

## **TREATMENT OF RADIOACTIVE WASTES USING LIQUID-LIQUID EXTRACTION TECHNOLOGIES-FEARS, FACTS, AND ISSUES**

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### **ABSTRACT**

Liquid-Liquid Extraction (LLE) technologies have been extensively used in the nuclear industry worldwide since the 1940's for the recovery and purification of uranium and plutonium. These and related LLE technologies continue to be successfully utilized outside of the U.S., but their use for nuclear applications in the U.S. has been essentially discontinued. Numerous LLE technologies have been developed and successfully demonstrated in the past 15 years for the treatment of radioactive wastes; yet DOE, contractor, and stakeholder acceptance for these technologies has been limited. A lack of understanding of the technology by decision makers and the public as well as perceived or exaggerated environmental issues have led to the limited acceptance. Liquid-liquid extraction technology offers many advantages over other chemical separation techniques that should be considered in the technology selection process.

A review of LLE principles and discussion of issues and expressed concerns, as well as technology advantages and disadvantages will be provided. The potential for future selection of LLE technology and implementation in U.S. DOE waste treatment plans will be assessed.

### **INTRODUCTION**

At the start of the third millennium, the nuclear industry finds itself with much to offer humankind but with seemingly intractable problems to solve. On the one hand, the looming specter of global warming and limited fossil-fuel resources presents great opportunity. On the other hand, increasing public skepticism of the nuclear industry's environmental and safety record, due in part to revelations of environmental neglect and accumulated waste following the end of the Cold War, threatens the existence of the nuclear industry. Meeting the challenge of environmentally responsible management of waste would certainly remove a key roadblock to further use of nuclear energy.

The large volume of Cold War legacy wastes poses one of the most urgent needs for the nuclear industry and governments. The presence of long-lived radionuclides in aqueous wastes significantly increases the complexity and cost of treating the waste for disposal (1). Incidental wastes resulting from processes utilizing nuclear fission may contain radionuclides (especially

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cesium and strontium) or lanthanide elements (Ce, Sm, Eu, for example) that require remote handling of the waste and process equipment. Other wastes that contain actinide elements (U, Np, Pu, Am or Cm) require, at the minimum, extensive containment controls, if not limited shielding to handle the waste. Separation of the major radioactive elements from the waste allows downstream processing of the bulk non-radioactive fraction of the waste to be performed in less expensive equipment and facilities with hands-on operation and maintenance. Separation technologies also concentrate the radionuclides destined for geologic disposal into a smaller volume. This lowers capital and operating costs of treatment facilities as well as final storage and disposal costs. Toward this end, an array of robust separation technologies are either in use or are in various stages of development in the United States and around the world. Among such technologies, liquid-liquid extraction, ion exchange, and precipitation continue to dominate the nuclear industry. As if to underscore this point, the recent selection of candidate technologies for cesium removal from alkaline waste at the United States Department of Energy Savannah River Site resulted in evaluation of one liquid-liquid extraction technology, one ion exchange technology, and one precipitation technology (2). At the Idaho National Engineering and Environmental Laboratory (INEEL), both ion exchange and LLE technologies are candidates for treatment of existing tank wastes.

Liquid-liquid extraction was initially utilized for separations of organic compounds (3) and beginning in the 1930's gained favor in the petroleum industry, where it has since found high-volume applications. The ability to separate inorganic compounds such as uranyl nitrate by LLE gradually evolved over the past two centuries. Inspired by the high selectivity that can be attained, analytical chemists developed the concept of an extractant in the first part of the past century. Industrial use of LLE for metal ion separation began in earnest with the need to separate uranium from ore and to recover metals from spent fuel in the nuclear industry. It has since been profitably utilized in numerous large-scale applications including petroleum, hydrometallurgical, pharmaceutical, food, and nuclear industries (3,4).

Liquid-liquid extraction provides a facile method for separating components of a solution by utilizing an unequal distribution of the components between two immiscible liquid phases. In most cases, this process is carried out by intimately mixing the two immiscible phases, allowing for the transfer of components from one phase to the other, then allowing the two phases to separate. Since huge interfacial areas are quickly generated on mixing, mass transfer can be practically instantaneous. Typically, one phase will be an aqueous solution, usually containing the components to be separated, and the other phase will be an organic solvent containing an extractant, which selectively effects a high distribution for some specific components of the solution. The process is reversible by contacting the solvent loaded with solute(s) with another immiscible phase that has a higher affinity for the solute than the organic phase. The transfer of solute from one phase into the solvent phase is referred to as extraction and the transfer of the solute back from the solvent to the second phase is referred to as back-extraction or stripping.

Certain compelling advantages of LLE have made it a workhorse separation technique in the nuclear industry and offer potential solutions to current problems. Liquid-liquid extraction can be performed on a continuous basis, by assembling a number of contactors (equipment to mix and separate the phases) with the immiscible phases flowing countercurrently between contactors. Continuous operation, with the capability for high throughputs and separation efficiencies has led scientists and engineers to use LLE technologies for production-scale

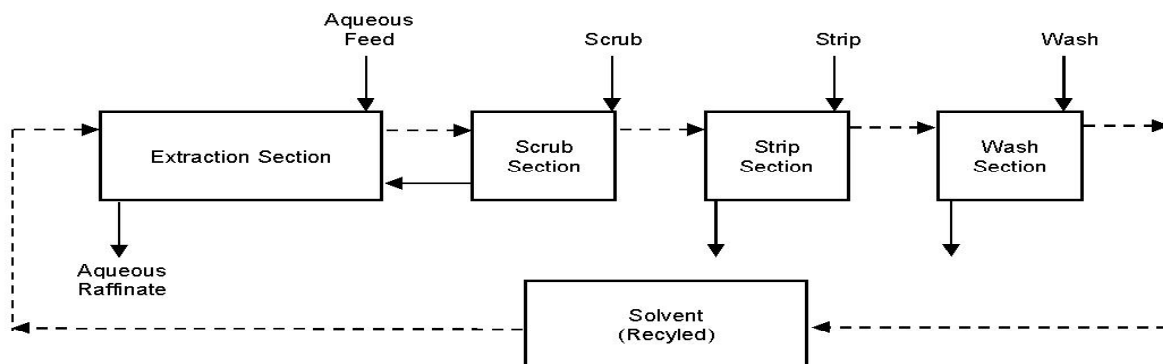
applications. The ability to change the number of stages, vary flow ratios, and introduce multiple scrubbing and stripping stages makes LLE extremely flexible to handle changing needs, even within an existing plant. For separations in the nuclear industry, all ambient temperature-liquid operation reduces process complexity and facilitates remote handling. A diverse chemistry, both off-the-shelf and emerging from the laboratory, gives enormous breadth to the possible problems that can be tackled. The chemistry of LLE for the separation of metals has grown markedly in the past 50 years and shows little sign of slowing down. Indeed, hundreds of potential extractants are introduced each year. One can only conclude that the chemical community generally recognizes the economic importance of LLE and its bright prospects.

Despite the strong record and many advantages of LLE technology, recent perceptions about the complexity of LLE processes have led to an increasing trend to avoid LLE processing to treat U.S. DOE radioactive wastes. Proposed separation facility costs at Hanford, INEEL, and the Savannah River Site have ranged from 400 M to over 1 B U.S. dollars. The sheer magnitude of this cost has stimulated intense study and debate concerning cost-saving alternative technologies. Other countries worldwide have dealt with the cost issues and many have included LLE as an integral part of their waste management/treatment strategy. Is LLE too complex, too costly, or otherwise inappropriate for treating legacy radioactive wastes in the U.S.? That question can best be answered by looking at the issues often raised regarding LLE. In the following discussion, we take the position that LLE technology has much to offer and should be included in testing programs related to treatment of nuclear wastes. Although we stress LLE's benefits that have been overlooked in recent U.S. technology development, the intent is to present a balanced perspective based on the state of the current technology, the challenges that must be overcome, and the changes taking place in the LLE field to meet these challenges.

## **PROCESS COMPLEXITY**

Liquid-liquid extraction processes usually employ organic extractants, which may be relatively simple molecules such as tri-n-butyl phosphate, or complex molecules such as macrocyclic polyethers. The development of new extractants can be a complicated science that must be interwoven with consideration for engineering and economic outcomes, including the complexity of the overall process as extended to both up- and down-stream impacts. Once the solvent composition has been developed and demonstrated, however, a viable process can be designed based upon well understood engineering principles. These principles have been developed and applied during over 50 years of operating liquid-liquid extraction processes for nuclear applications. The process will likely involve numerous extraction stages, which can be discrete contactors or lengths of an extraction column. However, the basic flow scheme is relatively simple, as shown in Figure 1. The solvent flows through the bank of contactors while the aqueous streams enter and leave various locations in the bank, flowing countercurrently to the solvent and facilitating extraction, scrub, strip and solvent wash applications. Once solvent and aqueous flows are established and controlled within specified ranges, (typically  $\pm 10-15\%$ ) a LLE process can run continuously for several months without shutdown. Usually operation time is limited by surge capacity between upstream or downstream unit operations. Extraction process performance is sensitive to a relatively small number of ions; therefore the process is usually capable of handling wide variations in feed composition, while providing a very stable and consistent feed product to downstream processes. Liquid-liquid extraction processes are also

capable of very high throughputs, with minimal generation of secondary wastes, because a relatively small volume of solvent can be used and continuously recycled



**Figure 1. General Liquid-liquid Extraction Flow Diagram**

Liquid-liquid extraction processes also offer the opportunity to customize solvent compositions to remove multiple ionic species in a single process. Processes have been successfully developed in recent years to remove and/or partition actinides, cesium, strontium, technetium, lead, mercury, and numerous combinations of the above elements.

It should also be recognized that overall process complexity includes impacts on up- and downstream operations. Secondary waste generation, a problem that just exacerbates the difficulty and expense of nuclear waste disposal, is discussed separately below. Upstream complexity and cost comes about if the feed to the LLE unit operation must first be treated in some way. Other than to require possible filtration of the feed to remove particulates, a common requirement also for ion exchange or membrane separations, successful LLE processes generally operate without adjustments to the feed stream. Indeed, the aim of extractant selection or design is to achieve this very quality, and for examples, one may consider the PUREX and TRUEX processes. Conversely, downstream processes typically deliver a stream readily used with minimal treatment.

All separation processes undergo some degree of "fouling," and for fairness, LLE has its share of difficulties. Fouling creates some process complexity in that operators must divert their attention and resources to remediation activities. Criticism of the LLE technique invariably includes mention of interfacial "crud," impaired phase disengagement, and other performance problems basically caused by impurities. Generally, these problems are efficiency issues dealt with in straightforward manner during extractant design and process development (4). Experience in both the nuclear and hydrometallurgical industries has been that fouling issues are best

uncovered by pilot-scale testing on actual process feed streams. Chemical analysis of fouling problems then leads to obvious solutions. This approach would be no different for process development using other technologies such as ion exchange, precipitation, or membranes.

## **SECONDARY WASTE GENERATION**

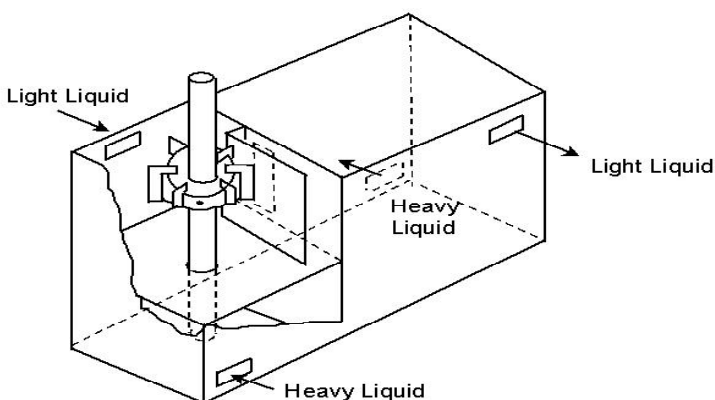
The primary streams exiting a LLE process are the raffinate, product, solvent wash, and ultimately the recycled solvent. During continuous operation, the solvent is recycled, which again, provides for high throughput with a minimum inventory of solvent. The raffinate stream is basically the feed stream minus extracted ions plus the scrub stream (used to remove undesired semi-extractable species from the solvent prior to product stripping). It will typically be a slightly higher volume than the original feed, but, in the case of radioactive waste treatment, may be treated as a low-level waste, and much less costly to immobilize for storage/disposal. The product stream contains the primary radionuclides and is typically a smaller volume, dilute acid stream. The product stream may contain complexants used to help remove the radionuclides from the solvent. In most cases, the product stream can be evaporated and when immobilized, will be only a small fraction of the original radioactive waste volume. Solvent wash streams (used to remove degradation products and prepare the solvent for recycle back to the extraction section) may be treated separately, or combined with the raffinate stream for low-level waste disposal. Final solvent disposal will require incineration at a low-level or mixed waste treatment facility. The useful lifetime of the solvent therefore becomes an important consideration in evaluating LLE technologies. Many of the solvents in use or under development today have been demonstrated to be chemically and radiolytically stable for several years under processing conditions.

Solvent extraction technology often excels in its ability to minimize secondary waste. An early example was the replacement of the bismuth-phosphate precipitation process by solvent extraction, first REDOX and then PUREX. Not only did the solvent extraction permit recovery of uranium in addition to plutonium, it dropped the rate of waste production by more than 30-fold (5). It is no accident that more recently developed processes such as TRUEX, DIAMEX, SREX, SRTALK, and CSEX have received intense interest for implementation in the nuclear industry world-wide. Like, PUREX, these processes operate by a common principle in which the feed is treated with minimal adjustment and stripping is accomplished at low ionic strength. By stripping with dilute solutions or even water, the processes concentrate the contaminants into a matrix that does not create yet another separation problem. In many cases, the strip solutions concentrated in the separated contaminants can be directly vitrified or otherwise treated for disposal. In the hydrometallurgical copper industry, solvent extraction-electrowinning has been widely adopted in large part because stripping can be accomplished by sulfuric acid to produce an ideal feed for electrowinning. In summary, the most successful applications of solvent extraction have generally been those that have minimized secondary waste. When this can be accomplished, the economics of solvent extraction are often superior to other techniques.

## **CONTACTOR TYPES**

There are three basic types of liquid-liquid contacting equipment: mixer-settlers, centrifugal contactors, and columns. The mixer-settler is simply a tank with two chambers; one with a

mixing impeller to mix the phases, the other to let the light and heavy phases separate by gravity (shown in Figure 2).



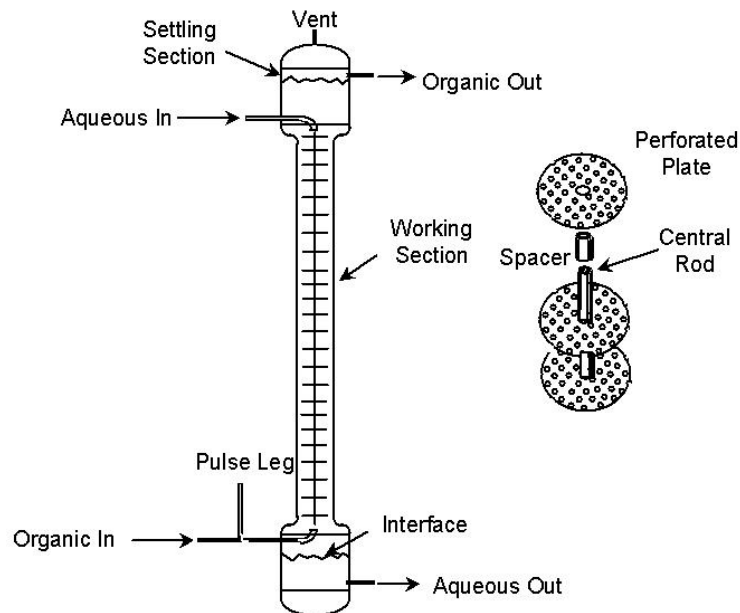
**Figure 2. Mixer-Settler Schematic**

Extraction columns utilize countercurrent flow through a column containing either packing or plates to disperse the phases. Phase separation is accomplished in a disengaging section, usually of larger diameter than the column, located at either or both ends of the column (shown in Figure 3). Centrifugal contactors are similar to mixer-settlers, except use a spinning rotor to mix the solutions and the centrifugal force on the inside of the rotor to separate the phases (shown in Figure 4.). Each type of contactor has inherent advantages and disadvantages, and all have been used in processes with radioactive elements. In past fuel reprocessing processes in the U.S., mixer settlers and centrifugal contactors were used at Savannah River Site and pulse columns were used at Hanford and Idaho. British Nuclear Fuels LTD. (BNFL) THORP facility, which was commissioned in the early 1990's, uses pulse columns for plutonium and uranium recover and purification. Recent developments in centrifugal contactors have resulted in increased acceptability in the nuclear industry, because of smaller facility size requirements, high throughput, rapid achievement of steady-state operation, decreased exposure of the solvent to the waste, and low solvent inventory. Centrifugal contactors also facilitate the high solvent turnover needed to make expensive reagents such as crown ethers and calixarenes economically viable on the process scale. High turnover of such designer reagents is difficult to achieve in ion exchange applications.

## SCALE-UP

Scale-up of extraction equipment is well understood and predictable. Since the function of an extraction contactor is to mix two immiscible phases together, and then separate them, known engineering principles can be applied.

In mixer-settlers, the mixing step is a function of mechanical energy input per volume. This may also have some dependency on the geometry of the mixing zone and the physical properties of solution, but these are usually fixed in a process. The settling step is a function of geometry (interfacial surface area), time, and physical properties of solutions.



**Figure 3. Pulse Column Schematic**

Pulse columns with a known height of a theoretical stage unit (HTU) can be easily scaled from small diameter columns (1-2 inches) to larger columns (1-2 feet). For fixed operational variables such as pulse amplitude-frequency, the primary variable in scaling pulse columns is the superficial (volume) velocity, expressed in gal/hr-ft<sup>2</sup> or m/s. Pulse column flooding curves are usually plotted as volume velocity vs amplitude-frequency product and these curves provide a

region for operation. Typically, columns are operated at approximately 80% of flooding for a given amplitude-frequency product.

Centrifugal extractors have an operating region similar to pulse columns. The operating range is defined by the volume throughput of both phases as a function of energy input (rotor rpm). Because centrifugal contactors operate with high stage efficiencies (typically >90%), designing process flowsheets is greatly simplified. Experimental contactors have been designed and operated at rotor sizes as low as 1-2 cm diameter. These small sizes allow testing with small feed volumes; however, because solution flows are very small, and often non-continuous due to solution surface tension, actual stage efficiencies may be low (60-80%). In larger-scale contactors (4-cm and larger), solution flows are sufficient to maintain continuous flow and stage efficiencies often approach 99%. Therefore, flowsheet scale-up from 1-2 cm contactors to larger sizes is conservative, i.e., larger contactors operate at higher stage efficiencies than small-scale contactors.

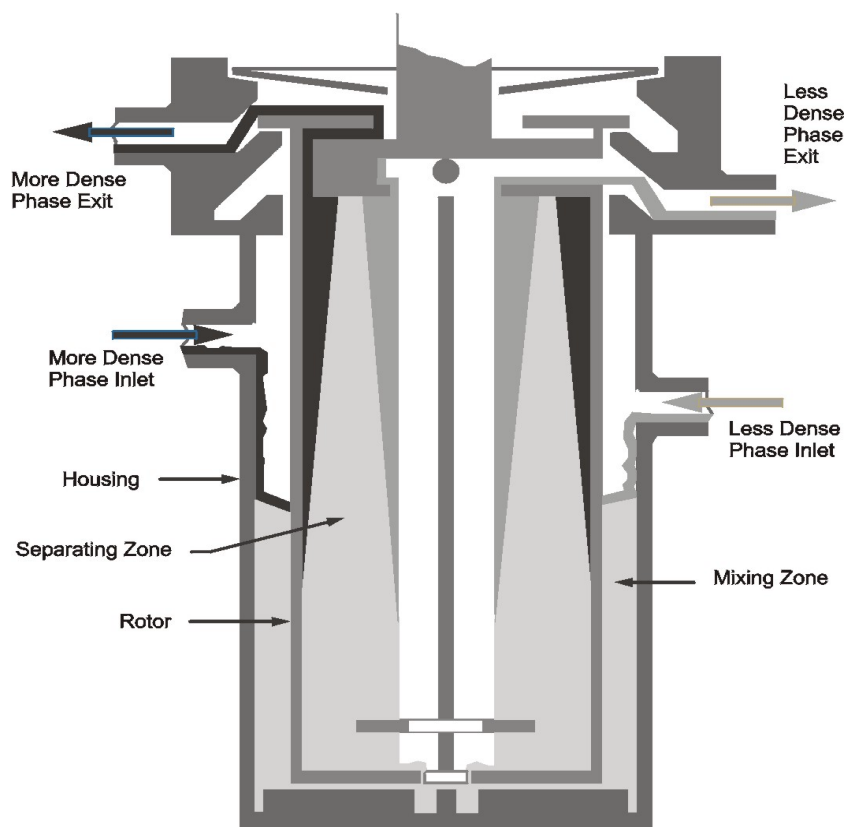


Figure 4. Operating Stage of ANL Centrifugal Contactor

## TECHNICAL MATURITY

Liquid-liquid extraction technologies have been in practice worldwide since the 1930's. They received widespread acceptance and use in the nuclear industry beginning in the early 1950's to recover uranium and plutonium from spent nuclear fuel. This practice continues in the United Kingdom, France, Japan, and Russia, but has been discontinued in the United States since the



early 1990's. With nearly 50 years of research and industrial-scale operating experience, LLE has literally been the workhorse of the nuclear industry, to recover fissionable material. The basic technology is therefore one of the most mature technologies available for waste treatment. The understanding of extractants and solvents varies from very mature to relatively immature. However, considerable work has been performed in the last 10 years to characterize and demonstrate new extractants. Many of these new extractants offer significant improvements in selectivity and solvent stability.

## **EFFICIENCY**

One of the main reasons LLE technologies have been used so widely around the world in the nuclear industry is that high separation efficiency can be obtained at high processing rates. The nature of a countercurrent process allows for much higher separation efficiencies than can be obtained in batch or crossflow processes. The decontamination factor in a countercurrent extraction process can be approximated by taking the extraction distribution coefficient,  $D$ , (concentration of solute in organic phase divided by the concentration of the solute in the aqueous phase) raised to the  $n^{\text{th}}$  power, where  $n$  is the number of extraction stages. For example, if the extraction distribution coefficient is 10, a decontamination factor of  $10^4$  can be achieved in only four extraction stages (assuming  $D$  is constant). Numerous LLE technologies have recently been demonstrated on actual acidic radioactive wastes at the INEEL using a total of 20-24 stages of centrifugal contactors for a complete flowsheet (extraction, scrub, strip and solvent wash) (6-10). A summary of the results from these tests is shown in Table 1. The transuranic extraction process (TRUEX) is very effective at removing actinides, with removal efficiencies ranging from 99.8% to 99.97% for Am and Pu respectively. The strontium extraction process (SREX) effectively partitioned Sr and Pb from INEEL tank waste with removal efficiencies of 99.994% and >94% respectively. Nearly all of the Pu and U in the waste were also removed in the SREX process, because of tributyl phosphate, which is a component of the SREX solvent. A trialkyl phosphine oxide extraction process (POR) was effective at removing plutonium from the INEEL waste (99.97%) but not as effective at removing americium (56.4%). Three variations of a process based on chlorinated cobalt dicarbollide (ChCoDiC) have been demonstrated. The first contained only ChCoDiC in a non-nitroaromatic diluent and demonstrated a cesium removal efficiency of >99.998%. The same process with the addition of polyethylene glycol (PEG) to the solvent facilitated coextraction of strontium had separation efficiencies of 99.3% for Cs and 96.2% for Sr. A universal extraction process (UNEX) based on ChCoDiC, PEG, and CMPO extractants in a non-nitroaromatic diluent demonstrated removal efficiencies of 99.96% for actinides, 99.995% for strontium and 99.4% for cesium.

A clear need for new extractants in the treatment of nuclear waste by LLE arises from the severe demands on separation performance that any technology must satisfy. Feeds are extraordinarily complex and concentrated in salts, acid, or alkali (1). By contrast, contaminants to be removed are present at trace concentrations. For example, at the Savannah River Site, cesium at an average concentration of ca.  $7 \times 10^{-4}$  M is to be removed with a DF of 40,000 from an alkaline waste containing 7 M sodium. Recent flowsheet testing of an alkaline cesium extraction process, developed at Oak Ridge National Laboratory and tested in centrifugal contactors at Argonne National Laboratory on a Savannah River Site supernatant simulant resulted in removal efficiencies of >99.997% (11).

**Table 1.** Summary of the removal efficiencies obtained from demonstrations of various liquid-liquid extraction processes with actual acidic radioactive waste

Component	TRUEX Process	SREX Process	POR Process	ChCoDiC Process	ChCoDiC Process (No PEG)	Universal Extraction Process
Gross Alpha	99.8%	94.0%	98.9%	b	b	99.96%
<sup>241</sup> Am	99.8%	2.0%	56.4%	b	b	B
Pu	99.97%	99.95%	99.97%	b	b	B
U	99.8%	99.6%	b	b	b	B
<sup>90</sup> Sr	b	99.994%	0.03% <sup>a</sup>	96.2%	0.09%	99.995%
<sup>137</sup> Cs	b	0.4%	0.03% <sup>a</sup>	99.3%	>99.998%	99.4%
<sup>99</sup> Tc	89%	b	66.4%	b	b	B
Al	b	0.4%	0.01% <sup>a</sup>	0.03% <sup>a</sup>	b	B
B	b	<23%	1.3% <sup>a</sup>	b	b	B
Ba	b	63.6%	b	b	b	>87.2%
Ca	b	5.0% <sup>a</sup>	0.08% <sup>a</sup>	1.4%	<28%	B
Ce	b	b	68% <sup>a</sup>	b	b	B
Cr	b	b	<15%	b	b	B
Fe	0.7%	1.7%	2.1% <sup>a</sup>	<2.1%	<40%	6.9%
Hg	73.7%	>89.2%	92%	26.4%	37.5%	0.16%
K	b	31.5%	b	45.2%	50%	20.4% <sup>a</sup>
Na	0.07%	0.4%	b	2.1%	0.7%	0.7% <sup>a</sup>
Pb	b	>94%	0.6% <sup>a</sup>	98.5%	b	>98.5%
Zr	42% <sup>a</sup>	>82%	28% <sup>a</sup>	<5.8%	b	97.7%

<sup>a</sup> Data from flowsheet testing with simulated waste.

<sup>b</sup> Data was not obtained.

## COST

Liquid-liquid extraction facilities typically require nominally larger capital expenditures than other separation processes. This is a disadvantage for LLE processes in the nuclear industry, because the Department of Energy is constrained to limited capital dollars, but less so with operating dollars. This leads to consideration of more costly life cycle processes, that require smaller initial capital investment. The primary cost is facility size and/or remote handling. Mixer-settler equipment requires large volumes and facility footprints. Extraction columns require less floor space but need >10 meters in headspace. Centrifugal contactors require much less floor and head space, but require remote capabilities for eventual motor change out. One of the main costs associated with these separation processes, however, is the space required for tankage. Operational flexibility mandates a certain amount of redundant tankage and surge capacity, but these must be evaluated against higher initial capital investment. Tankage requirements may be minimized by using centrifugal contactors because they require little or no surge capacity between upstream or downstream processes and require a much smaller solvent inventory. Operational costs of LLE facilities are equivalent to or less than other nuclear industry processes. Liquid-liquid extraction processes are operated at or near ambient temperature and do

not require extensive off-gas systems or require frequent equipment change out due to corrosion. The usually significant reduction in highly radioactive waste volume also reduces the size and cost of downstream, remotely operated facilities, as well as final high activity waste volumes which are costly to store and dispose.

### **LIQUID-LIQUID EXTRACTION vs ION EXCHANGE**

The current trend in the United States Department of Energy facilities appears to favor ion exchange processes over LLE processes. This choice is often biased by perceptions about LLE that are not based on experimental data or operational history. Both technologies have advantages and disadvantages and the choice should be made based on which process will work best for a given application. For example, LLE processes are better suited for treatment of large volumes of waste, at high throughputs, since the amount of secondary waste is minimized by recycle of the solvent. Ion exchange processes are well suited to removal of ions from dilute solutions of limited volumes. In both cases, there are numerous choices of extractants or sorbents that are highly selective for many metal ions of interest. There are cases where one technology will not effectively remove an ion where the other technology will. One example of this is the removal of strontium from acidic solutions of high ionic strength. Two LLE processes based on crown ether or cobalt dicarbollide/PEG extractants are highly effective. The only effective sorption method for Sr under these conditions is extraction chromatography using the same extractants on solid supports, but these have significantly less capacity than the analogous LLE process. With respect to facility size and complexity, a simple once through single column system is less complex than a bank of centrifugal contactors. However, if you use multiple columns or regenerate the sorbent in the columns, the ion exchange system can quickly become more complex than a single bank of contactors and require more manpower to operate the system.

Liquid-liquid extraction processes can be designed to achieve higher separation efficiencies than ion exchange processes, because of the semi-continuous operation of ion exchange processes. To achieve high separation efficiencies in an ion exchange process, the sorbent must be replaced or regenerated as soon as breakthrough is detected. This utilizes only a small fraction of the ion exchange sorbent capacity and increases secondary waste generation.

There is also a perception that the “solvents” used in LLE pose a more significant environmental threat than ion exchange resins. This is simply not true. Because the solvents can be recycled efficiently within the process, the solvent inventory is relatively small. Also, if centrifugal contactors are used, radiolytic degradation of the solvents can be minimized. Ion exchangers are directly loaded with the radioactive target ion, and are thus subjected to significant radiolysis. The required mass of ion exchanger to achieve a given separation is often much more than that of an analogous extraction solvent. The likely disposal path for either organic-based ion exchangers or extraction solvents is incineration. The primary incineration products are the same in either case – carbon dioxide and water. Spent inorganic ion exchange sorbents must be immobilized in grout or glass and sent to a long-term storage or disposal facility, resulting in additional waste volume and disposal cost.

## **POTENTIAL FOR FUTURE USE OF LLE**

At the present time, the future use of LLE for treatment of radioactive wastes is in question in the U.S. but there are significant development and demonstration efforts in progress. The largest obstacle in the U.S. is perhaps the negative image LLE has received because it has been tied to reprocessing of spent nuclear fuel. The potential for reprocessing in the U.S. in the next 10-15 years is very low, but LLE continues to be used in the UK, France, Japan, and Russia to reprocess spent nuclear fuel. A common argument against LLE is related to proliferation; however, numerous technologies exist to render the recovered fissionable material from reprocessing as self-protecting or non-recoverable for use.

The Savannah River Site and the INEEL are both evaluating LLE technologies for the treatment of tank wastes. The technology selection process is in progress at both sites and the final outcome has not been determined at this time. Demonstration of numerous LLE technologies have been successfully completed in countercurrent pilot plants using simulated and actual wastes. Liquid-liquid extraction technology has been successfully implemented to treat radioactive waste on a large scale in Russia. The Mayak production association, in Ozersk, Russia has used LLE to process over 500,000 L of high-level tank waste.

## **SUMMARY**

Liquid-liquid extraction technologies are obviously not the solution to all waste treatment problems. They do offer significant advantages over other separation processes such as ion exchange or precipitation in many situations, particularly where high volume, high throughput and high separation efficiencies are required. Because LLE is more of a “chemical” process as opposed to a “physical/chemical” process it is less understood by the public and management than other technologies. The average person relates ion exchange technology to their home water softener, but has no comparable analogy for LLE. The scientific community has done a less-than-adequate job of presenting LLE to management and the public in a manner that they can understand or embrace. The highly successful use of LLE technology for spent fuel reprocessing and fissile material recovery throughout the world for over 50 years indicates that LLE is an appropriate technology for treatment of legacy nuclear wastes, but also ties a stigma of nuclear reprocessing to it. We must remove this stigma and demonstrate to decision-makers and stakeholders the advantages LLE offers in waste treatment. This would best be accomplished by demonstrating the successful use of LLE in a full-scale process to solve waste treatment problems; however, it is somewhat of a catch-22 situation. The technical community needs to draw on successful applications and uses of LLE outside the nuclear industry, including “green” applications to demonstrate the potential and advantages over competing separation technologies.

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**WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ**

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