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Tests of a Higgins Contactor for the Engineering-Scale Resin Loading of Uranium

R. D. Spence
P. A. Haas

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CHEMICAL TECHNOLOGY DIVISION

HTGR FUEL RECYCLE PROGRAM

Refabrication Development — Task 500

TESTS OF A HIGGINS CONTACTOR FOR THE ENGINEERING-SCALE
RESIN LOADING OF URANIUM

R. D. Spence
P. A. Haas

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TESTS OF A HIGGINS CONTACTOR FOR THE ENGINEERING-SCALE RESIN LOADING OF URANIUM

R. D. Spence and P. A. Haas

ABSTRACT

The loading of uranium on weak-acid ion exchange resin is a basic step in the production of fuel particles for high-temperature gas-cooled reactors (HTGRs). In the work reported here, an engineering-scale continuous resin loader (2-in.-ID Higgins contactor) was tested with existing engineering-scale process equipment. The Higgins contactor was first successfully used to convert Na^+ -form resin to the H^+ -form; then it was evaluated as a uranium loader. Results show that the 2-in.-ID Higgins contactor can easily load 25 kg of uranium per day, indicating that a 4-in.-ID contactor could load 100 kg/day. Process control was achieved by monitoring and controlling the density, pH, and inventory volume of the uranium feed solution. This control scheme is amenable to remote operation.

1. INTRODUCTION

An HTGR fuel block consists of fuel rods contained in holes machined in a carbon block of hexagonal cross section. These fuel rods are composed of fissile and fertile fuel particles (called kernels) embedded in a carbon matrix.¹⁻³ The present work concerns only fissile kernels (i.e., kernels that consist of a core of uranium oxide and carbide surrounded by successive layers of porous carbon, dense carbon, silicon carbide, and dense carbon). To prepare these kernels, ^{233}U or ^{235}U is loaded on weak-acid ion exchange resin, the loaded resin is pyrolyzed and converted to yield the uranium oxide-carbide core, and the core is coated with the successive layers just mentioned. This report deals only with cold development of the first step, loading the weak-acid resin with natural or depleted uranium.

The complete loading process involves converting a commercial cation exchange resin, which is in the Na^+ -form, to the H^+ -form and producing uniform-sized microspheres of resin that are fully loaded with UO_2^{2+} , ready for the pyrolysis step.⁴⁻⁷ The commercial resin must be screened to obtain a particular size range, shape-separated to obtain spheres, contacted with acid-deficient uranyl nitrate to load the uranium, and dried to produce the loaded microspheres ready for pyrolysis. Engineering-scale studies of the contacting process have been reported elsewhere.^{4,6} In those studies, resin was contacted with acid-deficient uranyl nitrate (ADUN) in a batch fluidized contactor. The ADUN was generated by extracting nitric acid from the uranyl nitrate solution with an organic amine. The organic amine was regenerated with an aqueous base solution (e.g., NaOH) to give a waste stream containing the nitrate ions (e.g., NaNO_3 solution).

Compared with a batch loader, a continuous loader offers the advantage of high capacities within the dimensions dictated by nuclear criticality. It also provides steady-state operation and countercurrent flow, maximizing uranium loading or conversion efficiency. The feasibility of continuous uranium loading has previously been demonstrated on the laboratory scale using a 1-in.-ID Higgins contactor.⁵ The purpose of the work reported here was to build and test a scaled-up version of this laboratory contactor for use with the engineering-scale loading system. The 2-in.-ID Higgins contactor constructed for this purpose was first used to convert Na^+ -form resin to the H^+ -form; then it was evaluated as a uranium loader to demonstrate contactor capacity and process-control procedures.

The presence of ^{232}U in the recycle HTGR fuel makes remote operation of resin loading necessary. Therefore, extension of the process-control procedures to remote operation was considered, although remote operation was not actually attempted. Also, the reliability of the column parts and of the operation (from the standpoint of minimal maintenance and remote operation) is important and is discussed in this report.

The reprocessing and refabrication of HTGR fuel will be done as a service for various customers with fuels of different isotopic compositions. For this reason, we would expect the Higgins contactor to be operated in a start-and-stop manner. Startup and shutdown procedures, which need to be simple and reliable, were therefore investigated.

2. DESCRIPTION OF HIGGINS CONTACTOR

A Higgins contactor can be described as a semicontinuous, counter-current ion exchange column which operates as a normal packed-bed ion exchange column, except that at regular intervals the flow of solution ceases and the resin bed is moved in the direction opposite to the solution flow. At these intervals, fresh resin is introduced at one end of the bed while loaded resin is removed at the other end. A truly continuous countercurrent ion exchange column has been developed since the inception of the Higgins contactor, but this concept was not used in the present work.⁸ In the Higgins contactor developed in the 1950s and 1960s,⁹⁻¹⁴ the ion exchange resin was continuously recycled by using a regenerating section after the loading section. In the study

described here, the loaded resin is the contactor product. Thus, as in the laboratory-scale Higgins contactor,⁵ no regenerating section is involved.

2.1 Basis for Selection of Size

The process equipment used to supply ADUN for the batch loader was used in the present work.^{4,6} The batch loader demonstrated a capacity of 25 kg of uranium per day (suggested size for a commercial HTGR recycle facility; the capacity of a fresh fuel facility would be 100 kg/day).⁴⁻⁶ Although the nitrate extraction--amine regeneration could handle a higher capacity, the scale-up of the 1-in.-ID Higgins contactor was purposely kept in this range so that the capacity of the engineering-scale contactor could be fully tested. Consequently, the 2-in.-ID Higgins contactor described in detail in the following section was selected as the design for the engineering-scale tests of continuous uranium loading of weak-acid resin.

2.2 Description of Equipment

A schematic of the 2-in.-ID Higgins contactor constructed for this project is shown in Fig. 1. This figure illustrates the connections between the liquid ports and the ADUN system. Figure 2 is a photograph of the Higgins contactor. The overall height of the column is approximately 10 ft; the lengths of the loading, wash, and pulse sections are 10.2, 2.2, and 2 ft, respectively. (Note that there is a small "dead" space, 0.5 ft long, between the pulse section and the effluent exit.) The resin feed tank consists of a 6-in. to 2-in. Pyrex reducing union below a 1-ft section of 6-in. Pyrex glass pipe.

Fig. 1. Schematic of 2-in.-ID Higgins contactor.

Fig. 2. Photograph of 2-in.-ID Higgins contactor in the process of loading uranium on Amberlite IRC-72 resin.

The basic column structure is 2-in. Pyrex glass pipe. The U-turn is made with two sweep elbows. The process stream inlet and outlets are made via Plexiglas ports. The interface between the process streams and the resin is maintained by using a screen on the process line (except for the pulse line, which does not protrude into the column). These process lines, which are made from either 3/8- or 1/4-in. stainless steel tubing, terminate after extending across the column diameter and are sealed on the end. The portion of the tube that is exposed to the resin bed is bored with holes and covered with wire mesh to allow free solution flow across the tube walls while screening out resin.

The resin valves (2-in. ball valves) are interfaced with the glass pipe by means of 2-5/8-in.-long sections of 2-in. stainless steel pipe. Flanges designed to work in conjunction with 2-in. glass pipe flanges were welded onto one end of the stainless steel pipe. The resin valves are rotated by means of pneumatic rotary actuators with spring return. The resin valve between the resin feed tank and the pulse section operates out of phase with the other two resin valves (see Fig. 3). "Normally closed" solenoid valves provide "on-off" control of the process streams. The circuit used to control these valves was the same as that used on the 1-in.-ID laboratory-scale Higgins contactor.⁵ The schematic in Fig. 4 demonstrates how the control circuit works for one cycle. Note that all of the valves require power in order to remain open (except the resin valve between the resin feed tank and the pulse section); therefore, when the column is not operating, or in case of power failure, the active portion of the column is isolated (i.e., all valves are closed except the feed tank resin valve). (In case of compressed air failure, the spring return actuators ensured that the resin valves

Fig. 3. Schematic drawing of continuous column with valves in load and pulse positions.

Fig. 4. Schematic demonstrating control circuit operation.

would be in the "load" mode. The solenoids could still function, which means that the pulse liquid would be filling and overflowing the resin feed tank.)

After the column had been fabricated, it was leak-tested by operating with process streams of water, but no resin. Several very small leaks around pipe threads escaped detection until uranium was being used. For this reason, pipe threads should be avoided in future designs, if possible.

3. PROCEDURES AND ACCESSORY EQUIPMENT

3.1 Resin Conversion to the H^+ -form

The method for preparing resin feed, shown in Fig. 5, is an integral part of the HTGR reference fuel process. The engineering-scale Higgins contactor was first connected with the existing batch equipment used to convert Na^+ -form resin to the H^+ -form (see Fig. 6) and tested in several runs to determine the feasibility of continuously performing this conversion on Amberlite IRC-72. This operation demonstrated the feasibility of using a Higgins contactor to continuously convert commercial Na^+ -form resin to the H^+ -form, and provided the operators an opportunity to gain valuable experience with the contactor under simpler operating conditions than those used for uranium loading.

Two differences between continuous acid conversion and continuous uranium loading should be noted: (1) as the resin is converted from the Na^+ -form to the H^+ -form, it undergoes a large volume decrease (50%); and (2) continuous acid conversion involves single-pass operation of the feed, whereas continuous uranium loading involves continuous

Fig. 5. Process for preparing resin feed.

Fig. 6. Batch resin conditioning system flowsheet.

recycle of the effluent. Neither of these differences seriously affected the two goals stated above, although the volume change might have masked some problems evident in uranium loading.

Excellent acid conversion of Amberlite IRC-72 was obtained using the Higgins contactor (0.5 to 5 ppm of sodium in the final product for about 10% excess HNO_3). The contactor demonstrated 79 to 99% utilization of the feed acid without any attempts to optimize acid utilization. An automatic control technique presented in Sect. 4.1.1 would provide stable, reproducible conversion of resin on the Higgins contactor with efficient utilization of the feed acid.

3.2 Uranium Loading

After its satisfactory operation in acid conversion, the Higgins contactor was used in a series of experiments to determine its application to continuous uranium loading. This was done by connecting the Higgins contactor to the existing nitrate extraction--amine regeneration system referred to previously⁶ and shown schematically in Figs. 7 and 8 (Fig. 8 illustrates the batch contactor originally used). Figure 9 shows how the Higgins contactor was substituted for the batch contactor. The results obtained in these experiments confirmed that the Higgins contactor can successfully load uranium on acid-form resin on a continuous basis for periods up to 20 hr. Remote operation is feasible with the instrumentation indicated in Fig. 9.

3.3 Mass Transfer Considerations for Higgins Contactor

While Higgins columns are characterized by alternating periods of solution and resin flow, they exhibit mass transfer behavior equivalent

"ADUN" INDICATES ACID-DEFICIENT URANYL NITRATE ($\text{NO}_3/\text{U} < 2$, MOLES/MOLE)

Fig. 7. Flowsheet for loading uranium on resin via nitrate extraction.

ORNL DWG 76-926 R1

Fig. 8. Process flows of uranium and ion exchange resin during batch loading of resin.

ORNL DWG 77-666

Fig. 9. Flowsheet for continuous loading of uranium on ion exchange resin from acid-deficient uranyl nitrate prepared by nitric acid extraction.

to that for true countercurrent flow at the average flow rate for each phase. The net exchange reactions for feed resin conversion and uranium loading, respectively, are (where R indicates the resin equivalent to one exchange site):

In the case of feed conversion, the H^+ is supplied as nitric acid solution and the solution product is NaNO_3 . For uranium loading, the UO_2^{2+} is supplied as ADUN and the product solution is less acid deficient or contains free HNO_3 .

Calculations for mass transfer require the following information:

- (1) flow rates and compositions for material balance calculations,
- (2) the equilibrium relationship between the resin and solution, and
- (3) the kinetic relationships for mass transfer. The kinetic relationships will be very complex since they include the hydrolysis reactions of uranium in solution, the reactions of the resin, and mass transfer in the bulk liquid, the solution, the resin pores, and the solid resin.

Since the void volume between resin spheres is 40% and additional porosity exists inside the resin sphere, the most convenient units are bulk volume flow rates and concentrations in equivalents per liter of bulk volume. The uranium concentration can be expressed as a fraction of the total resin exchange capacity, that is, the fraction of the resin that is in the UO_2^{2+} -form. The typical total cation exchange capacities measured for our resins have been:

<u>Resin</u>	<u>Form</u>	<u>C_y (equiv/liter)</u>
Amberlite IRC-72	H ⁺ , UO ₂ ²⁺	3.2
	Na ⁺	1.6
Duolite C-464	H ⁺ , UO ₂ ²⁺	2.6
	Na ⁺	1.7

The equilibrium limitations for the two conversions are quite different. The conversion of Na⁺-form resin to the H⁺-form [Eq. (1)] is quite favorable so that only a small excess of acid is required for completion. Thus the Higgins column should provide good conversions using only 105 or 110% of the stoichiometric amount of HNO₃.

The equilibrium data of Shaffer et al.¹⁵ for Eq. (2) were plotted in a form convenient for countercurrent mass transfer calculations (Fig. 10). For the units used in Fig. 10, the operating line for the Higgins column is straight with a slope equal to the equivalents of NO₃⁻ divided by the equivalents of resin. This slope results from a uranium material balance as follows:

$$VC_y dy = LC_x dx \quad , \quad (3)$$

where

V = resin flow rate, liters/min,

y = uranium concentration of the resin, equiv U/equiv resin,

C_y = cation exchange capacity of the resin, equiv/liter,

L = solution feed rate, liters/min,

C_x = nitrate concentration, equiv/liter,

x = solution U/NO₃⁻ ratio, equiv UO₂²⁺/equiv NO₃⁻.

ORNL DWG 77-391

Fig. 10. Equilibrium loading of uranium vs ADUN U/NO_3^- ratio (Amberlite IRC-72 or Duolite C-464 at 30°C or 40°C).

Rearranging Eq. (3) yields:

$$\frac{dy}{dx} = \frac{LC_x}{VC_y} \quad (4)$$

The products, LC_x and VC_y , are the flow rates of nitrate and resin equivalents per unit time. These do not change as a result of uranium loading, water wash or pulse flow rates, or resin volume changes. Therefore, the ratio dy/dx is a constant throughout the steady-state operation of a Higgins column, and an equilibrium diagram in units of y and x results in a straight operating line. Other units for the resin and solution concentrations would not give a straight operating line.

The feed solution must have a favorable (high) U/NO_3^- ratio in order to provide a high loading of uranium on the resin. Also, an excess of solution is needed to give a large value of LC_x or a large dy/dx . In terms of conventional transfer unit or theoretical stage models, the operating line and the equilibrium line will pinch at the resin exit or solution inlet end of the column. For the typical operating line shown in Fig. 10 (slope = 3.5 equiv/equiv), about 50% of the uranium loading occurs in one stage at the resin inlet; over 90% occurs in two stages. Most of the Higgins column is filled by resin which is more than 90% loaded with uranium. If the LC_x/VC_y ratio is decreased, the point at which the pinch occurs will change from the resin exit to a point where the resin is about 20% loaded and the uranium loading of the exit resin will decrease. If the feed solution is allowed to become less acid deficient (that is, x or the U/NO_3^- ratio decreases), the uranium loading will also decrease as a result of the less favorable

equilibrium. The resin loading system must be controlled to keep both the acid deficiency or the feed solution value of x and the ratio of LC_x/VC_y at favorable values.

Neither the Na^+ - to H^+ -form conversion nor the uranium loading requires more than a few theoretical stages (each was accomplished in single-stage batch contactors with inefficient use of solutions). The loading of the last few percent of the uranium proceeds slowly as a result of some combination of slow mass transfer to the center of the resin beads and slow reaction of the less available exchange sites. Therefore, the HTU or HETS values as defined for conventional counter-current mass transfer may increase greatly as uranium loading nears completion. For this reason, the Higgins column may have fewer theoretical stages or transfer units for uranium loading than the same column would have for some other ion exchange process.

4. RESULTS

4.1 Process Control

The two applications of the contactor — acid conversion and uranium loading — offer a contrast in process control. The conversion of the resin to acid is highly favorable, requiring only a slight excess of acid for complete conversion. Therefore, the Higgins contactor was operated with only a single pass of the acid feed and the pH of the effluent was used as the control parameter. In contrast, the uranium loading application was not a single-pass operation. The ADUN surge tank was the central collection and supply point for the resin loading cycle and the extraction-regeneration cycle. (Note that the contactor

effluent was returned to this tank via evaporation.) Thus, the contactor was operated with set times instead of manually. The process control effort was directed at maintaining a feed stream suitable for effective resin loading. (The feed stream determines the final equilibrium loading of the resin, that is, the uranium content of the product.)

4.1.1 Resin conversion to the H^+ -form

In theory, either the resin flow rate or the acid flow rate could be varied to control the conversion operation. The concentrations of the feed acid and the feed resin are constants. The pH of the effluent solution is easy to measure, while the percent conversion of the resin is not. Since the load time directly affects the resin flow rate, control of the effluent pH by varying the load time is very logical and effective.

The traditional Higgins contactor operation is to set the solution flow rates and the resin flow rate. The resin flow rate is controlled by varying the load time and the pulse time. The load time controls the number of pulses per hour, and the pulse time controls the size of each pulse (see Fig. 11 for the effect on effluent pH of varying these two parameters plus the acid feed rate). The product of these two yields the resin feed rate. The timers required frequent adjustments to maintain the effluent pH within desired limits; with experience, however, the correct timer settings could be determined. Since the effluent pH was the variable of concern, switching to manual control of load time based on observation of the effluent pH was a logical alternative. This alternative method required the operator to observe

Fig. 11. Record of effluent pH for acid conversion of Amberlite resin.

the effluent pH and to initiate a pulse when the pH reached a certain value (typically 1.0). This manual feedback control proved to be effective and provided consistent and reproducible operation. The traditional operation requires damping action to be applied by the operator. The amount of damping would be highly dependent on the skill and experience of the operator. Although testing of the second method was limited to the use of manual controls, results showed that only an automatic response is required. An automatic feedback control system would duplicate the manual feedback control and would maintain good operating conditions unattended. For example, if the acid throughput suddenly decreased or increased, the original system would not respond automatically; the proposed system, on the other hand, would automatically decrease or increase the resin throughput to match the new acid throughput. The efficiency of acid utilization was 79 to 99% without any attempts at optimization. The proposed procedure for controlling the effluent pH can be easily used to achieve a high efficiency.

4.1.2 Uranium loading

The fact that the resin exit of the Higgins contactor operates at pinch conditions (for high loading) was clearly pointed out in the discussion on mass transfer considerations (see Sect. 3.3). This, along with the fact that contact time must exceed a minimum, changes the operating scheme for uranium loading compared with conversion to U^{+4} -form.

The work relative to the batch loading of uranium indicates that a 2-hr contact time is required for complete conversion.⁴⁻⁶ This was

verified at the end of one Higgins contactor run from the observation that loading was incomplete after 80 min but complete after 120 min.

Since complete conversion of the resin and maximum utilization of the uranium are the main objectives, the uranium effluent was continuously recycled to the ADUN surge tanks. Complete conversion of the resin depended on keeping the uranium-to-nitrate concentration ratio of the inlet feed within a certain range ($x \sim 1.25$ equiv U per equiv NO_3^- , see Fig. 10) and establishing a satisfactory slope for the operating line. The former fixes the equilibrium conversion of the resin exiting the column, while the latter ensures that this conversion is approached (assuming enough contact time is allowed).

The extent of resin loading is not easily and quickly determined. Therefore, the process variables of the feed ADUN were selected as the control variables. These variables are not directly related to the resin loading because of the intermediate steps of nitrate extraction and evaporation; however, the control scheme can be simplified by holding certain operations constant and affixing one process variable to each important control parameter.

The overall rate of loading was controlled by setting the caustic rate for regenerating amine (see Fig. 9) and the ADUN feed rate to the Higgins contactor at fixed values based on the desired loading rate. The uranium-to-nitrate concentration ratio of the ADUN feed was maintained by controlling the pH of the ADUN, the uranium concentration of the ADUN feed was maintained by controlling the ADUN density, and the volume of the ADUN was maintained by controlling the water evaporation rate.

The uranium depletion of the ADUN by resin take-up was balanced using a high-density solution of uranyl nitrate hexahydrate (UNH). The uranium concentration of the ADUN is proportional to the density of the ADUN according to the following relationship:⁶

$$[u] = 3.10 [\rho(\text{ADUN}) - \rho(\text{H}_2\text{O})], \quad (5)$$

where

$[u]$ = uranium molarity,

$\rho(\text{ADUN})$ = ADUN density, g/cc,

$\rho(\text{H}_2\text{O})$ = density of water at the same temperature, g/cc.

Thus the uranium concentration was controlled by observing the density of the ADUN and varying the UNH flow rate accordingly.

Since the uranium concentration was controlled independently, the U/NO_3^- ratio was determined by the acid concentration. Any change of acid concentration results from an imbalance of acid input from resin loading to acid takeout from nitrate extraction. Since the organic extraction rate is relatively constant, the resin loading rate (i.e., the resin flow rate) determines the pH of the ADUN. Thus, the U/NO_3^- ratio was controlled by observing the pH of the ADUN and varying the resin flow rate accordingly.

The evaporator in the ADUN system serves to prevent a continuous net addition of ADUN volume. (Note from Fig. 9 that the ADUN effluent, not the ADUN feed, passes through the evaporator. The high acid deficiency of the ADUN feed causes precipitation and plugging of the evaporator. The less acid-deficient effluent does not experience this problem.)

The ADUN volume was controlled simply by observing the volume and increasing or decreasing the evaporation rate (via increasing or decreasing the steam pressure) accordingly.

Table 1 summarizes the control scheme outlined above, and Fig. 12 demonstrates the variability of the selected parameters. Operation of the Higgins contactor using this control method was successful. The most significant disadvantage of the method was the slow response times. For example, since resin has approximately a 2-hr residence time in the loading section, changing the resin feed rate as a means of controlling the pH will result in a highly damped response. Nevertheless, a skilled operator can maintain satisfactory operating conditions. The most severe circumstances that could result from the poor judgment of an operator would be incomplete loading of the resin or the precipitation of uranium oxide. The first error could be remedied by further loading the affected resin and the second by decreasing the system pH to dissolve any uranium oxide.

Table 1. Control scheme for continuous resin loading

Parameter	Control method	Measurement
H ₂ O	Evaporator	Volume
U	U addition	Density
U/NO ₃ ⁻	Resin flow rate	pH

The process measurements — pH, density, and volume — presented no difficulty and could be made consistently and accurately. In-line pH

Fig. 12. Ac-d-deficient uranyl nitrate concentrations and volumes for a continuous (Higgins column) loading run.

meters and an in-line density meter provided continuous records and quickly responded to any process changes. All of these measurements are amenable to remote operation. The in-line pH meters tend to drift and require daily recalibration; thus far, however, this difficulty has not been encountered with the in-line density meter.

4.2 Solution Flow Results

Three problems with the solution flows were experienced during contactor operation: (1) seepage of resin into the pulse flow line, (2) failure of the three-way solenoid valves, and (3) clogging of the effluent exit.

The seepage problem occurred because the pulse flow line had no screen interface to keep resin out. This problem proved relatively minor since column operation was never hindered and the line was easily cleared by a pulse of water. A simple gooseneck between the column inlet and the solenoid valve would prevent such seepage.

Originally, the two inlet streams (feed and wash) passed through a three-way solenoid valve before entering the contactor. These valves allowed flow into the column during the loading period and recycle during the pulse sequence. Hence, the pumps did not have to function without flow during any portion of the Higgins cycle. Unfortunately, the three-way solenoid valves failed during our uranium loading tests. This failure could have been prevented and a lower pressure drop provided by replacing each three-way valve with two two-way valves. Operation was not hindered, however, by simply eliminating the recycle streams and replacing each three-way valve with a single two-way valve. (This is the setup shown in Fig. 1.)

Failure of the three-way valves can be attributed partially to the control system operation. As mentioned previously, the system without power is designed to isolate the contactor; consequently, the solenoid valves are with power in the load mode (which is essentially 100% of the cycle time). Although the two-way solenoid valves performed satisfactorily in the engineering-scale tests, long-term operation may reveal serious problems related to solenoid failure. Redesign of the contactor using "normally-open" valves may be necessary.

The original effluent exit tended to plug with resin, causing a steadily increasing resistance to liquid flow in the load mode. Eventually, the exit became completely blocked and liquid flow ceased. Inspection showed that resin particles were being trapped between the screen and tube of the outlet, and these particles served to trap additional particles (i.e., tended to provide a filtering action). The presence of fines aggravates (and maybe causes) the situation. Johnson well screens^{*} are used in Higgins contactors to alleviate this problem.¹⁶ A new outlet, fabricated from a Johnson well screen, performed satisfactorily without plugging.

^{*}A Johnson well screen consists of triangular wire wrapped around and welded to support rods. This construction allows a very open structure for free flow but stops particles on the outside surface where they are easily scraped off by the resin pulsing. With this arrangement, particles could never get trapped between two surfaces, as they were with the former outlet.

4.3 Resin Flow Results

In Higgins column operations, the pattern followed for pulse liquid flow typically consists of an initial surge to a high rate for up to 2 sec followed by a leveling off to a fairly uniform rate. The resin displays a lag similar to Bingham plastic flow and then assumes a steady, but sometimes jerky, movement. The downleg flow is typical of stick slip flow, whereas the upleg flow is typical of aerated flow.

The resin typically enters the loading section as a solid mass. This mass is broken up by the column obstructions and the resin movement, although it is not unusual for a few of the chunks formed to survive to the resin exit. Sometimes these chunks create problems by restricting the resin exit. Understandably, any obstruction, diameter change, or direction change increases the resistance to resin movement. No difficulties were encountered with acid conversion; however, problems were experienced in uranium loading with resin movement and jamming of the resin valves.

The retarded resin movement caused high fluidization of the upleg. An attempt was made to control the fluidization by reducing the column diameter from 2 in. to 1 in. at the U-bend. However, it was unsuccessful since the resin flow was reduced to practically zero, even after the pulse pressure was increased from ~20 psig to ~40 psig. Consequently, several modifications of the column and resin exit line were made in order to reduce the overall resistance to resin movement. The original 2-in.-ID bottom was reinstalled with a new resin drain.

The original drain in the U-bend called for a tube extending practically across the column diameter and exiting the column at the top instead of the bottom. Such a design prevented resin from clogging the drain line and allowed almost as much drainage, via vacuum, as a downward design. Since fluidization seemed to originate from the resin drain, this obstruction was removed. The new design simply called for a downward drain, eliminating the obstructing tube (a focal point for voids) in the highly critical horizontal section of the U-bend. The column was further streamlined by recutting the column gaskets and refabricating the resin exit. The gaskets had originally extended slightly into the column interior, making slight restrictions at several points in the column.

The original resin exit line was a 1/2-in.-diam, plastic tube 15 to 20 ft long. Resin did not move as freely as liquid in this line, and sometimes the liquid would drain from the line, leaving resin in a compact mass. Such a compaction prevented both liquid and resin flow. A 1-in.-diam stainless steel tube 5 to 6 ft long for the resin exit line combined with a larger valve on the resin collection tank allowed free resin drainage from the exit line and, in addition, reduced the resistance to resin movement.

After these changes had been made, resin moved through the column at the desired rate without high fluidization.

In summary, the only difficulties encountered were those related to the overall resistance to resin movement in the column. The greater the resistance to resin movement, the higher the fluidization in the upleg and the lower the rate of resin movement (until the point was reached

where no resin movement was occurring). Further reductions in resistance to resin movement could be accomplished by elimination of any column obstructions and by redesign of the resin exit. The obstructions in the present column are the wash distributor, feed distributor, pH check flow exit, and effluent exit described in the equipment section. These internal interfaces to the resin bed could be redesigned so that obstruction to the resin movement is eliminated. For example, a ring design (i.e., a tube encircling the inner circumference) rather than the present diagonal design would not interfere with resin movement. The column exit could be redesigned so that the resin is not pulsed through a restriction (i.e., the diameter of the resin exit is the same as that of the column, and resin is allowed to overflow into a collection tank).

Finally, during the uranium loading of Amberlite resin, resin lodging beneath the ball of the resin valves sometimes prevented the spring (the pneumatic rotation never jammed) from closing the valve. Manual assistance always completed rotation. Very small particles in the Amberlite resin, constituting only a small volume fraction of the screened resin, are probably the culprits in this case. More efficient screening in a resin feed processing facility⁷ or the use of a resin that is relatively free of these fines (e.g., Duolite) is a possible solution. A better solution would be to change to plug or butterfly valves. "Squeegee" butterfly valves work very well in Higgins contactors.¹⁶ (The leading edge of the disk of "Squeegee" butterfly valves is sharp, providing a sweeping, or "Squeegee," action on the elastomer seal.) More satisfactory operation is also obtained with

actuators that operate pneumatically both ways.¹⁶ The spring return can be retained as a fail-safe feature.

Even though resin was being caught in the mechanical action of the resin valves, shape separation of loaded resin resulted in less than 1% rejects. This indicates that no significant attrition of the resin occurred on passage through the Higgins contactor.

In addition to the resin being pushed through the column, resin movement also includes the gravity drainage of resin from the feed tank to the pulse section. This gravity flow provides resin at the bulk rate of approximately 20 liters/hr, which is more than adequate compared with the flow rates through the column (< 5.0 liters/hr; see the Appendix). (About 2 min is required to completely fill the pulse section.) Occasionally, bridging of the resin in the Pyrex reducing union did occur, preventing resin from draining into the pulse section. This problem was handled easily by manual probing; however, redesign of this reducing union will be necessary for a remote system. A reducing union with a more gradual change in diameter than the existing reducing union should be sufficient to eliminate the difficulty. If not, provision can be made for injection of water into the union to break up or prevent bridging.

4.4 Column Capacity

The important points to be discussed in this section are: (1) the maximum solution and resin flow rates that can be obtained for the

2-in.-ID Higgins contactor while maintaining complete loading* of uranium on the resin, and (2) the effect of scale-up to a larger column on these maximum flow rates.

4.4.1 Solution flow

As was pointed out in Sect. 3.3, the minimum solution flow rate for complete conversion of the resin was determined by the mass balance. The maximum solution flow rate was determined by the column flow resistance (i.e., the pressure drop through the column) and the existing equipment (i.e., the delivery pressure of the solution). Since the loading portion of the Higgins cycle operates as a packed bed, the pressure drop through the Higgins contactor was compared to the traditional pressure drop correlation for packed beds given below:¹⁷

$$\frac{\Delta p}{L} = \frac{2f_m G^2 (1 - \epsilon)^{3-n}}{D_p g_c \rho \phi_s^{3-n} \epsilon^3}, \quad (6)$$

where

Δp = pressure drop, lb_f/ft^2 ,

L = length, ft,

f_m = modified friction factor,

G = mass rate per unit area, $\text{lb}_m/\text{sec} \cdot \text{ft}^2$,

ϵ = void volume,

D_p = particle size, ft,

* Approximately 95% conversion can be considered complete in the present system with operation at pinch conditions.

$$g_c = 32.17 \frac{\text{lb}_m \cdot \text{ft}}{\text{lb}_f \cdot \text{sec}^2},$$

$$\rho = \text{liquid density, lb}_m/\text{ft}^3,$$

$$\phi_s = \text{shape factor, (ft}^2\text{) sphere/(ft}^2\text{) actual.}$$

Empirical correlations for the two quantities f_m and n are given by Leva¹⁷ as functions of the modified Reynolds number, N'_{Re} , defined as follows:

$$N'_{Re} = \frac{D_p G}{\mu}, \quad (7)$$

where

$$\mu = \text{liquid viscosity, lb}_m/\text{ft} \cdot \text{sec}.$$

For

$$N'_{Re} \leq 13, \quad (8)$$

the correlations can be represented by the following simple equations:

$$f_m = 100/N'_{Re}, \quad (9)$$

$$n = 1.0. \quad (10)$$

Substituting into Eq. (6) and rearranging,

$$\frac{\Delta p}{L} = \frac{200\mu(1-\epsilon)^2}{g_c \phi_s^2 \rho \epsilon^3} \frac{G}{D_p^2} \quad (11)$$

Note that the pressure drop for a given liquid is directly proportional to the mass flow rate and inversely proportional to the particle size squared. Upon scaling up, one does not scale up the mass flow rate per

unit area, G , but rather the mass flow rate itself to match the increased area. Therefore, Eq. (11) is useful for estimating the effects of different flow rates and particle sizes on the pressure drop both in the 2-in.-ID column and in scaled-up columns. In addition, it tells us that, if a conversion rate can be achieved in the 2-in.-ID column, the same rate per unit area can be achieved in a larger column with no increase in pressure drop.

The pressure drop through the washing and loading sections of the contactor was measured for varying flow rates of water. The column was filled with uranium-loaded, 625- to 765- μ -diam Amberlite IRC-72. For these measurements, Eq. (11) reduced to:

$$\Delta p'/L = 1.04G, \quad (12)$$

where

$\Delta p'$ = pressure drop, ft H_2O .

A regression of the wash flow data resulted in the following equation (95% confidence limits are given):

$$\Delta p'/L = (1.35 \pm 0.07) G^{1.11 \pm 0.04} \quad (13)$$

The pressure drop data for the wash flow experiments, as well as Eqs. (12) and (13), are plotted in Fig. 13. Pressure drop data taken for ADUN flow alone through the loading section are also plotted in this figure. Although the agreement between Leva's correlation and the present empirical results is not exact, the agreement demonstrated in Fig. 12 supports the conclusions about scale-up drawn from Leva's

Fig. 13. Pressure drop in 2-in.-ID Higgins contactor as a function of flow rate.

correlation. (It should be noted that the empirical results are for varying flow rates in a single cross-sectional area, not varying cross-sectional areas.)

The recommended pressure limitation for 2-in.-ID glass pipe is 55 psig. If we assume only a 10.2-ft length (> 90% of the pressure drop is in the loading section) and use 55 psig as the limiting pressure drop (at present, nothing is known about the pressure limit of the resin), the pressure drop per unit length is calculated to be 12.4 ft H₂O/ft. From Eq. (13), the maximum allowable solution flow for the 2-in.-ID Higgins contactor is 7.37 lb_m/sec·ft², or approximately 4 liters/min.

Criticality limits scale-up of the Higgins contactor to a 4-in. diameter. The recommended pressure limit for 4-in.-ID glass pipe is 35 psig, which converts to 7.92 ft H₂O/ft (based on a 10.2-ft length). From Eq. (13), the estimated maximum solution flow for a 4-in.-ID Higgins contactor is 4.92 lb_m/sec·ft², or approximately 11 liters/min. If the column is lengthened, the allowable pressure drop per unit length decreases proportionately. This affects the corresponding maximum flow rate according to the following relationship:

$$L_2/L_1 = (G_1/G_2)^{1.11} . \quad (14)$$

For example, doubling the column length means that the maximum flow rate is only 53.5% of the original value (i.e., a 4-in.-ID, 20.4-ft-long contactor would have a maximum solution flow of 5.9 liters/min compared with 11 liters/min for a 4-in.-ID, 10.2-ft-long contactor.

4.4.2 Uranium loading

The results for the Higgins contactor runs in which uranium was loaded on cation exchange resin are presented in the Appendix. In general, a slightly higher loading was achieved with the Higgins contactor than for batch loading. (Note that a higher loading was achieved with the Amberlite resin than with the Duolite resin, verifying the same observation for batch loading.) A high loading was achieved, 1.4 kg of uranium per hour, which easily exceeds the desired rate of 25 kg of uranium per day (size of a commercial recycle facility) for a 2-in.-ID contactor. The desired loading was not obtained in some of the runs at flow rates that resulted in operation close to pinch conditions on a low part of the equilibrium curve.

4.4.3 Scale-up

Criticality limits scale-up of the column diameter to 4 in. Increasing the diameter to this size would quadruple the loading rate demonstrated on the 2-in.-ID contactor and easily meet the capacity requirement of 100 kg of uranium per day for a commercial fresh fuel operation.⁴⁻⁶ Increasing the length of the column is limited only by the pressure drop limitations. A tendency for the resin particles to adhere to each other, and thus form a solid mass, has been noted; and the operating pressure probably affects this behavior. For this reason, the pressure limitations cannot be conclusively evaluated. However, the analysis of the diameter scale-up indicates that the desired capacity can be achieved without increasing the length. A summary of the column capacity is given in Table 3 in the Appendix.

4.5 Startup and Shutdown Procedures

Startup of the Higgins contactor presented no difficulty. Two procedures gave successful results for both acid conversion and uranium loading. In one of these, the column is filled with resin (to the feed point) and operated as a packed bed until all the resin in the loading section has reacted (control is maintained by observing the process variables of the effluent stream); then normal operation is started. The other procedure consists in filling the loading section with feed solution and then introducing the resin by pulsing or by gravity feed. The resin should be introduced in short bursts in order to prevent pushing uranyl nitrate solution (for uranium loading) or acid solution (for acid conversion) out the resin exit.

Because acid conversion is a single-pass operation, it is more amenable to the first procedure. In the second procedure, the acid feed must be allowed to stagnate in the column while resin is being introduced. Care must be exercised to keep from pushing acid out the resin exit. Once enough resin has reacted with acid in the downleg, acid and wash flow, as well as resin pulsing, can be slowly started and then allowed to build up to normal operation while maintaining control of the effluent pH. In contrast, the first procedure allows normal acid and wash flow rates until the proper effluent pH is reached with minimum operator effort (since the resin shrinks during acid conversion, the resin must be replenished during startup); then, normal operation is started.

In contrast, uranium loading is not a single-pass operation and the second procedure is more applicable. With no resin in the column, the solution and resin flows are started as would be the case during normal operation. The column gradually fills with resin via pulsing action. Startup is different from normal operation in that the first resin through the column encounters the maximum amount of uranium passing through the loading section, and each succeeding segment throughout the startup section encounters slightly less, until "steady-state" normal operation is achieved. In this case, the resin rate, not the effluent pH, controls the startup.

Shutdown presented no problems; simply turning off the column flows and the control system was sufficient. Sometimes the column was flushed with wash water after shutdown, while in other instances, it was left untreated.

Removing the resin from the column was another matter. In this case, care was taken to ensure that all of the resin was loaded. For uranium loading, feed was flushed through the column until the pH of the effluent approached that of the feed (if wash flow was used, the two pHs were slightly different). In Run H9, this final loading was observed to require between 80 and 120 min. After ensuring complete loading, the column was flushed with wash water. Most of the column resin could be pulsed out (i.e., pushed out with liquid in one long pulse). Once the resin plug reached the upleg, resin was lost from the face of the plug by gravity. Any obstructions or restrictions helped break up the plug, resulting in more resin being left in the

bottom. Up to this point, only the existing solenoid valves and pneumatically controlled ball valves were needed to remove most of the resin from the Higgins contactor; however, some resin remaining at the bottom of the column "U" had to be removed by the resin drain (see Fig. 1). The resin drain would have to be redesigned if this part of the operation were to be handled remotely. One possible solution would be to install a solenoid valve on this line. The resin could then fall into a small container which could be pressurized to pneumatically transport the resin to storage (or elsewhere).

4.6 Resin Washing, Resin Drying, and Product Quality

4.6.1 Washing

The wash flow rate of 100 to 200 cc/min proved sufficient for our purposes in these experiments with the Higgins column. The liquid in the resin storage contained only a small quantity (10 to 150 ppm) of uranium, probably from solution retained in the porous structure of the resin. Since the volume of the wash section was greater than 1300 cc, the stated wash flow rate results in a space time of 2.6 to 5.2 min (assuming a void fraction of 0.40). Since the load time was never greater than 5 min, the entire wash section may not have been purged between pulses, even with plug flow; however, the amount of resin per pulse varied from 175 to 325 cc, indicating that between 4 and 7.5 pulses were required for the resin to clear the washing section. With a load time of 5 min and a void fraction of 0.40, the resin was washed 15 to 107 space times before exiting. Larger volumes per pulse may present a problem with regard to sufficient washing of the resin or

to pulsing of the feed solution out the exit. During the pulsing operation required for uranium loading, the slippage* was 1.4 to 1.7.

4.6.2 Drying

The resin loaded on the Higgins contactor was dried by means of the microwave system established for the batch loader.⁶ The only requirement was that the resin be transported pneumatically from the resin storage tank to the microwave oven. A procedure was developed whereby the resin was dried periodically during long periods of operation of the contactor. After approximately 11 liters (dictated by the size of existing equipment) of loaded resin had been collected, the contactor was pulsed twice in succession and the pulse flow was turned off manually while the loaded resin was being transferred to the microwave oven. (Transfer required longer than 5 min, which was the maximum loading time used, i.e., one pulse would have occurred during transfer; completing the transfer before the end of a load cycle would eliminate the need for a double pulse and turning off the pulse flow manually.) After transfer was complete, normal contactor operation was resumed, and the resin was dried according to established procedure.

* Defined as the ratio of total volume pulsed to the bulk volume of the resin pulsed. It should be noted that bulk resin includes 40% voids as well as internal pores.

4.6.3 Quality

The quality of the resin loaded with uranium on the 2-in.-ID Higgins contactor was generally good. A high loading of uranium could be obtained with routine operating procedures. Shape separation of some of the product gave $\sim 0.3\%$ rejects (i.e., nonspherical particles), indicating that no significant attrition of the resin had occurred on passing through the contactor. Visual inspection and subsequent carbonization did not reveal any problems relative to cracking or weakening of the spherical shape.

5. ADDITIONAL RESULTS REQUIRED

Continuous loading of uranium on an engineering scale has been successfully demonstrated with a 2-in.-ID Higgins contactor. However, the following are needed as a prerequisite to designing Higgins contactors for remote operation with ^{233}U or improving the long-term dependability of these contactors:

1. demonstration of dependable resin valve performance under remote operating conditions;
2. establishment of the extent of materials reliability, particularly polymeric materials (used in seals and gaskets), under irradiation;
3. demonstration of dependable lifetimes of solenoid valves, particularly "normally closed" solenoid valves;
4. data concerning the mechanical and physical properties of resin under increased pressure; and

5. a cold engineering demonstration of loading 100 kg of uranium per day continuously.

6. CONCLUSIONS

A 2-in.-ID Higgins contactor was built to demonstrate continuous loading of weak-acid ion exchange resin with uranium on an engineering scale in conjunction with existing nitrate extraction process equipment. Process control procedures were demonstrated using pH, density, and volume measurements of the ADUN. These measurements, which were easily obtained and found to be reliable, could be obtained remotely. In addition, several problems involving the mechanical operation of the contactor were handled. Some possible improvements were noted, and these are enumerated in Sect. 5. The capacity of the column was tested within the limitations of our equipment, and the objective of 25 kg of uranium per day (size of a commercial recycle facility) was easily met. Simple startup and shutdown procedures were demonstrated, suggesting that the handling of different campaigns (i.e., feedstocks of different isotopic compositions for different customers) would present no difficulty. Established procedures were used in washing and drying the resin. Shape separation analysis indicated negligible degradation of the resin passing through the Higgins contactor.

The conclusions drawn from this work can be summarized as follows:

1. Adequate process control has been demonstrated for continuous loading of cation exchange resin with uranium.
2. Remote operation appears to be feasible with the existing instrumentation.

3. The existing equipment can supply an acceptable resin product at the rate of 1.4 kg of uranium per hour.
4. Scale-up to 100 kg of uranium per day (size of a fresh fuel facility) can be achieved by using a 4-in.-ID Higgins contactor. The batch loader demonstrated in a previous study has a maximum capacity of 25 kg/day.
5. The Higgins contactor efficiently converts Na^+ -form resin to the H^+ -form on a continuous basis.
6. The dependability of the resin valves and some other requirements for remote or long-term operation remain to be demonstrated.

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9. APPENDIX

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Table 2. Results obtained in Higgins contactor runs

Run ^a	Resin flow rate (ℓ/hr) ^b	Extent of U loading [wt % (dry)]	Rate of U loading (kg U/hr)	$\frac{LC_x^c}{VC_y}$ $\left(\frac{\text{equiv NO}_3^-}{\text{equiv resin}}\right)$
H3	3.2	48.99	1.2	2.0
H4	2.0	49.95	0.8	2.4
H5 ^d	3.3	45.52	1.1	3.3
H6-1 ^d	3.3	46.13	1.0	3.0
-2 ^d	3.2	45.08	1.0	2.9
-3 ^c	3.6	44.25	0.9	2.7
-4 ^c	3.1	44.16	0.9	3.1
H7-A	4.7	47.60	1.6	2.0
-B	5.1	47.50	1.7	1.8
-C	4.4	46.20	1.3	1.7
-D	3.4	48.17	1.1	2.0
-E	2.6	49.07	0.8	1.5
H8-A	4.0	47.17	1.3	1.6
-B	4.1	47.57	1.4	1.7
H9-1	3.6	49.78	1.4	3.4
-2	3.8	50.78	1.4	3.6

^aAll runs were made with Amberlite IRC-72 resin unless otherwise noted.

^bBulk volume flow rate (i.e., voids are included in volume).

^cSlope of operating line; see Eq. (4).

^dDuolite C-464 resin was used in this run.

Table 3. Summary of continuous contactor capacity calculations
(Loading section length, 10.2 ft)

Parameter	2-in.-ID column	4-in.-ID column
Cross-sectional area, cm^2	20	80
Max. solution flow rate (est.), cc/min	4000 ^a	11,000 ^b
Max. superficial velocity, cm/min	200	138
ADUN flow suitable for complete loading, cc/min	1700	6,800
Corresponding superficial velocity, cm-min	85	85
Inlet uranium conc. ($x = 1.25$), $\underline{N}$	0.50	0.50
Outlet uranium conc. ($x = 0.975$), ^c $\underline{N}$	0.39	0.39
Uranium rate (fully loaded), kg/hr	1.4	5.6

^aBased on a pressure drop of 55 psig.

^bBased on a pressure drop of 35 psig.

^c $LC_x/VC_y = 3.5$; see Fig. 10.

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