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CONTINUOUS OR SEMICONTINUOUS LEACHER
FOR LEACHING SOLUBLE CORE MATERIAL FROM SHEARED
SPENT NUCLEAR FUEL TUBES

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CONTINUOUS OR SEMICONTINUOUS LEACHER

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Abstract

There is no known existing device applicable to the effective dissolution of sheared liquid-metal fast breeder reactor fuels. New concepts and models representative of current approaches to the continuous or semicontinuous dissolution of these fuels were developed to assess the magnitude of the difficulties to be encountered in controlling the flow, residence time, agitation, and transfer of the sheared material through such a device. The performance of many of the models developed was successfully demonstrated, but the model of the compartmented rotary drum continuous leacher with sloped transfer ducts is representative of the most effective and straightforward design required to meet all of the performance requirements.

Introduction

The results of surveys^{1,2} of all known existing equipment for the dissolution of sheared nuclear power reactor fuel revealed that an effective device applicable to the dissolution of sheared liquid-metal fast breeder reactor (LMFBR) fuel of the type currently planned does not exist. That dissolution equipment of the type currently used to process spent fuel from conventional light-water-cooled nuclear power reactors is not suitable for processing spent fuel from liquid-metal fast breeder reactors is supported by four major factors.

One, an economic LMFBR fuel cycle demands a short turnaround time, and this means that the fuel must be processed soon after its discharge from the reactor. The fuel cooling time will be so short (perhaps as short as 30 days) that the presence of short-lived fission products may possibly cause serious heat dissipation problems in the dissolution equipment. Heat generation values as high as 200 W/kg of fuel can be expected.

Two, the plutonium content of LMFBR fuels is significantly greater (20 to 25%) than that of the currently used nuclear power reactor fuels, thereby necessitating a careful study of nuclear criticality in the dissolution equipment. The dissolver environment must also be severe enough to assure complete dissolution of the plutonium content of LMFBR fuel.

Three, because of high specific power requirements within the reactor, the LMFBR fuel rods are much smaller in diameter than those used in conventional light-water-cooled reactors. These small-diameter rods result in a high cladding-to-fuel-weight ratio that must be accounted for in the dissolver.

Four, current economic considerations are producing a trend toward larger processing plants and are indicating the need for an increased use of automated continuous equipment.

During the nearly two decades that the nuclear power industry has been in existence, little effort has been expended on the design of continuous or semicontinuous dissolvers, while much has been directed toward establishing the technology of bath dissolution, especially as applied to commercial power reactor fuels. Unfortunately, even these efforts were frequently directed toward a specific fuel design rather than a broad class of fuel types. Nevertheless, several worthwhile concepts and designs of dissolvers have emerged. Those concepts and designs of dissolvers that are applicable to a method of processing sheared or chopped LMFBF fuel in which the cladding remains essentially intact are the subject of this discussion.

Most of the concepts of dissolvers especially applicable to LMFBF fuel that are discussed herein involve continuous or semicontinuous operation. These concepts form a base from which additional development can provide the required reliability of operation, assurance of criticality and temperature control, and the certainty of applicability to a variety of LMFBF fuel types (oxide, carbide, nitride, and bonded and unbonded fuels) essential to achievement of the objectives of the LMFBF program. These objectives are (1) application of the present aqueous processing technology to LMFBF fuels, (2) development of new technology that will permit adaptation of existing facilities to LMFBF fuel processing, and (3) design of new facilities to meet long-term processing needs.

Shear-Leach Process

At the present, the aqueous method of processing appears to be the most widely accepted. It is the method used in the first commercial processing facility in the United States: Nuclear Fuel Services, Inc., in West Valley, New York. This method has also been chosen by the United States Atomic Energy Commission as the most likely one to be used for processing LMFBR fuel. Within the overall scope of aqueous processing, the practice of segmenting fuel by shearing and subsequent dissolution of the core material from within the segments of fuel cladding is known as the shear-leach process. Its capability for adaptation to a wide variety of fuel materials, sizes, and configurations and its proven reliability make this process the obvious choice for use in the head-end operation of LMFBR fuel reprocessing.

In the shear-leach process, spent fuel from nuclear power reactors is mechanically reduced to a size compatible with the geometry and dissolution conditions of the dissolver to be used. Frequently, the fuel assembly is taken apart by removing the end hardware and then removing the fuel rods from within the sheath surrounding them. The sheath can be slit or sometimes the rods can be pulled out of the sheath. The fuel rods can then be sheared either singly or in bundles. It is also possible to shear the entire assembly directly, but this procedure adds to the burden of non-nuclear material to be handled by the dissolver. In existing facilities, the sheared material is collected in a container which is eventually loaded into a batch type of dissolver.

The product produced during the shearing of LMFBR fuel is comprised of short tubular sections partially filled with core material, short

lengths of small-diameter spacer wires, and core fines which have been dislodged from the fuel tubes (about 50% when 1-in. lengths are sheared). If the entire assembly is sheared, fragments of the sheath will also be produced.

The design of the dissolver must provide for proper exposure of the fuel to the dissolvent. This implies control of the residence time of the fuel in the dissolver and the temperature in the dissolver. In addition, there must be provisions for exposing new fuel surfaces by agitation and for collecting off-gases for disposal or reflux. In continuous equipment, separate controls for agitation of the fuel and for advancement of the fuel through the vessel appear to be desirable.

New concepts of devices for dissolving LMFBR oxide fuels must also account for the increased quantity of off-gas generated by these high-burnup fuels to alleviate possible foaming conditions in the equipment. Because of the high heat generation rate exhibited by LMFBR fuels with short cooling times, it is probable that heat will have to be removed, rather than added, to control the dissolution rate. It is also likely that a greater proportion of the core material will be dislodged from the cladding because of its high-burnup history, its subjection to a high-temperature oxidation treatment prior to dissolution (to remove sodium and fission-product gases), its short shearing length, or a combination of these reasons. The resulting core fines will undoubtedly react vigorously with the dissolvent.

Early Dissolver Designs

Prior to the development of the shear-leach process, which dates from 1957, considerable work had been done on dissolution as applied to both the batch and the continuous dissolution of uranium metal and uranium-aluminum-alloy fuels. This work contributed to subsequent efforts made to design devices for dissolving sheared fuel because many of the parameters had already been investigated. The terms "dissolver" and "leacher" are used interchangeably in this discussion of early designs, but the term "leacher" is a more appropriate designation of devices used for sheared fuel.

Typical dissolvers from this early period include batch or continuous pot dissolvers, upflow column dissolvers, downflow or trickle column dissolvers, and slab dissolvers. Generally, the latter three types of dissolvers were operated continuously. However, these units are not suitable for modern processing because, in addition to other factors, they have no provisions for the removal of inert cladding materials that enter with the feed.

At least two early dissolvers were designed for fuels with a sizable amount of inert material to be removed.³ One was a critically safe rotary-slab dissolver or "Persian wheel" for continuous charging and discharging. The other was a spiral-vibrating-tray design in which the fuel was to be fed to the bottom of the unit through a center pipe, vibrated upward along a spiral path flooded with dissolvent, and discharged near the top. This design was intended for use with unclad or aluminum-clad fuels. Although the first of these two dissolvers was

built and apparently operated successfully, additional testing showed that some buildup of solids eventually occurred on the walls of the unit. Two other disadvantages that make this design unsuitable for processing LMFBR fuels are that it is not amenable to air sparging to provide additional agitation and the collection of off-gas would be difficult. The second dissolver, the vibratory model, was never constructed.

Designs for several continuous dissolvers for sheared fuel were developed at Oak Ridge National Laboratory (ORNL). A vertical vibrating spiral tray describes the working mechanism of one of these continuous dissolvers that was akin to the previously discussed design intended for use with unclad or aluminum-clad fuels. A unit for use with sheared fuel was fabricated, but preliminary tests were unsuccessful because of repeated mechanical failure brought about by the vibrations. If this problem could be overcome, the vibrating-spiral-tray dissolver would be a viable concept for application to LMFBR fuel reprocessing.

A type of continuous dissolver that has received considerable attention is the inclined spiral-screw dissolver. Two working models of this type of dissolver to be used for UO_2 and UO_2 - ThO_2 fuels were fabricated and tested at ORNL. The first model, illustrated in Fig. 1, consisted of an enclosed screw which rotated inside a cylindrical housing. In this unit, nitric acid is introduced at the upper end and is discharged into the fourth flight to cascade down through the remaining flights. As the screw is rotated, the fuel is advanced countercurrent to the flow of acid. The use of perforated areas on the inner drum and baffles on the outer drum permitted zoning of the treatment into dissolving and rinsing sections. This unit was operated successfully and was quite

Fig. 1. Continuous, Enclosed-Screw Rotary Leacher.

effective in the dissolution of UO_2 fuels but it did not provide the necessary agitation required for the slower dissolving UO_2 - ThO_2 fines. Other disadvantages included poor heat transfer from the inner cylinder to the outside housing and a lack of vapor space to accommodate the expansion of reaction gases.

A modified version of this inclined spiral-screw dissolver, illustrated in Fig. 2, was fabricated. The screw is not enclosed in this unit, and an overhead plenum is provided for gas expansion. The inclined vessel is filled with dissolvent just short of overflowing into the discharge chute at the upper end. This model also proved to have disadvantages. Small pieces of fuel, cladding, and other material lodged between the outer edge of the screw and the wall of the vessel. A wire brush was welded to the outer edge of the screw as a corrective measure, but spacer wires and other small particles became enmeshed in the brush. Some thought was given to enclosing the screw in a perforated cylinder and spiraling the wire brush around the cylinder to provide agitation for the fines that sifted through the perforations, but this was never done. However, the wire brush approach to removal of fines is not compatible with the oscillatory or reverse rotation mode of operation used in previous experiments with UO_2 - ThO_2 fuels.

Further improvements to screw-type dissolvers have been made in recent conceptual designs and models, and these are discussed in the following section. It should also be noted that the continuous dissolver designs just described do not have provisions for separate control of fuel agitation and transfer. The ideal dissolver or leacher should provide adequate residence time to permit complete dissolution of the fuel

Fig. 2. Continuous, Flooded Rotary Leacher.

and proper agitation to prevent local acid deficiencies, and these two features should be separately controlled.

New Leacher Concepts and Models

Eight concepts for continuous leachers were formulated at ORNL as part of the overall program to assess the magnitude of technical problems which arise when the aqueous method is applied to the processing of LMFBR fuels. These concepts include designs for modified versions of the continuous rotary-screw leacher discussed in the preceding section, an inclined vibrating tray, rotating single and multi-compartmented drums, rotating drum with elliptical baffles, rotating drum with compartmented trough transfer, and a compartmented rotating drum with sloped axial transfer ducts. One-fourth scale models were built to demonstrate each of these concepts, and these are operating models that demonstrate the various methods of agitation and transfer of the solids being processed.

Continuous Rotary Screw

The first model, shown in Fig. 3, consists of a spiraled feed screw enclosed in a perforated cylinder. A roller chain is loosely wound around the outside of the perforated cylinder in a spiral and attached with pins at every pitch length. Sufficient sag between pitch lengths is provided to allow the chain to drag on the bottom of the housing. The lower half of the housing enclosing the rotating perforated cylinder is cylindrical, but the upper half of this housing has straight sides to alleviate any jamming that might occur between the chain and the housing.

Fig. 3. Continuous Rotary-Screw Leacher With Perforated Cylinder and Spiraled Chain Flights.

The housing is inclined at an angle of 20° from the horizontal, and the fuel loading tube is at the lower end and the discharge chute is at the upper end of the unit. It is filled with liquid dissolvent to a level that will not permit overflow into the discharge chute, and an overflow at the lower end allows a continuous flow of acid through the unit counter-current to the fuel flow.

Sand and short pieces of wire were placed in the annulus of the model between the perforated cylinder and the housing to simulate the core fines and spacer wires that will sift through the perforations of the rotating cylinder. When the cylinder was rotated, some sand and wires passed through the chain. However, this material eventually moved along the annulus and was discharged into the discharge chute.

Although this model was successfully demonstrated, for actual fuel dissolution, the rotating cylinder should be oscillated to provide the required agitation of the fines and rotated according to a predetermined time cycle to advance the fuel through the leacher.

The function and construction of the second model of the continuous rotary-screw leacher, shown in Fig. 4, are identical to those of the first model except that the roller chain was replaced with a series of loose-fitting hinged flights. Two flight segments were provided for each pitch length, but for simplicity, only two pitch lengths of flights are shown in Fig. 4. Each flight segment is hinged to the perforated cylinder with pins in oversized holes so that the segments will scrape the bottom of the housing as the cylinder rotates. These hinged flight segments can rise upward and pass over any solids that might tend to wedge between the housing and a flight segment. This model provided a more positive transfer of material through the annulus than the first model.

Fig. 4. Continuous Rotary-Screw Leacher With Perforated Cylinder and Spiraled Hinged Flights.

Inclined Vibrating Tray

The concept of an inclined vibrating tray continuous leacher was formulated, and the model of this concept is shown in Fig. 5. This concept basically consists of a vibrating tray that is inclined at an angle of 12° from the horizontal and enclosed in a tank of liquid dissolvent. The tray has a spring-loaded operating shaft which extends through the wall of the tank. Except for a portion at the lower end of the shaft, the tank walls have been omitted in Fig. 5 for clarity.

Sheared fuel is introduced at the lower end of the inclined tray and discharged through a chute at the upper end of the tray. The tank is filled with dissolvent to a level that will not overflow into the discharge chute, and a liquid overflow is provided at the lower end of the tray. Fresh acid is introduced at the upper end of the tray to provide a flow countercurrent to the flow of sheared fuel traveling up the incline.

The model shown in Fig. 5 is provided with a trough in lieu of the discharge chute. Small steps have been attached to the inclined tray to provide the "perch" required to force the fuel up the incline when the compressed spring on the operating shaft is released. The motor is mounted outside the tank, and it has an actuating arm that compresses and releases the spring-loaded shaft. The time required for the fuel to travel up the inclined tray is controlled by the compression of the spring and the length and frequency of the stroke of the operating arm. Any change in the 12° angle of inclination of the tray will also affect the rate of fuel travel. The performance of this model was successfully demonstrated with simulated fuel.

Fig. 5. Inclined Vibrating Tray Continuous Leacher.

Rotary Drum

The model of a concept based on the principle of the concrete mixer is shown in Fig. 6. This design has a single compartment containing four longitudinal baffles inside a rotating cylinder. The baffles are sloped at an angle of 2° to provide axial transfer of the sheared fuel through the unit. A stationary feed tube is located at one end of the leacher, and a stationary discharge tube is located at the other end. The discharge tube extends inside the rotating cylinder to collect the leached hulls as they slide off the edges of the baffles. The dissolvent can be introduced into either end of the leacher. The ends of the longitudinal baffles are perforated to reduce the amount of dissolvent present at the sealed joints and to prevent the discharge of dissolvent with the leached hulls.

Initial testing of this model showed that material flowed through the leacher too rapidly and the 1/8-in.-thick radial baffles were added inside the cylinder to slow the flow. The rate at which material flows through this unit can be changed by changing the slope of the longitudinal baffles, the number of radial baffles, or the rotational speed of the cylinder.

Further testing of this model demonstrated that the bulk of the sheared material was transferred at a reasonable rate but that some of the material passed through rapidly and was discharged prematurely. This phenomenon, known as forward mixing, can result in high losses of fuel and cannot be tolerated in the processing of LMFBR fuels.

Fig. 6. Single-Compartment Rotary Drum Continuous Leacher.

Compartmented Rotary Drum

The model shown in Fig. 7 is similar to that shown in Fig. 6 except that the rotating cylinder is compartmented. This modification represents an attempt to eliminate the forward mixing which occurred in the unit with a single compartment. The model shown in Fig. 7 has six compartments, and each compartment has four longitudinal baffles. Transfer plates between each compartment are provided to transfer fuel from one compartment to the next. The results of demonstration runs with this model showed that forward mixing still occurred but to a lesser degree.

Compartmented Rotary Drum with Elliptical Baffles

A further attempt to eliminate forward mixing resulted in the development of the partial model shown in Fig. 8. The design of this model is the same as that of the model shown in Fig. 7 except for the transfer plates between compartments. In the model shown in Fig. 8, the transfer plates are elliptical baffles, sloped at an angle of 45° , that are located between each compartment. These baffles or transfer plates are mounted on a common shaft which can be rotated or moved axially through center holes in the partitions to control the agitation and transfer of fuel from one compartment to the next. During the agitation cycle, the baffles are positioned to prevent forward transfer of fuel into the next compartment. When fuel is to be transferred, the baffles are rotated and positioned axially to provide the desired rate of transfer.

During tests of this model, the only forward mixing observed during the agitation cycle involved fuel that passed through the openings between

Fig. 7. Compartmented Rotary Drum Continuous Leacher.

Fig. 8. Compartmented Rotary Drum Continuous Leacher With Elliptical Baffles.

the baffles and the holes in the partitions. During the fuel transfer cycle, several revolutions of the cylinder were required to completely empty a compartment. This feature represents a lack of control of the transfer rate between compartments and is undesirable because it will allow fuel to be transferred through the leacher much too rapidly to attain the desired residence time. The use of straight baffles in alternate positions would eliminate this problem, but twice the present total number of compartments would be required.

Compartmented Rotary Drum With Feed Trough

An effort to develop a means of controlling the transfer rate of fuel from one compartment to the next resulted in the model of the compartmented rotary drum with a center feed trough shown in Fig. 9. The design of this model is the same as that of the preceding model except that the elliptical baffles have been replaced with a sectionalized semicircular trough. This trough can be rotated and moved axially through center holes in the compartment partitions to transfer fuel from one compartment to the next.

When the fuel is to be agitated, the feed trough is inverted and the cylinder is rotated for the desired length of time. When fuel is to be transferred from one compartment to the next, the trough is rotated 180° and the cylinder is rotated until all of the fuel in each compartment has been loaded into a section of the trough. The trough is pushed forward until each trough section has been advanced one compartment, and it is then rotated 180° to dump the fuel. To keep fuel from being wedged between mating parts, the cylinder is not

Fig. 9. Compartmented Rotary Drum Continuous Leacher With Center Feed Trough.

rotated while fuel is being transferred from one compartment to the next.

Fuel was successfully agitated and transferred with no back or forward mixing during demonstration runs of this model, but some fuel did lodge on top of the inverted trough during the agitation cycle. However, the main disadvantage of this concept is that the required operating shaft will complicate the design of the fuel entry and leached-hull discharge tubes at each end of the leacher.

Compartmented Rotary Drum with Transfer Ducts

The design problems posed by the preceding concept were overcome by developing a concept in which there are no internal moving parts. This concept consists of a compartmented cylindrical drum which rests horizontally on rollers driven by a reversible motor, as shown in Fig. 10. The cylinder and compartment partitions are identical to those of the two preceding models. There is a hole in the center of each partition, and a sloped conical-shaped duct with a slot in it connects the center hole in the first partition (small end of cone) with the center hole in the second partition (large end of cone). This duct arrangement is used to connect each of the compartments for fuel transfer, and the slot in each duct is sloped from the axis of the drum. Each compartment contains a baffle which is sloped to match the slot in the transfer duct. This is referred to as the transfer baffle, and it is positioned to provide a smooth transfer of fuel into the duct. A second baffle parallel to the axis of the drum is positioned adjacent to the sloped or transfer baffle at the discharge end of the duct in each compartment. This

Fig. 10. Compartmented Rotary Drum Continuous Leacher With Sloped Axial Transfer Ducts Shown in Agitation Mode of Operation.

straight baffle is referred to as an agitation baffle, and it is perforated to permit cross-flow of liquid dissolvent.

Openings are provided in each end of the cylindrical drum for the introduction and discharge of material. Fresh acid can be introduced at either end of the drum, and the product overflows at the opposite end. The compartment partitions are perforated to allow crossflow of the dissolvent. Sheared material may be charged continuously or batch-wise into the first compartment, agitated for a predetermined time, and then transferred into the next compartment. This cycle is repeated until all compartments are charged. Thereafter, during each transfer cycle, leached hulls are discharged from the last compartment and the first compartment is emptied and ready for a new charge.

During the agitation cycle, the cylindrical drum is rotated in the direction which permits the straight or agitation baffles to pick up the sheared fuel and dump it over the transfer ducts to the bottom of the drum. This mode of operation is shown in Fig. 10. When the drum is rotated during the agitation cycle, the non-perforated sloped transfer baffles produce a pumping action of the liquid dissolvent, thereby providing dissolvent flow countercurrent to that of the sheared material. Provision of concurrent or countercurrent flow of the dissolvent is dependent upon whether or not the straight agitation baffles are perforated.

When fuel is to be transferred from one compartment to the next, the rotational direction of the drum is reversed. This allows the sloped transfer baffles to pick up the sheared material and drop it through the slots in the sloped transfer ducts from whence it flows into the next compartment. The transfer mode of operation is shown in Fig. 11.

Fig. 11. Compartmented Rotary Drum Continuous Leacher With Sloped Axial Transfer Ducts Shown in Fuel Transfer Mode of Operation.

The operation of the agitation and transfer cycles was successfully demonstrated in tests of the scale model. There was no forward mixing of sheared material between compartments, and the material was transferred from one compartment to the next in one revolution of the drum. Patent rights for this design have been applied for by the United States Atomic Energy Commission in the United States and six foreign countries.

Conclusions

The new concepts and models of a continuous leacher that were developed provide insight into the problems encountered in controlling the residence time, agitation, and transfer of a wide variety of materials (hulls, wires, and fines) through such devices. These concepts were developed with foreknowledge of the special requirements of LMFBR fuel reprocessing, and their further development should be straightforward. The conclusions reached from observation of the working models are summarized as follows.

1. The continuous rotary-screw models provide the desired control over the flow of material through the leacher, but they do not provide sufficient agitation of the fines.

2. The inclined vibrating tray, rotary drum, compartmented rotary drum, and compartmented rotary drum with elliptical baffles provide sufficient agitation of the fines, but none of these designs permit positive control of the rate at which fuel is transferred through the leacher.

3. The compartmented rotary drum with a sectionalized center feed trough meets all of the performance requirements, but difficulties

could be encountered if sheared material were to become wedged between mating parts. A different method of introducing sheared material and discharging leached hulls will be required for this concept.

4. The compartmented rotary drum with sloped conical transfer ducts meets all of the stated requirements, and it has no moving parts other than the rotating drum. The unit is designed so that the sheared material and the dissolvent can be brought together in one stage and the resultant treated material can be advanced one processing stage at a time, as desired, without forward mixing of the material. Separate positive control over the agitation of sheared material during the required residence time in the vessel and the rate of lateral flow of material through the vessel is provided in this design.

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