INL/CON-05-00088
PREPRINT

Design Attributes and Scale Up Testing of Annular Centrifugal Contractors

Chemical Engineering Division of the AIChE Spring National Meeting

D. H. Meikrantz
J. D. Law
T. A. Todd

April 2005

Paper 182-F

Design Attributes and Scale Up Testing of Annular Centrifugal Contactors

D.H. Meikrantz, J.D. Law, and T.A. Todd
Idaho National Laboratory, Idaho Falls, ID 83415

Presented at the Chemical Engineering Division of the AIChE Spring National Meeting April 10-14, 2005, Atlanta, GA

Abstract

Annular centrifugal contactors are being used for rapid yet efficient liquid- liquid processing in numerous industrial and government applications. Commercialization of this technology began eleven years ago and now units with throughputs ranging from 0.25 to 700 liters per minute are readily available. Separation, washing, and extraction processes all benefit from the use of this relatively new commercial tool. Processing advantages of this technology include: low in-process volume per stage, rapid mixing and separation in a single unit, connection-in-series for multi-stage use, and a wide operating range of input flow rates and phase ratios without adjustment.

Recent design enhancements have been added to simplify maintenance, improve inspection ability, and provide increased reliability. Cartridge-style bearing and mechanical rotary seal assemblies that can include liquid-leak sensors are employed to enhance remote operations, minimize maintenance downtime, prevent equipment damage, and extend service life. Clean-in-place capability eliminates the need for disassembly, facilitates the use of contactors for feed clarification, and can be automated for continuous operation.

In nuclear fuel cycle studies, aqueous based separations are being developed that efficiently partition uranium, actinides, and fission products via liquid-liquid solvent extraction. Thus, annular centrifugal contactors are destined to play a significant role in the design of such new processes. Laboratory scale studies using mini-contactors have demonstrated feasibility for many such separation processes but validation at an engineering scale is needed to support actual process design.

Introduction

Development of the annular centrifugal contactor (ACC) by researchers and engineers at the U.S. government laboratories began in the early 1960s. The basic design is credited to Webster, who worked at the Savannah River Laboratory (1,2). Bernstein et al., at Argonne National Laboratory, added the annular mixing zone (3). Another Argonne researcher, Ralph Leonard, led the development of many practical contactor concepts and improved upon the design and utility (4,5). Research and process development with contactors continues at the national laboratories and has progressed into commercial applications (6).

Commercialization of annular centrifugal contactor technology began eleven years ago with the technology transfer of a patent from the Department of Energy's Idaho National Engineering and Environmental Laboratory (7). Since that time, a number of design enhancements have been made and patented that led to a device better suited to a variety of liquid-liquid processes. Multiple sizes were designed to provide total throughput ranging from 0.25 to 700 liters per minute (8,9). Interchangeable heavy phase weir rings were incorporated into the rotor to allow separation of a wide range of density pairs. A low mixing sleeve was added to aid in direct separation of viscous and difficult liquids (10). Clean in place (CIP) capability was achieved by adding a hollow central shaft with spray nozzles to the rotor (11).

The resulting annular centrifugal contactor is a low rpm, 100-600g centrifuge powered by a direct drive, variable speed motor. A variable frequency drive is used to control speed, start-up, shut down and also conveniently display other electrical status information. Properly optimized, this centrifuge can efficiently separate two immiscible liquids of differing densities throughout a 100% change in feed ratio and flow rate without adjustment.

Laboratory testing of highly radioactive feeds using mini-contactors of similar design has been conducted on numerous flowsheets related to nuclear fuel cycle studies. Recent work at such locations as: Argonne National Laboratory, Idaho National Laboratory, Savannah River Laboratory, in France, and in China has successfully demonstrated high separation and decontamination factors for many elements using a variety of organic extractant solvents (12-16). Vandegrift et al also reported the results of nuclear fuel cycle multi-flowsheet testing at the 2004 Waste Management Conference (17).

Difficulties related to maintaining consistent flow and stage efficiency are observed due to the small inlets, outlets and orifices inherent in mini-contactors having 1 or 2 cm. diameter rotors. Such limitations make it difficult to predict how many stages will be required to meet the separation goals in both pilot and full scale processing applications. Therefore, testing with larger units has been suggested to support process facility designs. Leonard et al summarized some hydraulic performance problems while using 2 cm contactors and indicated that most are resolved in units of 4 cm diameter or greater (18,19). He also reported on the hydraulic performance of a 5 cm contactor and found no indication of the typical mini-contactor types of efficiency losses due to such phenomena as slug flow or phase inversion (20). However, single stage efficiencies were not directly measured on the 5 cm unit during this study. Testing at the INL will be conducted using 5-25 cm rotor contactors to further address these issues and several others.

Contactor Design and Operation

The annular centrifugal contactor can be operated as both a separator and a contactor, which make it a valuable tool in numerous processes. Its unique design provides mixing and separation in a single, compact unit. **Figure 1** gives a cutaway view of the centrifuge housing and rotor with the significant design features including the liquid flow path.

Two immiscible liquids of different densities are fed to the separate inlets and are rapidly mixed in the annular space between the spinning rotor and stationary housing. Please note that the areas above the liquid levels in the contactor housing and at the center of the rotor are vapor space. Incoming mixed phases are directed toward the center of the rotor bottom by radial vanes in the housing base. As the liquids enter the central opening at he base of the rotor, they are accelerated toward the inner diameter wall. The self-pumping rotor is divided into four vertical chambers and is dynamically balanced by the pumped liquids. The mixed phases are rapidly accelerated to rotor speed once trapped in a quadrant and separation occurs at elevated g force as the liquids are displaced upward by continued pumping.

The separating zone extends from the diverter disk to the lighter phase weir. It provides a residence time for the liquid-liquid interface to form and sharpen. The interface should be positioned half way between the lighter phase weir and the heavier phase underflow at the top of the separating zone by selecting the proper size heavy phase weir ring. Optimum performance is thus achieved despite changes in flow rate or liquid ratios because the interface position can shift a significant distance without loss of separation quality. Because the interface is free to adjust in position, it is important to keep the liquid discharges unrestricted in terms of liquid flow, vapor flow, and pressure. Equilibration of pressure between the centrifuge housing, discharge pipes, and receiver tanks ensures trouble-free operation over a wide range of process conditions.

Performance parameters that can be adjusted and optimized for various two-phase processes include: heavy phase weirs, rotor speed, input phase ratio, input flow rate and thus separation residence time, and mixing mode. Input phase ratios of 0.1 to 10 are readily processed with both good mixing and separation performance at all but the lowest feed rates. Evaluation of total feed rates of less than 10% of the hydraulic limit is advisable in extraction processes to ensure adequate mixing for high stage efficiency. For direct separations, in either high or low mix mode, there is no turn down limit for the feed.

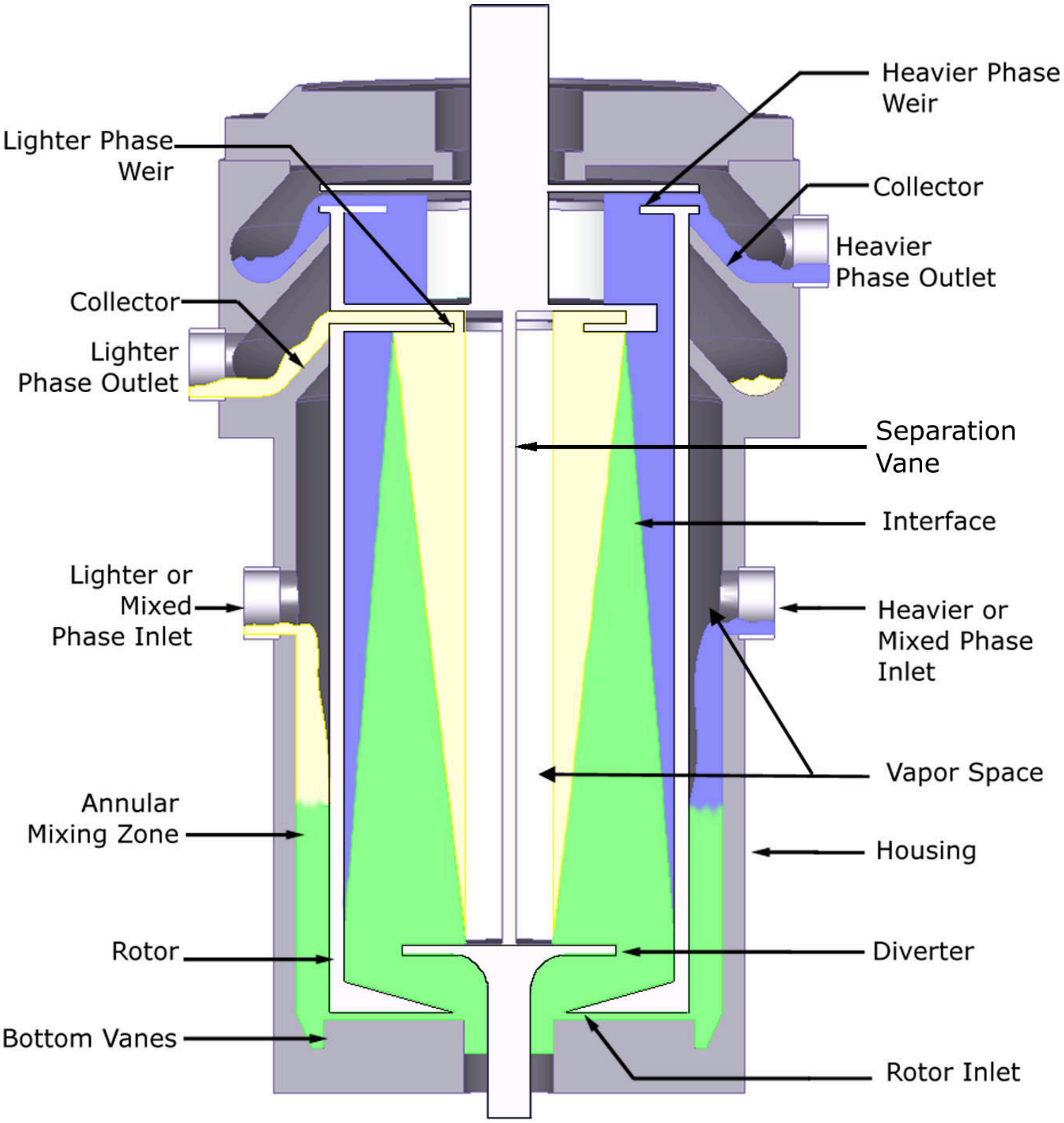

Figure 1. Cutaway view of the ACC

Low Mixing Sleeve

In processes requiring only two-phase separation or when shear sensitive liquids are being processed, excess mixing in the annulus should be avoided. Therefore, a low mixing sleeve, a cylinder slightly larger in diameter than the rotor, has been developed. It is attached to a modified bottom plate of the centrifuge housing and is easily exchanged for the high mix version. A view of these two mixing options and their location in the ACC is provided in **Figure 2**. The low mix sleeve encases the rotor preventing the feed liquids from being mixed by the spinning rotor's outer surface.

Feed is diverted to the new annulus formed between the outside of the low mix sleeve and the inside diameter of the lower housing. Radial vanes direct the feed liquids to the center of the rotor bottom and into the rotor entrance aperture. A lip seal is installed into the base of the low mix sleeve to prevent liquids from bypassing the rotor entrance and entering the high shear area between it and the spinning rotor. Limited mixing still occurs as the liquids enter and are pumped by the rotor, useful in certain washing applications.

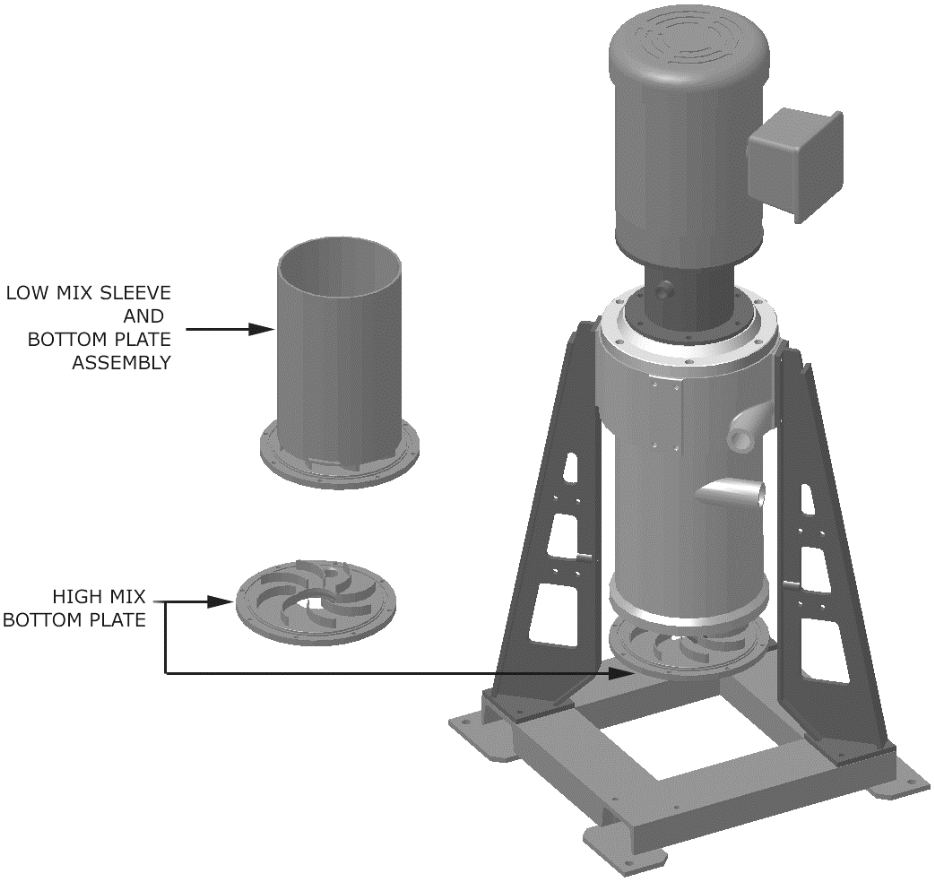

Figure 2. High and low mixing options

Clean-in-Place Design

The ACC, despite low rotor speeds, is an efficient collector of solids due to the relatively long separating residence time provided by this design. Solids accumulate uniformly along the inside diameter of the rotor cylinder from the diverter disk to the heavy phase weir. Over time, they will form a thick layer sufficient to interfere with flow of heavy phase through the heavy phase under- flow slots at the top of the separating zone. When this occurs, heavy phase discharge is diverted over the light phase weir and separation quality suffers. Frequent disassembly for cleaning or inspection is both impractical and inconvenient, especially in remote applications.

A clean-in-place (CIP) rotor design was implemented as shown in **Figure 3**. It employs a hollow rotor shaft that protrudes from below the contactor housing and extends into the upper rotor assembly. A set of high-pressure spray nozzles is used to clean each rotor quadrant, simultaneously. Flat patterned spray heads are used in the weir areas to avoid contamination of the product discharge areas by cleaning solutions. This process is performed off-line with the rotor drained and stopped. A rotary union, attached to the bottom of the rotor shaft, provides a permanent inlet for cleaning fluids and allows CIP cycles to be fully automated.

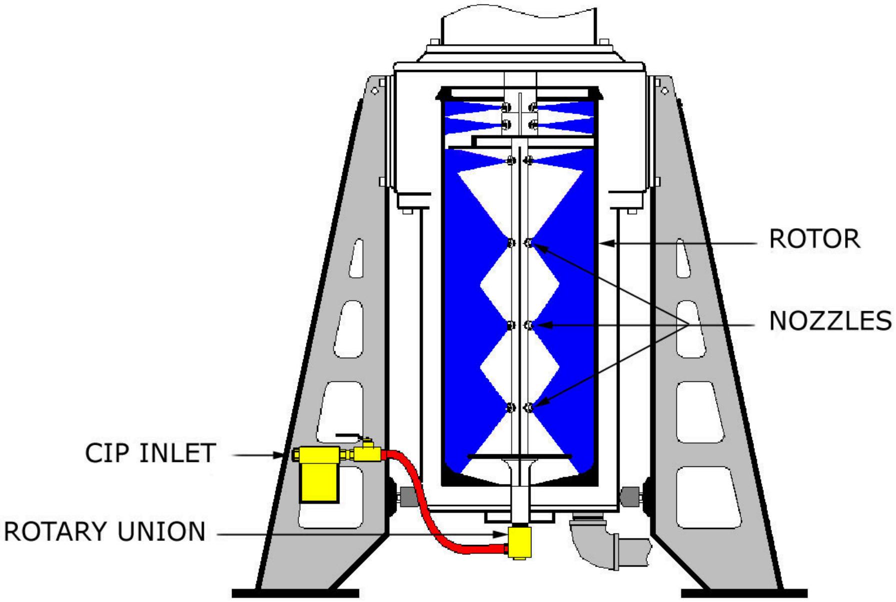

Figure 3. Clean-in-place cross-sectional view

Cartridge Style Bearing and Seal Assemblies

New designs that allow rapid and convenient maintenance of the lower bearing and mechanical seal have been developed to enhance the utility of large contactors. An isometric drawing of this new assembly is shown in **Figure 4**. The cartridge assembly has two major advantages when compared to separately mounted seals and bearings. Prior designs, based upon two-part rotary seals, required rotor removal to complete seal servicing because the upper seal half was attached to the lower rotor shaft near the diverter disk. As such it was inaccessible through the bottom of the housing. Instead, the cartridge is serviced from the outside and only requires the removal of four bolts to change. The rotor does not need to be removed and the service time is reduced by at least a factor of three. A spare cartridge assembly can be installed and the used assembly can be refurbished at facilities elsewhere. There is little chance of accidental seal damage when removing or installing the cartridge seal assembly as the seal faces are enclosed within the assembly. This feature is especially useful for large contactors that need the stability of a tail shaft and are employed in remote or hazardous environments.

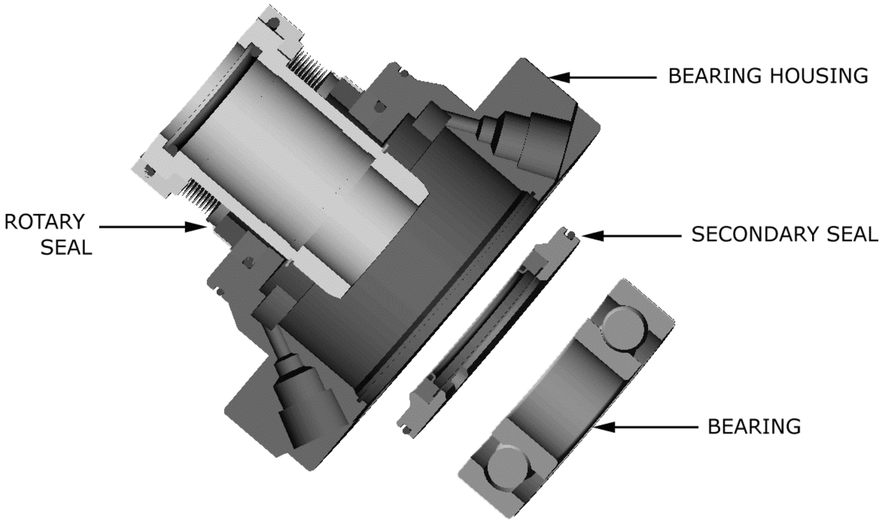

Figure 4. Cartridge bearing and seal assembly

Contactor Scale-up Testing

A testing laboratory for the study and evaluation of 5-25 cm diameter contactor performance at flow rates from 0.25 to 100 liters per minute is being established at the Idaho National Laboratory. The goal is to determine reliable operating parameter ranges, correlate laboratory data to engineering scale process flow sheets, and input design data for the successful implementation of contactor based fuel cycle tests and facilities.

In addition to the previously mentioned limitations of using mini-contactor tests for design input to large scale facilities, other scale-up issues need to be addressed. Although physical dimensions for scaling up contactors are linear, the process parameters that result are not. Contactor throughput increases by a factor of six as the rotor diameter doubles and increased separation residence time is also observed. Annular mixing energy increases with rotor diameter, even as rotor speed is lowered to maintain consistent relative centrifugal force (RCF) for separation. **Table 1** provides a brief comparison as an example of important scaling issues.

Table 1. Scale up comparison of several contactor rotor sizes

Rotor Size	RPM	RCF (g)	Mixing Speed ft/s*
2 cm	3600	85	12.4
5 cm	3600	339	31.4
5 cm	2767	200	24.1
12.5 cm	1713	200	35.9
25 cm	1211	200	50.8

* Linear rate at outside diameter of rotor.

The typical rotor speed used for most 2 cm mini-contactor tests is 3600 rpm. Despite the relative low RCF and mixing energy imparted on the liquids, good performance is possible. Single stage efficiencies of 95% are achievable indicating good phase mixing and separation. However, such rotor speeds are not required for larger sizes. Although the higher RCF values are attractive, the mixing energy can increase to levels that create small droplets not readily resolved by enhanced gravity alone. Once this occurs, an interface layer will build in the rotor that will eventually contaminate one or both discharged phases. Loss of contactor efficiency will result and if multiple units are interconnected, the problem is magnified. Droplet size analysis profiling at various operating conditions for each size can provide assurance that this third phase formation is avoided.

It is desirable to determine the range of optimal rotor speed, mixing, and throughput rate that will yield high stage efficiencies. Matching the parameters obtained using mini-contactors is a starting point but further characterization for each size contactor is required. For example, centrifugal contactors discharge at levels slightly elevated from the inlets because the rotor is a pump. Rotor speed drives the pumping rate generated by each rotor size but must also be balanced with mixing energy imparted to the liquids.

Near-term testing will be conducted on four 5cm and two 12.5 cm contactors. Hydraulic testing will be performed to determine the operating parameter range for both single and multi-unit applications. Heavy phase weirs sizes will be selected for candidate two-phase density pairs and flow ratios expected for flowsheets being studied. Preliminary mass transfer efficiency studies will also begin and preparations for conducting droplet size profiles for large contactors are planned. Initial tests using the low mix option will be conducted for improved coalescence under selected applications. The 12.5 cm units, equipped with CIP, will also be evaluated for potential use in feed clarification.

Summary

Numerous industry-driven design enhancements have been incorporated to provide a commercially robust annular centrifugal contactor. This technology is gaining a foothold in many facets of liquid-liquid processing due to its size, efficiency, simplicity, and ease of use. ACCs are destined to play a significant role in the design of future nuclear processes. Their flexibility promotes rapid change from one set of operating parameters to another. Recently added features extend their applications to certain solid liquid separations for feed clarification and improve utility in hazardous and remote operations.

Studies are in progress to characterize the performance curves and separating efficiencies of pilot and process scale contactors. The goals are to establish reliable operating parameter ranges and conditions that will allow engineers to design future high throughput separation facilities. In addition, design enhancements that promote improved utility in remote environments will also be considered.

Acknowledgements

This work was carried out under the auspices and financial support of the U. S. Department of Energy, Office of Nuclear Energy, Science and Technology through contract DE-AC07-99ID13727.

References

1. M. W. Davis and A. S. Jennings, “Equipment for Processing by Solvent Extraction,” *Chemical Processing of Reactor Fuels*, J.F. Flagg, editor, (Academic Press, New York, 1961) pp. 271-303.
2. D. S. Webster, A. S. Jennings, A. A. Kishbaugh, and H. K. Bethmann, “Performance of Centrifugal Mixer-Settler in the Reprocessing Of Nuclear Fuel,” *Recent Advances in Reprocessing of Irradiated Fuel, Nuclear Engineering, part XX*, (American Institute of Chemical Engineers, New York, 1969) Chem. Eng. Prog. Symp. Ser., No. 94, Vol. 65, pp. 70-77.
3. G. J. Bernstein, D. E. Grosvenor, J. F. Lenc, and N. M. Levitz, “ A High Capacity Annular Centrifugal Contactor,” *Nucl. Technol.* **20**, 200-202 (1973).
4. R. A. Leonard, G. J. Bernstein, A. A. Zeigler, and R. H. Pelto, “Annular Centrifugal Contactors for Solvent Extraction,” *Sep. Sci. Technol*. **15**, 925-943 (1980).
5. R. A. Leonard, “Recent Advances in Centrifugal Contactor Design,” *Sep. Sci. Technol.* **23**, 1473-1487 (1988).
6. D. H. Meikrantz, S. B. Meikrantz, and L. L. Macaluso, “Annular Centrifugal Contactors for Multiple Stage Extraction Processes”, *Chem. Eng. Comm.* **188**, 115-127, (2001).
7. D. H. Meikrantz, “Method for Separating Disparate Components in a Fluid Stream”, U. S. Patent 4,959,158, (1990).
8. D. H. Meikrantz, L. L. Macaluso, H. W. Sams, C. H. Schardin, and A. G. Federici, “Centrifugal Separator”, U. S. Patent 5,591,340, (1997).

9. D. H. Meikrantz, L. L. Macaluso, H. W. Sams, C. H. Schardin, and A. G. Federici, “Centrifugal Separator”, U. S. Patent 5,762,800, (1998).
10. D. H. Meikrantz, A. G. Federici, L. L. Macaluso, and H. W. Sams, “ Rotor Sleeve for a Centrifugal Separator”, U. S. Patent 5,571,070, (1996).
11. L. L. Macaluso and D. H. Meikrantz, “ Self Cleaning Rotor for a Centrifugal Separator”, U. S. Patent 5,908,376, (1999).
12. R.A. Leonard, S.B. Aase, H.A. Arafat, C. Conner, D.B. Chamberlain, J.R. Falkenberg, M.C. Regalbuto, and G.F. Vandegrift, “Experimental Verification of Caustic-Side Solvent Extraction for Removal of Cesium from Tank Waste”, Sol. Extr. And Ion Exch. **12** (4), 505-526 (2003).
13. R.S. Herbst, J.D. Law, T.A. Todd, V.N. Romanovskii, V.A. Babain, V. M. Esimantovski, B.N. Zaitsev, and I.V. Smirnov, “Development and Testing of A Cobalt Dicarbollide Based Solvent Extraction Process for the Separation of Cesium and Strontium from Acidic Tank Waste”, Sep. Sci. Technol. **37** (8), 1807-1831 (2002).
14. M.A. Norato, S.G. Campbell, M.L. Crowder, M.W. Geeting, G.F. Kessinger, R.A. Pierce, and D.D. Walker, “High Level Waste Demonstration of the Caustic-Side Solvent Extraction Process with Optimized Solvent in the 2-Cm Centrifugal Contactor Apparatus Using Tank 37H/44F Supernate”, WSRC-TR-2002-00243, Rev.0, Nov. 1, (2002).
15. N. Simon, B. Tournois, S. Eymard, G. Volle, P. Rivalier, J. Leybros, J.-Y. Lanoe, N. Reynier-Tronche, G. Ferlay, J.-F. Dozol, “ Cs Selective Extraction from High Level Liquid Wastes with Crown Calixarenes: Where Are We Today?”, Conference Atalante, Nimes, June, (2004).
16. J. Wang, G. Tian, J. Chen, and C. Song, “ Application of Miniature Centrifugal Contactors for Treating High-Level Liquid Waste,” Sep. Sci. Technol. **36** (13), 2983-2996 (2001).
17. G.F. Vandegrift, M.C. Regalbuto, S.B. Aase, H.A. Arafat, A.J. Bakel, D.L. Bowers, M.A. Clark, J.W. Emery, J.R. Falkenberg, A.V. Guelis, L.D. Hafenrichter, R.A. Leonard, C. Pereira, K.J. Quigley, Y. Tsai, M.H. Vander Pol, J.J. Laidler, “Lab-Scale Demonstration of the Urex + Process,” paper WM-4323, Proceedings of WM’04, Tucson, AZ, Feb 20-Mar 4, (2004).
18. R. A. Leonard, C. Conner, M.W. Libertore, J. Sedlet, S.B. Aase, G.F. Vandegrift, P.V. Bonnesen, and B.A. Moyer, “ Development of a solvent Extraction Process for Cesium Removal from SRS Tank Waste,” Sep. Sci. Technol., **36** (5&6), 743-766 (2001).
19. R. A. Leonard, S.B. Aase, H.A. Arafat, C. Conner, D.B. Chamberlain, J.R. Falkenberg, M.C. Regalbuto, and G.F. Vandegrift, “ Experimental Verification of Caustic-Side Solvent Extraction for Removal of Cesium from Tank Waste,” Sep. Sci. Technol. **21** (4), 505-526 (2003).
20. R.A. Leonard, M.C. Regalbuto, G. F. Vandegrift, S. B. Aase, H.A. Arafat, and J.R. Falkenberg, “ Hydraulic Performance of a 5-cm CINC Contactor for Caustic-Side Solvent Extraction, ANL-02/18; Argonne National Laboratory: Argonne, IL. (2001).