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PNNL-38195

Development of a bench-scale dissolution concept for the direct extraction of nuclear fuel

August 2025

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U.S. DEPARTMENT OF ENERGY

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Abstract

Current used nuclear fuel reprocessing efforts utilize a hydrometallurgical approach in which used nuclear fuel is dissolved in hot nitric acid followed by solvent extraction into an organic solvent to harvest target nuclides. Previous studies have shown that the dissolution and loading process could be combined into a single organic dissolution/extraction step, producing loaded organic in a single step process. This single step process also includes the advantage of selectively targeting key nuclides in the dissolution while leaving undesirable constituents as part of the undissolved solids. This process is referred to hereafter as direct extraction. Ongoing research from multiple national labs has proven the effectiveness of this technique at research scale. Therefore, potential methods to implement direct extraction at both bench and industrial scale have been developed. The key features of potential dissolver system designs were identified via the team at Pacific Northwest National Laboratory and several designs based on industrial counterparts were assessed for feasibility. This report summarizes the advantages and disadvantages of multiple methods, concluding with a path forward to create multiple unique dissolver designs. The first design will be a single stage recirculating eductor mixer. The second design recommendation is a stator rotor static mixing flow loop design. Each dissolver could be utilized separately, simultaneously, or in series to answer questions surrounding reaction kinetics including residence time, provide a proof of concept for targeted extractions of specific nuclides, and inform needs for industrial scale implementation

Acknowledgments

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Acronyms and Abbreviations

PUREX	Plutonium Uranium Reduction Extraction
UNF	Used Nuclear Fuel
TBP	Tributyl Phosphate
PNNL	Pacific Northwest National Laboratory
ANL	Argonne National Laboratory
DEHiBA	N,N-di-2-ethylhexyl-isobutyramide
CHON	Carbon, Hydrogen, Oxygen and Nitrogen
CSTR	Continuous Stirred Tank Reactor
PFR	Plug Flow Reactor
HNO_3	Nitric Acid
SRNL	Savannah River National Laboratory
MTU	Metric Tons of Uranium
COTS	Commercial Off the Shelf
TRL	Technology Readiness Level
ORNL	Oak Ridge National Laboratory

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1.0 Introduction

The current standard for nuclear fuel reprocessing is the Plutonium Uranium Reduction Extraction (PUREX) process (Arm 2025; Jubin 2007). This method involves dissolving used nuclear fuel (UNF) in nitric acid (HNO_3) and then contacting it with an immiscible solvent containing tributyl phosphate (TBP) to extract plutonium and uranium. The extracted plutonium and uranium are then selectively stripped into separate streams to produce pure products. An advancement in this process has been the development of monoamide extractants to selectively recover uranium from used nuclear fuel (UNF) (Condamines and Musikas 1992; Miguirditchian et al. 2009; Malmbeck et al. 2019).

Teams at Pacific Northwest National Laboratory (PNNL), Savannah River National Laboratory (SRNL), and Argonne National Laboratory (ANL) have had success adapting the direct extraction concept developed by Hiroshi et al. (1966) for use with N,N-di-2-ethylhexyl-isobutyramide (DEHiBA). Direct extraction with DEHiBA as the solvent provides several noted advantages over traditional HNO_3 dissolution and subsequent solvent extraction, including selective recovery of uranium, a high preliminary degree of separation and the use of a solvent system that complies with the CHON principle (i.e., organic compounds consisting only of carbon, hydrogen, oxygen, and nitrogen). CHON based solvents are promising in solvent extraction efforts due to their effectiveness in minimizing secondary waste generation.

Ultimately, the design requirements of an industrial dissolver for aqueous dissolution in HNO_3 will be different than those of a room temperature dissolver utilizing organic solutions. Based on recent success and lessons learned, teams at PNNL and SRNL are now evaluating existing dissolver designs with the goal of developing a scaled down, industrially inspired system that will facilitate scalability. Section 1.1 describes the key features and requirements that guided the selection of promising dissolver designs. Section 2.0 outlines several dissolver systems and mixing methods that were ultimately removed from consideration, as they did not meet all of the requirements outlined. Lastly, Section 3.0 describes the promising candidates for a bench scale system that meet or exceeds the necessary criteria.

This report fulfills the milestone M4FT-25PN0301010510 *Bench-scale dissolver inspired by industrial equipment*.

1.1 Reactor Types

1.1.1 Batch Reactor

Batch reactors are a relatively common type of chemical reactor. In this system, constituents are added to a tank and allowed to react under constant conditions until completion. Batch reactors may or may not include a mixing apparatus as part of the design. A key feature of the batch reactor is that throughout the course of the reaction, there are no materials added or removed from the tank. These reactors are useful in obtaining the basic information on reaction kinetics including residence times, off gas evolution, and reaction thermodynamics (Perry, Green, and Maloney 1984). Batch reactors are typically used when product prices are high enough to justify lower output rates in exchange for higher yields (e.g., the pharmaceutical industry).

1.1.2 Continuous Stirred Tank Reactor (CSTR)

Continuous stirred tank reactors (CSTRs) are defined by a continuous stream of reactants and products constantly entering and leaving a main tank body. As the name implies, CSTRs include a mechanism for mixing reactants within the main tank. A CSTR can be configured as either a single vessel with constant feed and product streams, or as multiple mixing vessels arranged in series. In the latter configuration, each stage's product stream becomes the feed of the subsequent stage until the desired final product is achieved. While CSTRs typically yield less product than a batch reactor, they offer superior process control. When the reaction kinetics are well-understood, CSTR flowsheets allow for a high degree of control of product stream quality and composition (Perry, Green, and Maloney 1984).

1.1.3 Plug Flow Reactor (PFR)

Plug flow reactors (PFRs) are another commonly used reactor type where the constituents enter at the beginning of a flow stream and exit together as a single product stream. Unlike a CSTR, the reactions in a PFR occur parallel within a single flow direction rather than within a continuously stirred tank. Reaction rates at the inlet of a PFR are very high and gradually decrease as the concentration of solute reacted into the solvent increases. PFRs are theoretically more efficient than an equivalently sized CSTR, driving reactions further towards completion within the same residence time. An ideal PFR assumes that only radial mixing occurs, with reactants in each volume element mixing independently of those upstream or downstream. This configuration greatly simplifies the reaction kinetics since any single region of the stream can be treated independently. However, the design of a PFR must carefully control the flow regime throughout the reactor to maintain ideal flow characteristics. If laminar flow conditions occur, non-uniform velocity profiles (fastest at the center and slowest at the walls) create a wide distribution of residence times across different flow streamlines. This variation means some reactants may exit prematurely while others remain longer than necessary, reducing the overall reaction efficiency (Perry, Green, and Maloney 1984).

1.1.4 Novel Reactor Types

Industrial applications often employ hybrid reactor systems that combine elements of multiple reactor types to address the specific requirements of the desired dissolution. One example would be the use of multiple CSTRs placed in series to approximate the mixing behavior of a PFR. The primary goal of a hybrid type design is to optimize reactor performance and the overall design footprint by tailoring the system to the specific reaction kinetics of the constituents.

1.2 Design Criteria and Desirable Features

The following list describes key features categorized as either required or desirable for the selection of a benchtop dissolver design. Required features are mandatory, while desirable features will guide the final selection among qualified designs.

1.2.1 Scalability

Scalability is a required design feature. The targeted bench-scale design may treat approximately 500 – 1000 g of voloxidized fuel powder with 5 – 10 L of organic solvent to achieve a final concentration of 0.42 M uranium. At full industrial scale, it can be expected that up to 7 metric tons of uranium (MTU), or roughly fourteen spent fuel bundles, would be treated with approximately 70,000 L of organic solvent per day to meet the targeted concentration of 0.42 M.

That is, the targeted full-scale design should be capable of treating approximately 1,000 MTU per year given potential plant downtime. Thus, the bench scale design must demonstrate a clear pathway to industrial scale operation.

1.2.2 Serviceability

A required feature of the design is ease of service. At an industrial scale, the dissolver would likely be installed inside a hot cell or black cell, making it inaccessible for hands-on maintenance once operational. Therefore, a hot cell viable design must be capable of remote serviceability, allowing for component replacement or complete dissolver exchange without direct, hands-on access. However, in a black cell viable design the dissolver would not be serviceable, and lessons learned at the bench scale would inform the final designed lifespan.

1.2.3 Mixing

A critical design requirement is the ability to effectively mix a biphasic system. Since solids in solution tend to settle and/or clump, adequate mixing should be able to maintain particle suspension throughout the entire dissolution process. Voloxidized fuel powder exhibits hydrophilic properties and may be susceptible to the formation of a hydration barrier in the presence of an aqueous phase (Guo et al. 2019). Thus, ample mixing is required to mitigate the potential of a hydration barrier forming around particulates, and therefore, keep solids in contact with organic solvents. Later sections of this report will explore various mixing methods, evaluating which to eliminate and which to incorporate into potential design concepts.

1.2.4 Phase separation; Undissolved Solids and Aqueous Phase

The ability to separate phases is a required feature of the design to support integration with downstream processes, mitigate radiolytic dose to the solvent, and remove the possibility of undissolved solids accumulating at levels approaching criticality safety constraints.

During direct extraction, three phases are present: the undissolved solids, the organic phase, and an aqueous phase produced during dissolution. A drawback to using the organic process solvent for direct extraction is its susceptibility to extreme radiolytic degradation (Kynman et al. 2025). The undissolved solids left in solution are expected to carry a large radiation dose, and therefore must be continually removed. Additionally, the undissolved solids could carry a criticality safety concern if left in place.

Theoretically, the ratio of the organic-to-aqueous phase volume is approximately 800:1. However, experiments have shown greater aqueous volumes of approximately 1% of the total liquid volume (Speelman et al. 2025). The composition of this aqueous phase is not well understood. Due to its small volume and the consumption of acid during dissolution, it is possible that it contains relatively little nitric acid or dissolved material. However, until known otherwise, it must be assumed that this aqueous phase contains isotopes contributing to a significant dose (i.e., Cs-137, Sr-90) and small amounts of U, Np, and Pu that present both criticality and material accountancy concerns when scaled to an industrial process. In the bench scale design, the separation of the aqueous phase must be achieved to answer these questions for industry.

1.2.5 Residence Time

Optimizing residence time in the final design would have several key benefits to the direct extraction process. Shortening the residence time would limit radiolysis of the organic solvent and

therefore create a more favorable environment for the lifespan of the solvent. Similarly, shorter residence times suppress the extraction of undesirable components, benefiting downstream operations, such as separating uranium from the loaded solvent. Lastly, for a given throughput, an optimized residence time largely coincides with a reduced footprint. By reducing the overall size of the equipment, the total plant footprint can also be reduced, or multiple process streams could be installed within a smaller total area.

While residence time is an important factor in the final design, substantial optimization can be expected to occur during practical implementation rather than the design phase. Theoretically, the direct extraction parameters are mostly known from previous efforts, but the lessons learned during the implementation of a bench-scale design will provide further opportunities for optimization to industrial scale (Speelman et al. 2025; Hall et al. 2025; Hall et al. 2023; Hall et al. 2022).

1.2.6 Continuous Flow versus Countercurrent Flow

Continuous flow-type reactors offer several advantages over batch systems. These systems integrate more effectively with downstream processes (e.g., solid separations), thereby increasing the total daily throughput. Among continuous flow designs, countercurrent flow reactors provide the added advantage of reducing solvent exposure to radiation dose. However, there is a notable trade-off: countercurrent type reactors for a uranium-only recovery may promote the extraction of unwanted constituents. That is, the countercurrent flow inherently exposes fuel powder to fresh solvent, a scenario that would typically maximize dissolution efficiency. Yet for uranium extraction using DEHiBA, undesirable components, such as Zr, Np, and Pu may more readily dissolve into the fresh solvent. Another concern with countercurrent flow would be the increased potential to expose fresh solids with an aqueous phase and form a hydration barrier around fresh solids. As noted in Section 1.2.4 the ratio of an aqueous phase in solution increases as the direct extraction progresses. Therefore, in a countercurrent flow design the probability of solids contacting the aqueous phase would be the greatest at the point where fresh solids are added. The design selection process must balance these competing factors. While countercurrent flow beneficially reduces radiation exposure to the solvent, it simultaneously increases the risk of extracting unwanted constituents that compromise downstream processing and the probability of fresh solids forming a hydration barrier by contacting an aqueous phase.

1.2.7 Commercially Available

Generally, commercial off the shelf (COTS) equipment is preferred for the selected bench scale dissolver designs. COTS equipment is at a higher technology readiness level (TRL) and therefore the effectiveness can be demonstrated in the environment where it will be utilized. Additionally, COTS equipment is more readily attainable than custom-built components for a bench scale design. On an industrial scale, however, it is likely that much of the equipment will require custom design and procurement. Therefore, this requirement would not impact efforts to scale up.

1.2.8 Criticality Safe Geometry

A full analysis of criticality safety for the industrial setting is outside the scope of this report. Efforts will be made to emphasize criticality safe geometries. Hydrocarbons present in organic solvents are a more effective moderator than water or nitric acid (Ayres and Trilling 1971). Thus, the criticality risks are greater in direct extraction versus nitric acid dissolution. In an industrial process, the addition of a neutron poison which is soluble in the chosen process solvent may be necessary which would later be separated before stripping the product. However, criticality

considerations are less of a concern at bench scale design and thus would not require a neutron posoin. At full-scale, a complete criticality safety assessment would be required.

1.2.9 Process Monitoring

Previous efforts of direct extraction utilized both the visible and Raman spectra to monitor the progress in real time through a flow through loop (Bessen et al. 2024). Similar process monitoring will need to be considered at the bench scale with the ability to be scalable to industry. Alternatively, to accommodate bench scale installation, grab samples may be taken at predetermined intervals from the bench scale design. Therefore, this is not a required feature of the design but will help to distinguish between various dissolver designs when making a final selection.

1.2.10 Off-Gas Collection and Analysis

Traditional nitric acid dissolution inherently produces NOx as a reaction byproduct. For environmental reasons, NOx off-gas must be treated before it is allowed to vent to the atmosphere. When the fuel is converted to UO_3 via voloxidation in preparation for direct extraction; much of the burden for treating off-gas is shifted away from the dissolver to the voloxidation unit operation. Nevertheless, NOx may be produced in the dissolver due to radiolytic generation. At the bench-scale, off-gas analysis will either occur in real-time or through the collection of the off-gas in sample bulbs. Industrial-scale operations will require real-time treatment of off-gas; therefore, bench-scale analysis will inform these future requirements.

1.2.11 Temperature Control

On an industrial scale, it is estimated that heat generation may need to be managed through heat exchangers or similar heat rejection systems, owing to the exothermic nature of the reactions. At the bench-scale, the addition of heat may be utilized to mimic full scale behavior. Generally, temperature control is not a significant factor in the process and not discussed further in this report. Temperature monitoring during selected processes may be used to better estimate the temperature generation that would occur through scale up to industry.

1.3 Graded Approach

A graded approach will be utilized to determine the most feasible design for a bench scale dissolver. The design criteria and desirable features described in Section 1.2 will be applied toward promising dissolver designs, which will be graded as satisfactory, passable, or unsatisfactory following their description. A satisfactory grade indicates that the given criteria or features require little to no compromises either at the bench scale or upon scale up to industry. Design criteria graded as satisfactory indicate that the specific dissolver design is ideal given that the required traits of the design can be readily implemented. Next, a passable grade indicates that the specific design criteria or feature requires compromise for implementation of the design, but is a solvable problem given careful considerations for design challenges. If a required design trait is given a passable grade, then the dissolver design is achievable. However, it may be less ideal than a comparable design with better overall grading. Lastly, an unsatisfactory grade means that the design criteria or feature requires a major compromise upon implementation of the design in practice. Unsatisfactory grades for required design traits indicate that other dissolver designs should take priority when considering a recommendation for fabricating a bench scale dissolver.

2.0 Down Selection of Dissolver Designs and Mixing Methods

2.1 Simple Batch Reactors

A batch reactor, defined in this case as a closed vessel with constituents added, has been removed from consideration for the direct extraction dissolver design. Batch reactors are considered when long residence times are allowed to achieve a high conversion where cost restrictions for size and space are not an issue. Aqueous dissolution of spent fuel with nitric acid (HNO_3) has occurred in batch reactors for this reason because nitric acid maintains the ability to be an effective solvent in the presence of a diverse and intense radiation field and the need for full dissolution of the fuel. Additionally, adding heat to greatly increase reaction kinetics is allowable with HNO_3 so residence times can be reduced to increase production for a given reactor size. While DEHiBA has shown an increased resilience to radiation relative to TBP with an increased radioprotective effect after uranium dissolution, prolonged exposure to high dose increases the likelihood of solvent breakdown and NO_x generation from the aqueous phase limiting reaction time and the benefit for using a batch reactor. (Kynman et al. 2025) Additionally, heat input with DEHiBA is limited due to increased combustibility concerns relative to HNO_3. A continuous reactor such as a CSTR or PF may minimize dose exposure protecting the integrity of the organic phase. To ensure integrity of the organic phase, the simple batch reactor design was removed from consideration.

2.2 Mechanical Agitation

Certain types of mechanical agitation have been deemed to be undesirable. Overhead mechanical agitation such as large impellers, paddles, and shear blades pose an issue with in-service maintenance, replacement and disposal. At a bench scale, removal and replacement is easy, at an industrial scale the weight, dose and contamination risk would likely make disposal of the entire assembly more appealing than replacing potentially failed components. The cost of replacing the entire assembly as a routine maintenance item is less desirable than replacing components of the assembly. Also, routine maintenance associated with the use of mechanical agitation would eliminate the use of a black cell in an industrial scale design. The limiting nature of mechanical agitation at industrial scales led to eliminating this type of mixing from a final design.

2.3 Air Sparging

When combined with large additions of air the DEHiBA solution presents a foaming hazard. While surfactants could be used to decrease the foaming issue, introducing additional constituents would likely increase the complexity of the chemical reactions during dissolution and the subsequent chemical separations steps. Typically, air sparging is added to the bottom of a batch type reactor or CSTR to keep solids suspended from the bottom of the tank. However, as the density of the solids increases the flow rate of air sparging necessary to properly suspend solids in solution also increases. The flow rate of air necessary to properly mix voloxidized fuel in solution would become a limiting factor as foaming issues would increase as the air flow rate increases. Thus, simple mixing by way of air sparging to suspend solids was removed from consideration.

An air lift pump dissolver has been previously established and studied for the dissolution of PuO_2 in a nitric acid media (Tesitor 1978). The air lift theory relies on the pressure differential due to density differences of the gas-liquid mixture versus the surrounding liquid. This dissolver design

requires no moving parts and theoretically the serviceable portions of the design could be located outside of any shielded containment. However, in the case of PuO_2 powder, the abrasive nature of suspended particulates required the dissolver body to be replaced after a year of service life. Additionally, to effectively suspend particulates, a flow rate of between 4 to 12 liters per minute of air sparging was necessary within 4 liters of liquid volume. This resulted in a turnover rate of 30 liters per minute within each unique dissolver vessel. It can be assumed that similar air sparge flow rates would be required to suspend voloxidized fuel powder and upon scale up to industry the vessel size, liquid volume, and thus air sparge rate would all increase accordingly. Similarly to simple air sparging, the air lift pump would also introduce a foaming hazard when combined with organic solution as the primary solvent. Again, surfactants could be used to counter the foaming issues but would introduce new complexities to the chemistry within the dissolver. Surfactants could also have the potential to interact with the voloxidized fuel as a flocculant and lead to aggregation of the particles. For those reasons, the air lift pump design was removed from consideration.

2.4 Pulse Jet Mixing

Pulse jet mixing is used in the nuclear industry to mix tanks of sludge in black cells given their low maintenance, long service life, and ease of installation. With no moving parts within the reactor and all mechanical systems (e.g. vacuum pump) outside radiological areas, long term serviceability is the key feature of this type of mixing. However, the cost of implementation is much higher on smaller scale systems compared to other mixers and do not necessarily scale easily from bench scale. While pulse jet mixing is useful for large scale systems, the geometry of the reaction tank matters to ensure solid suspension is achieved. Criticality concerns have plagued this type of mixing where developed solid heals have led to use of only batch reactors where the contents can be emptied out. As discussed *a priori*, batch type reactors are an undesirable design given concerns with dose of the solids breaking down the DEHiBA organic solvent so this may not be the best mixing method despite the serviceability advantages. While there are advantages of pulse jet mixing for large batch reactor designs, these advantages are nullified when scaled to a continuous bench scale dissolver design. Therefore, it was removed from consideration as a primary mixing method for the bench scale system.

2.5 Down Selection Summary

Given parameters identified in Section 1.1 and Section 1.2 the initial down selection of dissolver designs and mixing methods was presented. Batch type reactors were ruled out during down selection as the concerns around exposing the organic phase to a large radiation dose over an extended timespan were unable to be remedied given design constraints of batch reactors in addition to the large footprint for a given throughput. Mechanical agitation, air sparging and pulse jet mixing methods were also eliminated from consideration as typically these methods are added to an existing batch reactor to suspend solids into the solution. However, other reasons for eliminating these mixing methods varied from serviceability issues to complexities surrounding foaming concerns via air sparging. An air lift pump design for dissolution of PuO_2 within HNO_3 was discussed and ultimately removed from consideration primarily due to foaming concerns when adapted to DEHiBA direct extraction. Further discussion will present promising designs for a bench-scale application when considering the required design criteria.

3.0 Promising Dissolver Designs

3.1 Rotating designs

3.1.1 Rotating Screw

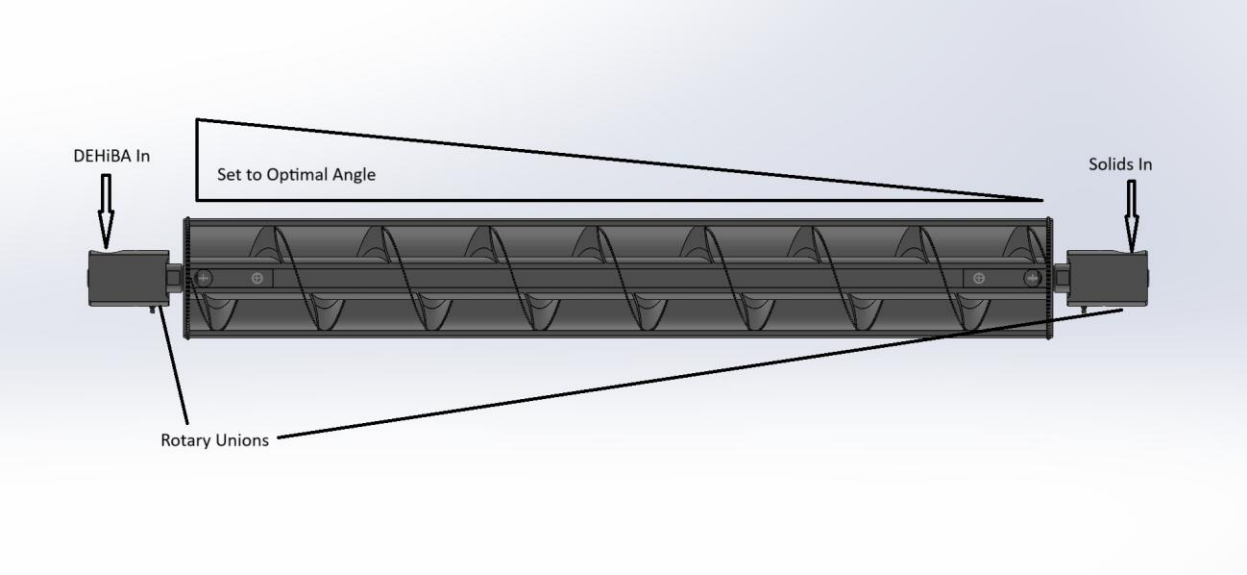

Figure 3-1. Custom designed rotating screw dissolver.

The rotating screw dissolver features an auger that would be installed in segments and welded to the interior of the drum. This would force liquid and solids to travel the full length of the auger. Rotary unions at either end allow for additions at either end, allowing for countercurrent flow of organic to solids. The length of this design could be modified to an optimal length following bench top testing once residence times are known. Moving parts would be outside of the assembly and this design would be easily scalable with implementing multiple in parallel. While the current design does not have outlets added, benchtop testing could implement multiple outlets across the length to identify optimal location for undissolved solids and loaded organic discharge.

A drawback to this design would be the inability to remove solids from the interior of the assembly. Fuel would eventually coat the interior of the assembly creating a potential criticality issue, and decreasing the efficiency of the assembly. Benchtop testing would help to get an idea of the rate of accumulation while exploring potential removal solutions. Additionally, the rotary unions will be the most likely point of mechanical failure which would in turn produce a leak. Secondary containment and redirection equipment would need to be a consideration to safely mitigate this concern.

3.1.2 Counter Current Rotary Dissolver

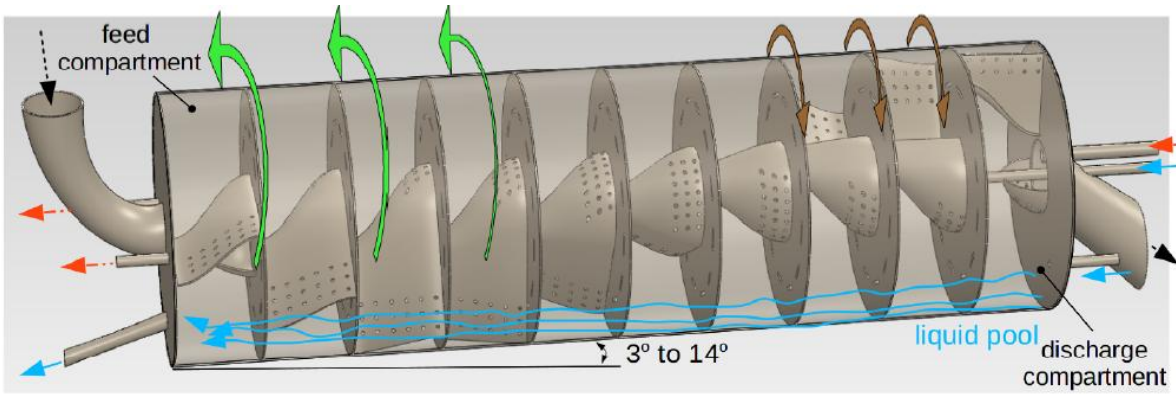

Figure 3-2. Countercurrent rotary dissolver design. Green arrows indicate leaching-mixing rotation direction while brown arrows indicate solids transfer rotation direction. CAD model image provided via ORNL report (de Almeida 2012).

An evolution of the rotating screw dissolver is the counter current rotary dissolver. This can simplistically be thought of as a stage-wise counter current dissolver while the rotating screw is a continuous dissolver. Previous efforts at Oak Ridge National Laboratory (ORNL) have proven the effectiveness of a countercurrent rotating drum for dissolution of large scale “chunks” of UNF along with the potential to dissolve voloxidized powder. (Odom 1972; de Almeida 2012; Lewis 1984) The design is based on a U.S. Patent filed in 1973 by Clyde Odom and Maurice Kunselman (Odom and Kunselman 1973). However, bench scale efforts could modify the design by decreasing the number of partitioned sections as well as decreasing the overall size of the partitions. The full-scale design described by ORNL has been proven with modeling efforts to accommodate up to 1 ton per day of dissolution. This could scale into industry via increasing the number of partitions, increasing the rotational speed of the drum, or placing several drums in parallel with each at the proven 1 ton per day benchmark. The rotation of the drum is driven by external rollers and there are no moving parts within the walls of the drum. Similarly, the speed of the rotation is optimized to eliminate concerns with erosion due to collision with the used fuel particulates. Therefore, remote handling and serviceability is all external to the drums radiation hazards. At the end of its usable life, the drum could be picked up and a new one placed onto the rollers. The contents of the drum are continuously mixed via the internal partitions. Likewise, the orientation and spacing of the partitions allows for undissolved solids to be picked up from one partition and dumped into the next partition when the rotation direction is reversed. Countercurrent flow of solvent means that the solid feed moves from contacting spent solvent to continually contact areas of fresh solvent. Similarly, this means that dose to fresh DEHiBA solvent is minimized with a short residence time. Commercially available rollers could be procured to ease the burden of designing an apparatus; however, the drum itself would be a custom design and build. Lastly, off gas collection and process monitoring would be achievable via simple additions at the headspace of the tank and at the liquid outlet flow.

A potential drawback to this design, and countercurrent flow, could be that the addition of fresh solid particulates occurs in the area of the tank with the highest concentration of aqueous phase. Voloxidized fuel powder exhibits hydrophilic properties and therefore an aqueous barrier could coat the powder and hamper the ability of future partitions, with higher solvent concentration, to contact the fuel powder (Guo et al. 2019). Eventually a collection of undissolved solids may coat the walls of the drums and surfaces of the partitions, adding to the concerns of serviceability. A simple washdown of the tank with nitric acid may solve this issue but could increase the secondary

waste with a concern for nuclear nonproliferation. This concern could be mitigated by taking advantage of leftover solutions from downstream processes, for example the scrubbing solution from solvent exchange, but this would require complex planning and coordination. Lastly, accurate particle size distributions would be paramount to the success of this design given the need for properly sized liquid flow through holes in partition walls and mixing flanges.

Overall, the countercurrent rotary dissolver design meets many of the design criteria while mixing custom built parts with commercial off the shelf equipment. While not without flaws, the bench scale design could prove to alleviate assumed negatives via optimizing residence times, recirculation of fresh DEHiBA within later partitions, or other process parameter optimizations.

3.2 Semi-Custom Eductor Mixing

3.2.1 Multistage Cascading Eductor

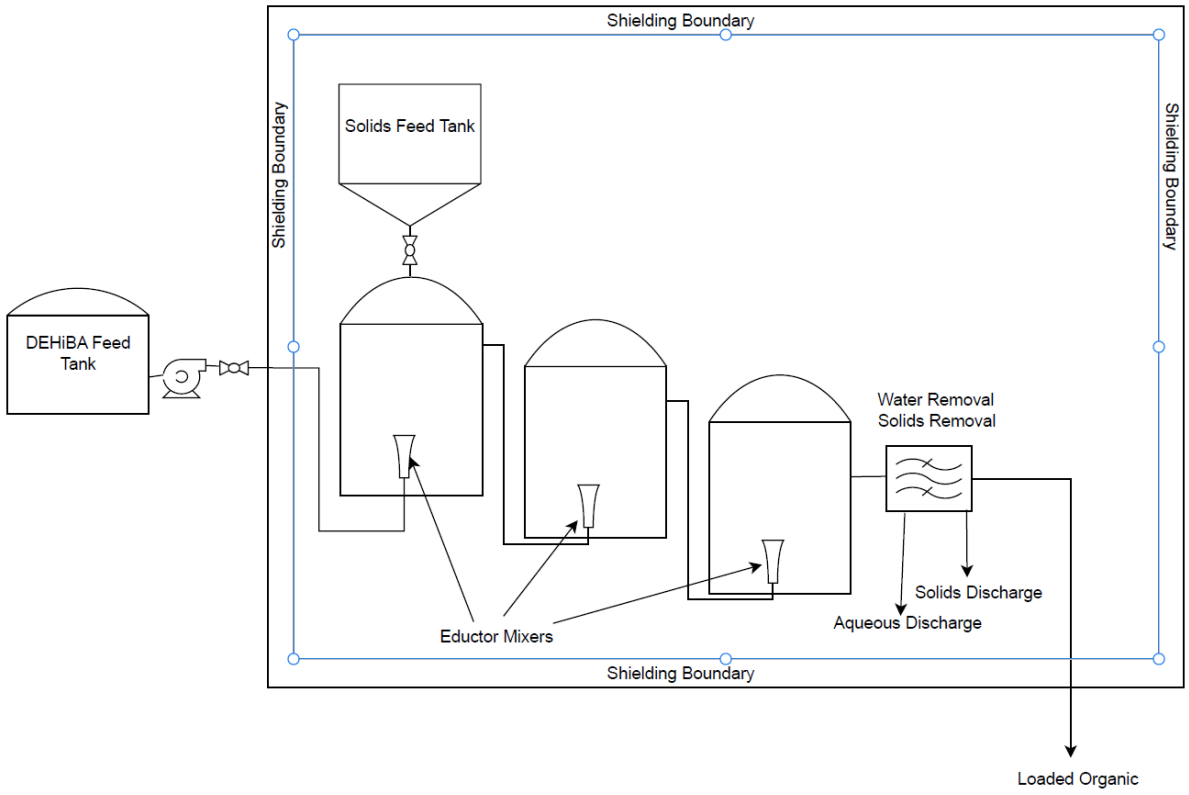

Figure 3-3. Conceptual multistage cascading mixing eductor.

Mixing eductors work within a very similar theory to the previously mentioned air lift design, however the low-pressure region is created via the velocity of fluid pumped through the nozzle. This causes the immediately surrounding liquid and solid particulates to be sucked into the nozzle and mixed into the incoming liquid stream. Therefore, there are no moving parts within the tank, and air is not added to the tank. Serviceability is thus limited to the pump outside the tank and efforts to replace or service the pump could be handled via remote methods. Scalability could be

solved by optimizing the tank and eductor geometry to an increased size and thus a greater amount of processed material. A simple mixing eductor flow loop could be custom designed and fabricated using only commercial off the shelf parts. Lastly, given the simple tank geometry and inherent flow loop present in the design both off gas removal and process monitoring could be added to the bench scale design without any interruption to the tank mixing.

The design outlined in Figure 3-3 would utilize a series of eductors installed into tanks to create a mixed output product. This design follows the principles of a CSTR set up in series where the product from one tank becomes the reactant for the next tank and so on. The first tank would receive the solids feed and be mixed while filling with DEHiBA until it reached the point of overflowing to the next tank. Operation of the eductors in series would be handled by the flowing liquid stream between tanks. Following the series of tanks, the solution would move to a solids and aqueous phase separation. Following that, loaded organic would be discharged for processing, solids and aqueous phases would be discharged separately.

Over time the eductors would eventually erode and need to be replaced, bench scale testing and modeling would be able to demonstrate the frequency of required in tank maintenance at full scale. Additionally, mixing studies would need to be done to ensure the single pump in combination with gravity feed to subsequent tanks would be enough to meet the optimal pressure for the eductors to properly mix.

3.2.2 Single Stage Recirculating Eductor

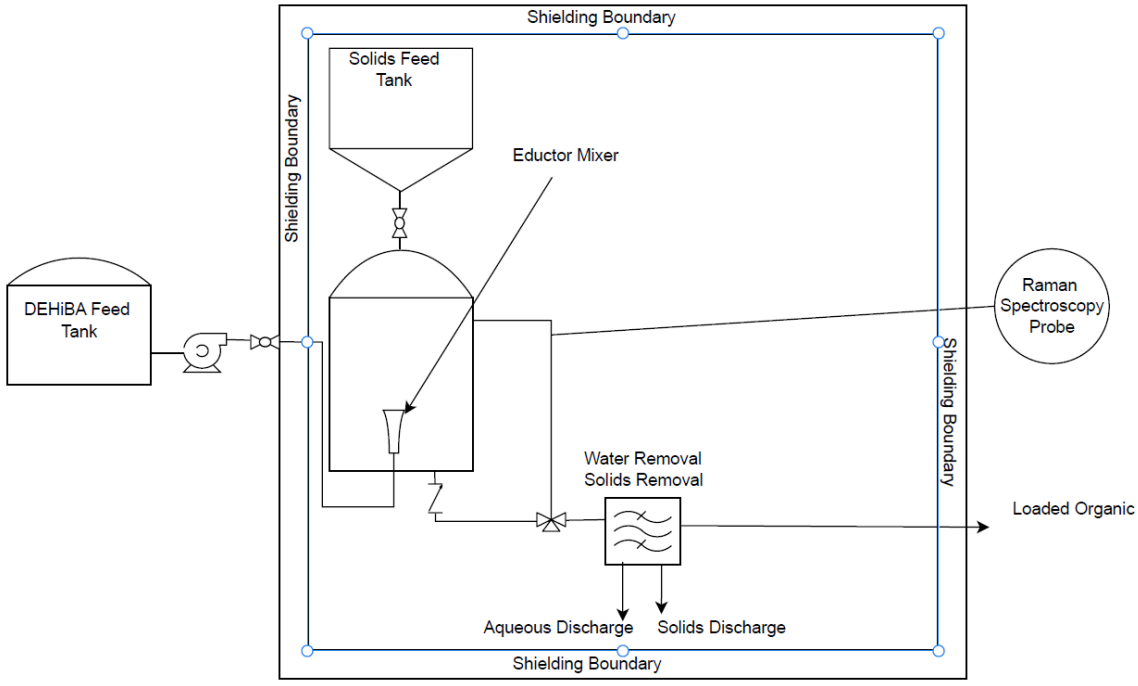

Figure 3-4. Simple single stage eductor mixer apparatus.

While the previous cascading eductor design (Figure 3-3) could theoretically be achievable at full industrial scale, the bench scale design for installation in a hot cell may be limiting due to space constraints depending upon the size of the hot cell. A simple single stage mixing eductor such as the design shown in Figure 3-4 would be better suited to a limited space design. This design holds many of the same advantages as the cascading eductor mixer shown in Figure 3-3 and would provide a potential proof of concept for the full-scale cascading design performed at the bench scale. This design meets the classification of a single stage CSTR.

3.3 Stator Rotor Static Mixing Flow Loop

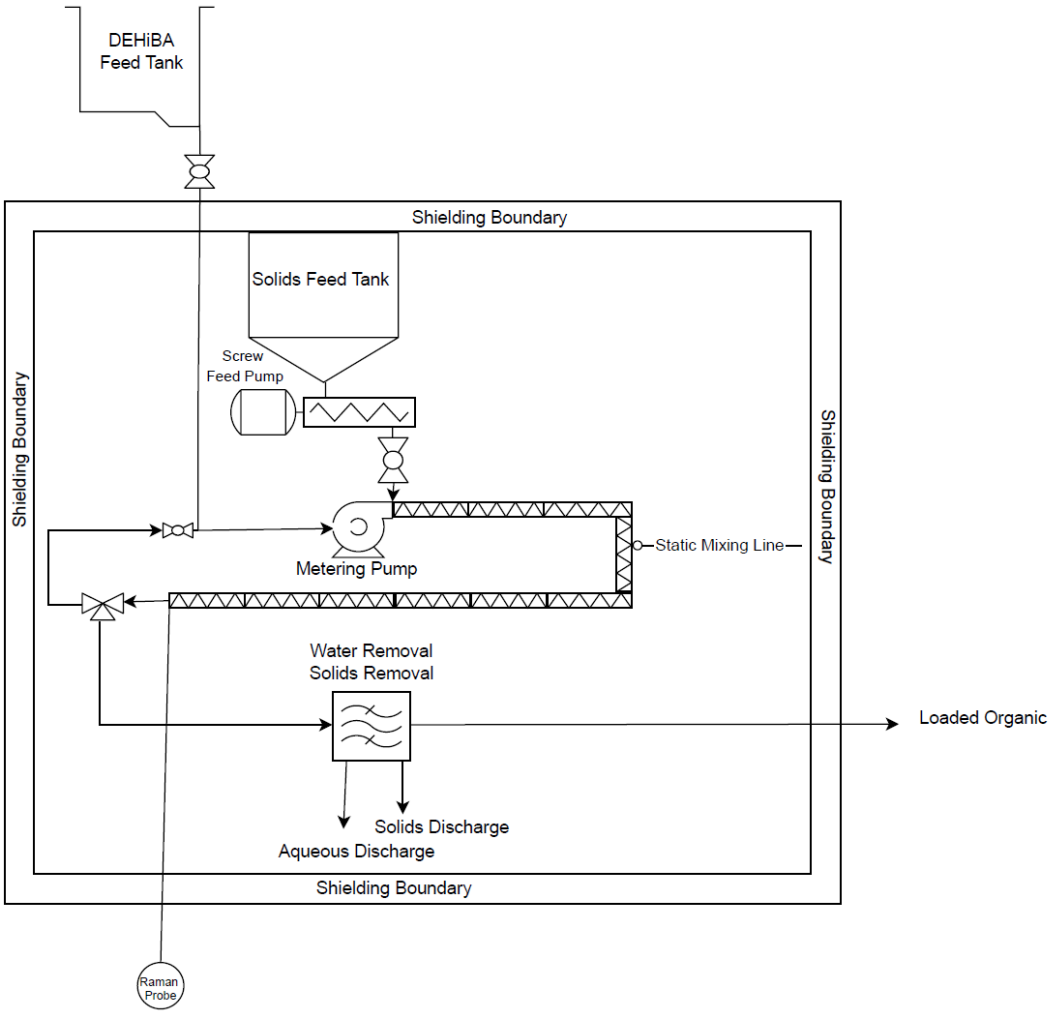

Figure 3-5. Stator rotor static mixer flow loop.

Stator rotor mixers are readily used in a variety of industries where a metered flow of solid to liquid reactants is necessary. Having the ability to meter both the solids and liquid feed would allow for target ratios to be achieved at the input location. Feeding the mixed slurry directly into a static line mixer provides constant mixing along the length of piping. (Shirzadi et al. 2024; Thakur et al. 2003) Given the design elements, this dissolver primarily resembles a PFR due to the single piping flow loop. However, the introduction of static mixing within the loop creates additional

mixing in the flow direction and mimics the characteristic of a CSTR within this section of the design. Given the flow occurs in a single primary direction concern of an aqueous barrier forming around solids are mitigated with this design. Upon addition, fresh solids contact fresh solvent with the lowest ratio of aqueous phase. Also, the static mixing portions of the flow loop continuously mix the solids with the solvent stream throughout the duration of extraction breaking up any formation of an aqueous barrier. For bench scale testing, Raman spectroscopy can be utilized at multiple points in the piping to get a better understanding of the conversion rate and required residence time. During bench scale testing, if the conversion percentage is not at the target by the end of the stretch of piping, a three-way valve would allow for recirculation of the slurry back through the static mixing line, allowing for operation as a flow through or semi-batch reactor. At a larger scale, once kinetics and flow rate have been evaluated further, the piping could be elongated to achieve the target residence time and perform as a flow through reactor. Once the desired conversion percentage is achieved, the liquid would be sent directly to solids and aqueous separation. If it is determined during bench scale testing that adding heat is desirable, implementing a feed tank heater or a heat exchanger along the static line mixer would be simple modifications. This design, as a continuous flow loop, would create an ideal scenario for conceptual testing at the bench scale in a fumehood, glove box, or hot cell environment due to the optimized size relative to the required components. Lessons learned at the bench scale would inform scaling efforts for industry to modify the design to a single pass flow through where the static mixing line length is optimized to match the desired flow rate and residence time.

The slurry containing loaded organic, undissolved solids and some amount of aqueous phase could be filtered using a centrifugal separator to create an output of loaded organic. It would be preferred to have the separation controlled by one step, however, inline back-pulse filters or cyclonic desanders could be used to optimize solids removal prior to removing the aqueous phase.

With mechanical parts this design would require servicing. Erosion of the pump and static mixing lines would need repaired or replaced. Bench scale testing and modeling would help to determine the frequency of the necessary maintenance. Additionally, the hopper feed of voloxidized fuel would present challenges. It would be recommended to store hopper in a dry, shielded environment and utilize hopper agitation to prevent bridging and other feed challenges. Additionally, both motors could be converted to air powered motors, running on plant air rather than electricity to decrease maintenance requirements and increase service life. Lastly, with similar mixers deployed across many industries, components are readily available commercially for both bench and industrial scale applications.

3.4 Phase Separation

Upon implementation of a bench scale design, it is recommended that varying methods of phase separation be tested to determine the most suitable candidate. Potentially multiple separation techniques could be utilized in series to create an optimal path to capture of the desired components. At bench scale the phase separation techniques should occur outside of the main dissolution process at smaller scales. Thus, the overall goal of bench scale phase separations would be to tie in the most effective technique to a finalized dissolution design.

Centrifugal separation could allow for both aqueous and solids removal at the same time, discharging loaded organic in one step while removing unwanted solids. However, centrifugal separators can introduce additional maintenance to the final design. Even with maintenance concerns, centrifugal separation techniques have historical background for use in a black cell type

reprocessing facility (Baker et al. 2022). This separation technique would be ideal for a flow through type dissolver design with the separation occurring at the outlet of the dissolver.

Inline back-pulse filters or cyclonic desanders could be used to optimize solids removal prior to removing the aqueous phase. This would allow for a sedimentation basin to function as a large-scale separatory funnel for aqueous phase removal, with the solids gone, dose poses less radiolysis concerns to the DEHiBA. The low maintenance aspect of this separation technique could provide a strong advantage to use for a final design, however a large total plant footprint would be required to accommodate these methods of phase separations.

3.5 Discussion

Several promising designs ranging from rotating drums to multistage cascading type flow loop designs were presented in the previous discussion. Table 3-1 summarizes key aspects of each design including the graded approach for design criteria outlined in Section 1.1 and 1.2 , viability for hot cell installation, and eventual scale up for industrial applications.

An eductor based mixing system described in Section 3.2 as either a multistage cascading design or a single recirculating type eductor was presented with several key advantages for a direct extraction flowsheet. Primarily, each design meets the required design criteria for scalability, serviceability, and mixing characteristics. These eductor-based systems may also become adaptable to a single stage uranium only extraction process or a dual stage type extraction where radionuclides of interest are selectively extracted within the unique stages of the process. Specifically, a multistage eductor process as described in Section 3.2.1 could provide the framework for an industrial scale design where each tank in the cascading series targets specific elements of interest. This setup would require careful planning and the ability to separate product streams between stages for targeted nuclide extractions. Alternatively, the single stage eductor as described in Section 3.2.2 could provide targeted extractions by simply routing solids removed from the first direct extraction back through the same dissolver with a fresh DEHiBA feedstock. This would provide a method to test a multistage targeted extraction flowsheet at the bench scale by limiting the overall footprint necessary for a bench scale design. Eductor-based designs become appealing if longer residence times are proven necessary to complete each stage of dissolution. Implementation of bench scale testing would prove invaluable for determining the residence time both at the bench scale and for scale up to industry.

The stator rotor static mixing flow loop design described in Section 3.3 provides another promising direct extraction framework. The required design criteria of scalability, serviceability, and mixing are all graded as satisfactory for this design. Additionally, careful planning of a bench scale iteration could provide a multistage extraction flowsheet. Several mixing loops could be placed into series with one another where they are separated by a targeted phase separation between the loops. If desired, the solids removed from the first mixing loop would directly feed into the second loop for a series of targeted extraction steps where each loop provides a loaded organic product containing the elements of interest as separate product streams. At the bench scale this same idea could be achieved by simply routing the removed solids back into the original dissolution loop with fresh DEHiBA. Therefore, decreasing the necessary footprint and overall complexity of a bench scale design while providing necessary insights for industrial scalability. A static mixing flow loop would become appealing to optimize residence times by altering either the solution flow rate or the static mixing line length. Alternately, the slurry could perform several passes through the static mixing loop before proceeding through solids removal. Therefore, the bench scale static mixing system could produce several tests of dissolution residence time without a major increase to the design size.

Table 3-1. Promising dissolver designs summary table

Designs	Design Criteria and Features			Hot Cell Viability	Industry Scale Up
	Satisfactory	Passable	Unsatisfactory		
Rotating Screw (3.1.1)	- Scalability - Mixing	- Serviceability - Residence Time - Critically Safe Geometry - Off Gas Collection - Temperature Control	- Flow - COTS - Process Monitoring	Viable hot cell design with proper sizing of mixer tank	Scale up would depend on desired throughput.
Counter Current Rotary Dissolver (3.1.2)	- Scalability - Serviceability - Mixing	- Residence Time - Critically Safe Geometry - Off Gas Collection - Temperature Control	- Flow - COTS - Process Monitoring	Viable hot cell design with proper sizing of mixer tank	Original designs were modeled for industry. Scale up would depend on desired throughput.
Multistage Cascading Eductors (3.2.1)	- Scalability - Serviceability - Mixing - Flow - COTS - Off Gas Collection - Temperature Control	- Residence Time - Critically Safe Geometry - Process Monitoring	N/A	Hot Cells may present space restrictions depending on size.	Scalable to industrial size
Single Stage Recirculating Eductor (3.2.2)	- Scalability - Serviceability - Mixing - Flow - Off Gas Collection - Temperature Control	- Residence Time - COTS - Critically Safe Geometry - Process Monitoring	N/A	Viable hot cell design with proper sizing of mixer tank	Scale up to a multistage design
Stator Rotor Static Mixer Flow Loop (3.3)	- Scalability - Mixing - Residence Time - Flow - COTS - Critically Safe Geometry - Process Monitoring	- Serviceability - Off Gas Collection - Temperature Control	N/A	Viable hot cell design with proper sizing of static mixer loop	Scale up would depend on desired throughput and residence time.

Satisfactory – Requires little to no compromises to place into service
Passable – Requires a moderate level of compromise to place into service
Unsatisfactory – Requires a high level of compromise to place into service

4.0 Conclusions

Direct extraction of uranium via N,N-di-2-ethylhexyl-isobutyramide (DEHiBA) has shown promising results via collaborative efforts at PNNL, SRNL, ANL, INL, and ORNL. Bench scale efforts in direct extraction via DEHiBA have been proposed in the 100 – 1000 g scale to inform efforts in potential industrial applications. Key features of a bench scale design are outlined in Section 1.1 and include items such as scalability, serviceability, phase separations, optimized residence time, and commercially available parts. Given the desired design criteria mixing and dissolution methods involving batch reactors were quickly down selected from a potential design given the need for protecting the organic phase for radiolytic degradation. Pulse jet mixing was eliminated from final considerations given concerns with properly suspending solids into solution, scalability within a small-scale design, and high costs associated with bench scale implementation. Simple mechanical agitation via stir paddles or similar methods were also eliminated given concerns over serviceability of parts at an industrial scale. Lastly, air sparging was eliminated from contention given concerns over foaming of organic solvents.

Promising designs for the bench scale recommendation included rotating type screw designs utilizing counter current flow where fresh the process solvent enters at the opposite side as the fuel powder. Mixing eductor designs optimized for industrial scale or a bench scale proof of concept were also discussed. Lastly, a commercial or custom flow loop type design utilizes a COTS stator rotor mixer paired with static mixer flow lines show considerable promise. Both the stator rotor mixer and the eductor mixer design could are a continuous (concurrent) flow type reactor. Having a concurrent flow of the process solvent and fuel powder is viewed as advantageous from both preventing the extraction of undesirable components as well as minimizing blinding of the fuel by water produced during extraction of the uranium.

5.0 Reccomendations

It is recommended to proceed with fabricating and testing bench scale designs for both an eductor mixing based system and the stator rotor static mixing flow loop. Given the need for hot cell viability, the recommendation is to proceed with a single stage eductor mixing setup rather than multi-stage. Upon final bench scale design, the static mixing flow loop could be custom sized to fit within the appropriate location and therefore could be placed into service following the conceptual design outlined in Section 3.3. As shown in Table 3-1 each design includes satisfactory or passable grades for all defined design criteria with no features graded as unsatisfactory. A benefit to producing both designs could be the potential to examine each design, either separately or in series, for targeted extractions. Given the unknown nature of various reaction kinetics, including residence time, future work will better define these characteristics under unique dissolver designs and flowsheets to determine the methods most appropriate for industrial scale. In particular, an emphasis should be placed on benchmarking the minimum time to achieve dissolution in these two designs. Each design could be created using COTS parts which is a highly desirable trait for cost-effective production of a bench scale design.

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