substitute for iron in arriving at a recipe for a ceramic solid.

TABLE 9. Volume of Waste from FTR Spent Fuel Processing
(Rounded to nearest 5)

<u>Option</u>	<u>Volume Ceramic Solid, ft³/MT^(a)</u>	<u>Volume of Ceramic and All Hardware, ft³/MT</u>
A Minimum SS/UPu ratio	50	80
B Bottom and plenum end removed	60	90
C Maximum SS/UPu ratio: dissolution of whole subassemblies	130	130
D Shear Leach	5	55 ^(b)

- a. Volume of ceramic is based on a bulk density of 2.9 kg/l with a ceramic weight percent composition of 13% Na₂O, 46% P₂O₅, 40% SS oxides and 1% FP oxides. (LMFBR wastes used in WSEP(5) are 11% Na₂O, 42% P₂O₅, 19% SS oxides and 28% FP oxides. This was used for option D.)
- b. Cladding hulls volume from shear leach of fuel portion from option B is 20 ft³/MT.

Since a comparison of waste volumes is not meaningful without considering the heat content of the waste (Table 10), the volume of waste was converted to the number of thermally safe containers per metric ton.

TABLE 10. Decay Heat in FTR Fuel^(a)
(Rounded to nearest 5)

<u>Option</u>	<u>kW/MT</u>
A Minimum SS/UPu ratio	155
B Bottom of plenum end removed	155
C Maximum SS/UPu ratio: whole subassemblies	165
D Shear Leach	10 ^(b)

- a. Decay heat in stainless steel was estimated to be 5 to 8 kW/MT in core region. Thus total fuel decay heat was estimated at 165 kW/MT 30 days after discharge from an average exposure of 45,000 MWD/MT at 159 MW/MT.
- b. The stainless steel remaining after the bottom end and the plenum have been removed by two cross section cuts.

Thermally safe in water, as defined here, is the maximum decay power which will give an 800°C temperature difference between center line of a container and the wall in water when the thermal conductivity is 1.0 Watt/M-°C (.059 Btu/hr ft²-°F/ft) for ceramic and 0.35 Watt/M-°C (0.2 Btu/hr ft²-°F/ft) for calcine. Likewise, thermally safe in air, is the maximum decay power which will give 500°C temperature difference between the center line and the container wall when it is cooled by air. Values for kW/ft of container were used as a reference because once the temperature difference and the thermal conductivity are specified, the value of kW/ft is independent of diameter of the container. In other words, the container will only dissipate a certain amount of heat per foot of length. The reference value used for a ceramic cooled in water was 3.05 kW/ft and for calcine, 1.07 kW/ft. For air cooled calcine, the thermally safe value was taken as 0.67 kW/ft. Use of these values for kW/ft is somewhat more conservative than values which have been measured in the Waste Solidification Engineering Prototypes* (WSEP) at this laboratory site. However, the wastes from electrolytic dissolution of FTR fuels are quite different from those used in WSEP. Values in WSEP for air cooled containers have been as high as 2.2 kW/ft for ceramic and 1 kW/ft for calcine.

As a general rule, the volume of calcine is about equal to the volume of ceramic, but the thermally safe value for calcine is about 1/3 that of ceramic. Thus under heat limiting conditions of solidification within one year there can be 1/3 the number of containers of ceramic as calcine.

The minimum number of thermally safe containers which can be produced from a particular waste is simply the relationship between decay power and the allowable kW/ft as shown in Figure 4. Some specific values of interest are listed in Table 11. Since these are independent of diameter, the diameter must be adjusted to give the fill volume which produces or is below the allowable kW/ft in a practical size. For example, about 24 inches in diameter is probably the practical limit for hauling and interring a container in a salt mine. Thus, the number of containers of aged wastes is set by the practical diameter if the decay power has subsided sufficiently to permit economical burial in a salt mine.

*

W. R. Bond, et al. Waste Solidification Program, Volume 8, "Spray Solidification Performance During Full Radioactivity Tests in Waste Solidification Engineering Prototypes," BNWL-1583, June 1970.

FIGURE 4: Number of Waste Containers Required to Obtain Thermally Safe Storage for Various Fission Product Decay Power Levels. (Based on K of Ceramic of 1.0 Watt/M-°C ΔT in Water of 800°C, in air 500°C and K of Calcine of 0.35 Watt/M-°C. FTR Fuel at 45,000 MWD/MT and 158 MW/MT.)

TABLE 11. Minimum Number of Containers of Waste per
Metric Ton^(a) to Dissipate Decay Heat

Option	100% fp's ^(b) 30 day cooled	25% fp's >2 yr aged	100% fp's >6 yr aged
A Minimum SS/UPu ratio	5.7	1.4 (1.9)	<0.2 (1.9)
B Bottom and plenum end removed	5.7	1.4 (2.2)	<0.2 (2.2)
C Maximum SS/UPu ratio	6.1	1.5 (4.8)	<0.2 (4.8)
D Shear Leach of Option B	6.7 ^(c)	2.4 ^(c)	<1.2 (<0.2) ^(c)

- a. The numbers given do not reflect any consideration of diameter. Since about 24 inches may be the practical limit for hauling and placing aged waste in a salt mine, this diameter becomes restrictive for aged SS wastes. The numbers in parentheses indicate the number of 24-inch diameter containers where the diameter could be larger than 24 inches on the basis of dissipation of the decay heat.
- b. Decay heat values from Table 7.
- c. One container for hauling hulls as high level wastes is included in the total shown.

At a specific decay power level, the diameter must be set to meet the allowable kW/ft for thermally safe conditions. Diameter is shown in Figure 5 as a parameter for relationships between kW/MT and volume in ft³/MT. The relative positions of options A, B, and C are shown. However, generally a diameter will be selected and the kW/MT decreased by aging of the waste as required to meet the selected diameter. To show a comparison, specific diameters are indicated in Table 12 for water cooling. For 6-year aged waste, the limit imposed by the salt mine requirements can become controlling depending on the choice of time for storage and shipment.

FIGURE 5: Decay Power vs Volume of Ceramic Waste at Selected Thermally Safe Diameters with Water Cooling

TABLE 12. Thermally Safe Diameters of Waste Containers in Water

Option			Decay Power, kW/MT	Ceramic Volume ft ³ /MT	Diameter, inches
A	Minimum SS/UPu ratio	30 day 100% FP	155	50	14
		30 day 25% FP	40	45	23
		>6 yr 100% FP	2.5	50	>100*
B	Bottom & plenum end removal	30 day 100% FP	155	60	15
		30 day 25% FP	40	60	30*
		>6 yr 100% FP	2.5	50	>100*
C	Maximum SS/UPu ratio	30 day 100% FP	165	130	21
		30 day 25% FP	40	130	43*
		>6 yr 100% FP	2.5	130	>100*
D	Shear-Leach	30 day 100% FP	165	5	< 6
		2 yr 100% FP	16	5	< 12
		>6 yr 100% FP	2.5	5	30*

*

All of these values exceed the practical diameter for shipping and burial, e.g., 24" diameter with a 9-foot fill contains 5100 lb of ceramic. Thus, the number of containers is determined by the volume a 24" diameter will hold and not by decay heat.

4.4 Material of Construction Considerations

There are no known special material problems introduced by processing LMFBF fuels. Material considerations apply as outlined under Section 3.0, State of Development for LWR Fuels. The need for dissolvents other than nitric acid has not been determined for LMFBF fuels. If additives such as fluoride were used, their corrosive effect on the separate dissolution equipment as well as potential distribution throughout the plant must be considered. The expected detrimental effects of fluoride on platinum precludes the use of fluoride directly in the electrolytic dissolver for dissolution of mixed oxides.

4.5 Special Considerations

4.5.1 Undissolved Solids

During the electrolytic dissolution of stainless steel, a portion of the metal remains as fine metal particles mostly originating from

grains of stainless steel released by intergranular corrosion during the dissolution process. The amount apparently varies with the design of the dissolver. From 3 to 8% of the weight of stainless charged to a dissolver has been found as undissolved solids. The small particles of undissolved solids range from about 80% as -200 mesh (<74 micron) in the dissolver solution to ~90% -325 mesh in the dissolver vessel.⁽³⁾ The metallic particles readily settle or they can be separated by centrifugation.⁽³⁾ Uranium loss to the solids is .001 to .002% of total uranium.

4.5.2 Dissolver Sludge and Electrode Fouling

In addition to the fine metallic solids, silicious materials also remain insoluble in electrolytic dissolver solution and plate out on metallic surfaces, e.g., electrodes, heat exchangers, etc. The current carrying capacity of electrodes is reduced by as much as 30% as the electrodes become fouled. Several chemicals have been tested to clean dissolver surfaces; dilute sodium hydroxide solutions were found effective.⁽³⁾ Sufficient data are not available to indicate the probable frequency of electrode cleaning. Every one to two weeks is anticipated. The factors affecting deposition or the prevention of deposition warrant further work if electrolytic dissolvers are going to be used extensively.

4.5.3 Dissolver Cycle Time

The fuel charging concept used for design can affect the overall time cycle. The usual procedure is to have the power off while batch charging the dissolver. As a result, the dissolution is inefficient. Some improvement can be made by using gravity feed of fuel bundles. However, experience and projections indicate that about 80 to 85% of the time can be used for power on. The remaining time involves charging, chemical additions, solution transfer, etc.

In addition to the regular time cycle, complete clean out of a dissolver can be time consuming if cleaning is needed. The dissolution of residual pieces of stainless steel in a partially filled dissolver is inefficient, simply because the acid surrounding the pieces conducts

current between the electrodes. Lowering the liquid level during clean out is helpful. Because the major concern will be complete recovery of fissile material from a campaign, cleanouts will be probable for each fuel type.

4.5.4 Criticality

The high concentration of plutonium (15 to 25%) in LMFBR fuels makes design of critically safe dissolvers mandatory. Although dissolution data on LMFBR fuels is being extensively investigated^(17,18) preliminary data (see 4.1, Projected Capacity) indicate that the uranium dissolves faster than plutonium and both fine and coarse particulate plutonium particles exist in the solution. The coarse particles easily settle. Until data exists to the contrary, it must be assumed that particles can settle in the dissolver and any location where particles may settle must be geometrically safe.

Since slab or annular concepts will not permit wide enough gaps to accept nuclear fuel without the use of poisons, individual barrels or tubes will probably be used. The usual nuclear factors must be considered for spacing and size of tubes. For conceptual scale up purposes in this report, six equally spaced tubes on the corners of a hexagon were used.

4.5.5 Metallic Sodium

In liquid metal fast breeder reactors, the fuel resides in metallic sodium throughout its reactor history; it is initially cooled in sodium and may even be shipped in sodium. The presence of metallic sodium introduces a new material which must be handled in the reprocessing plant. Although techniques for removing sodium are being developed and used primarily for reactor fuel inspection, they do not necessarily assure that sodium is removed from defective or vented pins. Therefore, any electrolytic dissolver must be either capable of accepting sodium or a processing step must be completed to assure the absence of sodium prior to dissolution.

Early in the power reactor development programs some data was obtained on sodium bonded fuels. Tests at Savannah River used glass

vials of sodium (10 to 30 g each) which were shattered in acid, and copper tubes containing sodium were dissolved in 195°F nitric acid.⁽¹⁹⁾ At this temperature, the reactions were violent and sodium oxide was expelled from the solution. Later, tests⁽²⁰⁾ at reduced temperatures were more favorable even when rods containing sodium were pressurized to expected fission gas pressures of 500 psig. The sodium stayed in the rod as a solid. Further tests oriented toward the dissolution of Hallam fuel showed that 900 grams of sodium could be handled in an electrolytic dissolver if the temperature is kept at <140°F and the rate of dissolution of sodium is no higher than about 1.2 g/min. However, to date the Hallam fuel has not been dissolved at SRP.

In other tests at Hanford⁽²¹⁾ it was concluded that the extremely violent reaction of Na and/or NaK with nitric acid prevents the charging of NaK-bonded fuels directly in a chemical dissolver. A planned sudden exposure of 22 grams of Na charged to <700 pound dissolver containing cold 60% nitric acid generated sufficient force to displace the dissolver while generating a peak hydraulic pressure of 850 psig. Other tests showed the shock waves were much less severe if the submergence in liquid was minimized. Other tests⁽²²⁾ with water showed up to 40 grams of NaK could be exposed under water at one time without damage but only 10 grams of NaK was preferable. Data indicated the dispersion of liquid metal was important; if well dispersed, up to 2.4 g/sec could be reacted. Otherwise, only about 2 to 3 grams would react safely per minute.

Adams⁽²³⁾ has summarized many of the problems in processing liquid bonded fuels and has recommended a mechanical approach for handling sodium rather than chemical.

Several factors make the dissolution of sodium in fast fuels more favorable than early tests on the Hallam or on the LWR fuels. The pins of a fast fuel are small. Only failed pins are of concern rather than sodium filled pins. The probability of defects is low and the amount of sodium in a defective pin is expected to be small, e.g., 0 to 7 grams of Na in a failed pin.

Recent tests at ICPP on EBR II fuels* using a horizontal electrolytic dissolver confirmed that small quantities of sodium can be handled safely. Each of 6 SS-clad EBR II rods ~14 inches long and 0.144 inches in diameter had about 0.7 grams of sodium inside along with the metallic fuel. At no time did the undiluted hydrogen concentration (1.4%) in the gas exceed the lower explosive limit of hydrogen (~4% if dry, >4% if wet). Sodium in the reservoir end as well as the bond dissolved smoothly at a dissolver temperature of 150°F without any evidence of a violent reaction.* The overall sodium content was .01 kg/kg U and the 304SS was 0.5 kg SS/kg U after removal of the blanket ends. (For comparison FTR contains 0.9 to 2.9 kg SS/kg U+Pu depending on the extent of disassembly.)

Because the dissolution tolerance to sodium has not been clearly defined, particularly for kinetics and heat transfer, generally there will be a reluctance to dissolve fuel containing a total inventory of a few grams (e.g., >5 grams) of sodium primarily because the potential hazard exists under inadvertent overheating to melt the sodium and rapidly expose it. The principal difficulty then arises as to how to assure the metallic sodium inventory which might enter a dissolver is low. Considerable development work will be required to define either measurement methods and/or process steps which will deactivate the sodium.

Electrolytic dissolution appears to have some advantages for handling metallic sodium because of the limited rate of exposure of sodium to acid during dissolution and because the low temperature operation which, with proper heat exchange from dissolving sodium to the solution, will safely react the sodium. However, from the available data one cannot generally state at this time that electrolytic dissolvers can handle sodium in defective LMFBR fuels. Sufficient test work could be accomplished to better define the safe limits if sodium removal techniques prove to be impractical.

*

Communication with R. A. Brown and G. F. Offutt, Idaho Nuclear Corporation.

4.6 Capital Cost

In order to complete a perspective assessment of electrolytic dissolution and its capability for extension to LMFBR fuel processing, an engineering estimate of the probable differences in capital costs was developed. More definitive engineering criteria are required for a complete and accurate estimate; however, for the present state of development for LMFBR fuels, simplifying assumptions were used. The capital costs of electrolytic dissolution for LMFBR fuel is economically competitive with shear leach costs only if the processing rate is 1 MT/day or less. For higher capacities, the shear leach capital costs are less than electrolytic dissolution.

The estimate was made on the basis that there would be an equal number of shear leach dissolvers or electrolytic dissolvers in a plant and that they require the same cell space. The incremental capital costs for electrolytic dissolution were estimated for the electrodes, electrical supply, heat exchangers and ancillaries for the electrolytic dissolver over and above the expected cost of a shear leach dissolver. In addition, a capital cost penalty for electrolytic dissolution was imposed for downstream capacity by requiring titanium evaporators for all capacity and requiring incremental waste solidification capacity for the larger volume of electrolytic dissolution wastes.

The cost range of shears for present day reprocessing plants is \$500K to \$1000K* depending upon customer specifications, e.g., all stainless steel, manipulating devices, etc. By using the normal total installed cost of 2.5 x direct purchase cost for mechanical equipment, the cost range for a shear can be \$1250K to \$2500K complete. A single shear usually has adequate capacity for presently anticipated reprocessing loads. Since the shear is the single item that would not be required in an electrolytic dissolver head end, the incremental shear capital costs and the incremental electrolytic capital costs can be compared roughly as shown in Table 13.

* Communication with Birdsboro Corporation, Birdsboro, Pennsylvania.

TABLE 13. Incremental Plant Equipment Capital Costs Attributable to Shear Leach and Electrolytic Dissolution

kg SS/MT (U+Pu)	Incremental Capital Costs per MT/day of Electrolytic Dissolution Capacity, K\$	Comparable Shear-Leach ^(a) Incremental Costs for a Shear Independent of Capacity to Greater Than 5 MT/day, K\$
500	1960	1250 to 2500
800	2570	1250 to 2500
1200	3290	1250 to 2500
2000	4920	1250 to 2500

- a. This cost range and capacity is for LWR-type shears and does not include any special modifications for LMFBF fuel; however, these are expected to be within the indicated cost range.

In particular, for processing 1 MT/day or less of LMFBF fuel the capital cost of electrolytic dissolution is comparable with shear-leach if the amount of stainless steel with the fuel is 500 kg/MT (U+Pu) or less. For LMFBF's, the GE follow-on design* approaches this condition, i.e., <500 kg SS/MT (U+Pu) after end hardware removal. For FTR fuel which has a flow duct around the fuel rods, this condition can be approached only by removing the duct and cutting of the plenum section of the rods to reduce the stainless steel to about 870 kg/MT (U+Pu). A much more favorable cost situation for electrolytic dissolution exists for LWR fuel since in LWR fuel the metal (Zr or SS) normally ranges from 250 to 350 kg metal/MT (U) before end fittings are removed. [Incremental capital costs for electrolytic dissolution range from \$3K to \$4K per kg SS/MT (U+Pu) in the fuel for each MT/day of capacity.]

A breakdown of the relative components of incremental costs for electrolytic dissolution is shown in Table 14. The relative percentage of the total incremental cost in Table 14 attributable to each component is interesting. Some 75% of the capital cost is involved in waste handling and disposal as shown on page 40.

*

W. R. Gee, Jr. et al. GEAP-5678, December 1968.

TABLE 14. Incremental Cost Components for 1 MT/Day Electrolytic
Dissolution Relative to Shear Leach

kg SS/MT Fuel ^(a)	Dollars in Thousands				
	Power and Electrode ^(b)	Heat Exchange ^(c)	Waste Evaporation ^(d)	Waste Solidification ^(e,f)	Total
500	500	70	250	1140	1960
800	670	110	260	1530	2570
1200	900	170	290	1930	3290
2000	1370	280	640	2630	4920

- a. 500 = ~GE Follow-on, 800 = ~AI Follow-on, 1200 = ~AI Reference, 2000 = ~FTR, all with end hardware removed.
- b. Platinum cost of \$110/Troy oz. for 10 mil sheet. Electrode area and power based on 2 amps/cm² and 0.45 g SS dissolved/amp hr. Fabrication cost of electrode = 0.5 pt cost, power supply @ \$70/kW.
- c. Based on a reference value for a normal dissolver with 50 ft² of heat exchange per MT/day. Increment above 50 ft² assessed for electrolytic requirements and a direct purchase cost of \$30/ft² of heat exchange surface.
- d. New titanium evaporator is assumed for all waste capacity @ \$2000/ft² of steam-liquid heat exchange area.
- e. Based on ~20 kg SS/ft³ of solidified waste/MT (U+Pu) and a scale up exponent of 0.6. Costs are incremental increase in waste solidification capacity relative to a unit processing 5 ft³ of ceramic-glass-solid/day/MT. Equipment affected by capacity was scaled up relative to \$437,000 as direct purchase costs for 5 ft³/day/MT.
- f. For purposes of cost comparison, the cost of storage facilities for cladding hulls was assumed to be the same as the incremental cost of on site storage space for larger containers of high level waste from electrolytic dissolution. The incremental cost of somewhat larger waste containers for electrolytic dissolution is considered an operating cost rather than a capital cost.

<u>Component</u>	<u>Percent of Total Cost (Low kg SS - High kg SS)</u>
Power and Electrodes	26 to 28
Heat Exchange	4 to 6
Waste Evaporation	13
Waste Solidification	57 to 53

4.7 Typical Flowsheet

A typical study flowsheet for FTR fuel is shown in Figure 6. Flowsheet quantities were set to meet an end acidity of 6M. Digestion of sludge is assumed in a separate step and the acid is combined with dissolver acid (no fluoride). As shown, the final dissolver solution required dilution to at least 6M total nitrate concentration before solvent extraction in order to provide reasonable zirconium decontamination.

Not shown on the flowsheet is a batch requirement for dissolver clean up particularly of electrodes. At least 5% NaOH is required at boiling for approximately 2 hours.

4.8 Future Development Requirements

From the survey of electrolytic dissolution, several development requirements are evident.

1. Capacity--From the experience on electrolytic cutting of metal using a flowing electrolyte, increasing the rate of dissolution by increasing the current flow appears feasible if the electrode is designed for acid flow through the electrode. Essentially it may be possible to increase the effective surface area of the electrode per unit surface of electrode. At the same time the flow of acid can reduce the effect of heating at the electrode.
2. Fouling--Further work on the phenomena of plating or fouling of the interior of the vessel and the electrode surfaces is needed to clarify plutonium losses. Simulated or actual fission products should be included in the study.

FIGURE 6: Study Material Flowsheet - Electrolytic Dissolution of FTR Fuel

3. Sodium Tolerance--Some uncertainty exists on the ability of electrolytic dissolution to accept sodium which may be logged within a fuel pin. Tests under high heating conditions typical of LMFBR fuels are required before operation of a full scale dissolver with potentially logged pins.

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