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SOLVENT-IN-PULP PROCESSING USING SIEVE PLATE PULSE COLUMNS

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EXTRACTION METALLURGY DIVISION

Reprinted from Chemistry and Industry

Published by Society of Chemical Industry

6 November 1971

Reprint Series RS 107

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Information Canada
Ottawa, 1972

Solvent-in-pulp processing using sieve plate pulse columns

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In the present context pulp is the unfiltered solution from ore leaching operations at mining sites. The possibility of utilising ion exchange or solvent extraction techniques for recovery from pulps has received much attention. Resin-in-pulp has been used in US plants but has never been used in Canada. So far neither the US experience nor any other studies have indicated that resin-in-pulp has any real advantage over the ion exchange purification of clear solution as practiced in Canada. However, solvent-in-pulp techniques appear more attractive because of the potential cost saving in capital and operating of filters, and because they decrease soluble metal losses encountered in a liquid-solids separation. In test work using sieve-plate pulse columns, throughput capacity has not been seriously affected by the pulp density in the range of 30 to 40 per cent solids, and the high solvent losses reported in other solvent-in-pulp systems have not been encountered. Both Eldorado Nuclear Ltd and the Mines Branch have done considerable work on this process. Solvent losses in the solvent-in-pulp system, employing sieve-plate pulse columns, have been remarkably low, about 0·1 lb/ton dry feed. This loss compares favourably with a liquid-liquid operation.

The first successful work on solvent-in-pulp recovery of metals to be reported was that on the extraction of uranium from a sulphuric acid leach slurry of a flotation concentrate containing pyrite and graphite,[1] using an amine extractant. The equipment used consisted of a 10in diameter extraction pulse column and three mixer-settlers for scrubbing, stripping and acid equilibration. A brief review of the highlights of this investigation, and subsequent work at the Mines Branch together with new information are described here with estimated processing costs.

Description of equipment

Solvent-in-pulp extraction was effected in a 10in diameter stainless steel pulse plate column as shown in Fig. 1. Stainless steel discs perforated with $\frac{3}{8}$in diameter holes, totalling 27 per cent of the disc area, were spaced 2in apart over a length of 12ft (2×6ft sections). Disengagement chambers were situated at the top and bottom of the extraction section. The pulse was applied by means of a diaphragm pump protected from acid attack by an organic leg. Solvent was fed to the column *via* the pulse leg through a 0·5in diameter pipe. Pulp feed was regulated by a variable speed screw type slurry pump.

The flowsheet for the complete pilot plant is shown in Fig. 2. The loaded organic phase was pumped through a plate and frame clarifier to remove entrained graphite and

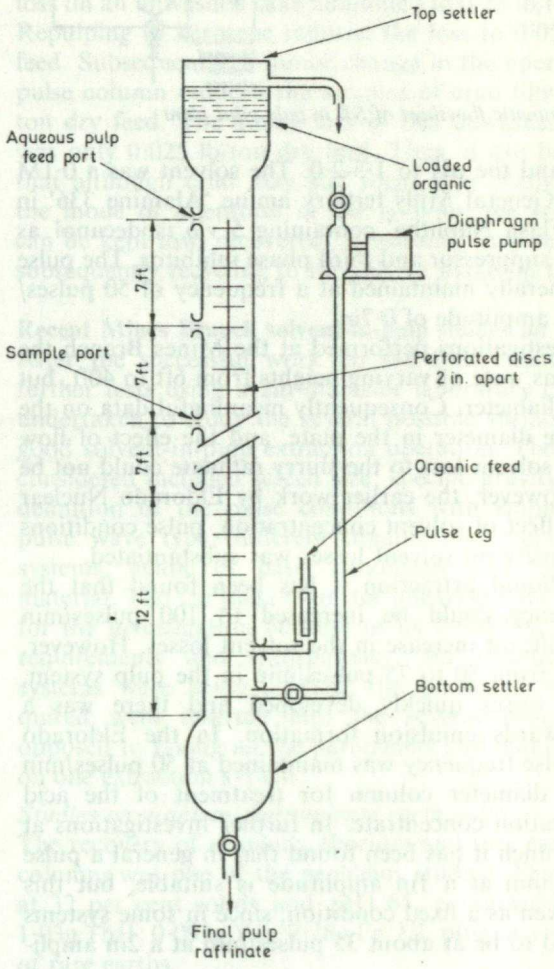

Fig. 1 *Sieve plate pulse column*

then to a mixer-settler for the removal of iron impurity by water scrubbing. The mixer-settler unit consisted of a baffled 30gal. steel tank located in one compartment of a baffled 4ft by 8ft by 2ft deep epoxy-coated wooden settler. A second mixer-settler unit was used for stripping the organic phase with plant carbonated barren solution. A final mixer-settler was used for acidification of the stripped organic phase (maintained at pH0·5 to minimise possible crud formation) prior to re-entry to the pulse column.

Operating experience

Acid slurry from the mill circuit was stored in a pachuca at approximately 60 per cent solids and a pH greater than 2. In the majority of the tests, the density was adjusted to 30 per

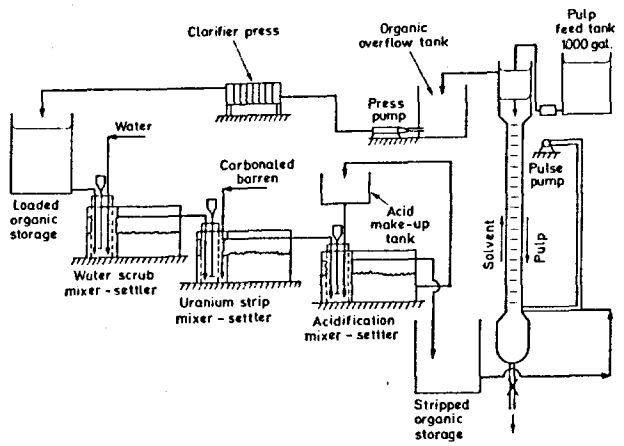

Fig. 2 *Diagrammatic flowsheet of SX in pulp pilot plant*

cent solids and the pH to 1·5-2·0. The solvent was a 0·1M solution of General Mills tertiary amine 'Alamine 336' in Shell '140 Flash Naphtha' containing 5 *v/o* isodecanol as an emulsion suppressor and third phase inhibitor. The pulse rate was generally maintained at a frequency of 50 pulses/min with an amplitude of 0·7in.

In the investigations performed at the Mines Branch the pulse columns were of varying heights from 6ft to 40ft, but only 2in in diameter. Consequently meaningful data on the effect of hole diameter in the plate, and the effect of flow rates on the solvent loss to the slurry raffinate could not be obtained. However, the earlier work by Eldorado Nuclear Ltd on the effect of solvent concentration, pulse conditions and pulp density on solvent losses was substantiated.

In liquid-liquid extraction it has been found that the pulse frequency could be increased to 100 pulses/min without significant increase in the solvent losses. However, on changing from 50 to 75 pulses/min in the pulp system, high amine losses quickly developed and there was a tendency towards emulsion formation. In the Eldorado work, the pulse frequency was maintained at 50 pulses/min in the 10in diameter column for treatment of the acid slurry of flotation concentrate. In further investigations at the Mines Branch it has been found that in general a pulse of 50 pulses/min at a 1in amplitude is suitable, but this cannot be taken as a fixed condition, since in some systems operation had to be at about 35 pulses/min at a 2in amplitude.

There were two sizes of sieve plates tested during the operation of the Eldorado pilot plant. These tests indicated approximately the same level of uranium extraction in both types of plates, but amine loss using the small holed plates ($\frac{3}{16}$in, 32 per cent free area) was much greater than with the larger holed plates ($\frac{3}{8}$in, 27 per cent free area). Therefore, further work in the 10in diameter column used plates with $\frac{3}{8}$in diameter holes.

In the work at Eldorado and the Mines Branch, with slurries of pulp densities of the order of 30-40 per cent solids (s.g. approximately 2·7), an increase in the solvent loss with increase in pulp density occurred. The work by Eldorado on the 10in diameter column showed that up to 35 per cent solids can be treated while maintaining the solvent losses at less than 0·1 lb/ton dry feed at flow rates up to 5gal./min of slurry. Work with the 2in diameter column showed that 30-40 per cent solids could be tolerated, resulting in an amine loss to the raffinate of slightly greater than 0·1 lb amine/ton dry feed. The lower losses in the 10in diameter column, compared to the 2in column, can probably be attributed to the greater wall effects in the 2in diameter column. Some typical results for the 10in diameter column are shown in Fig. 3.

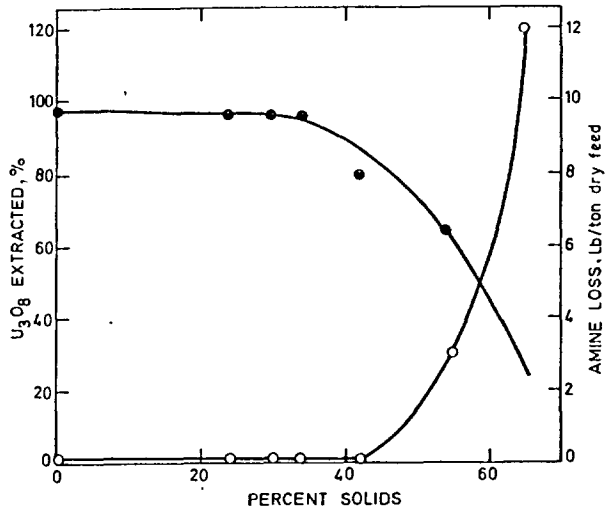

Fig. 3 *Effect of per cent solids.* ○ *amine loss;* ● *uranium extraction; flow 5gal./min*

The effect of varying the feed slurry flow from 1-7gal./min at 30 per cent solids on uranium extraction and amine loss is shown in Fig. 4.

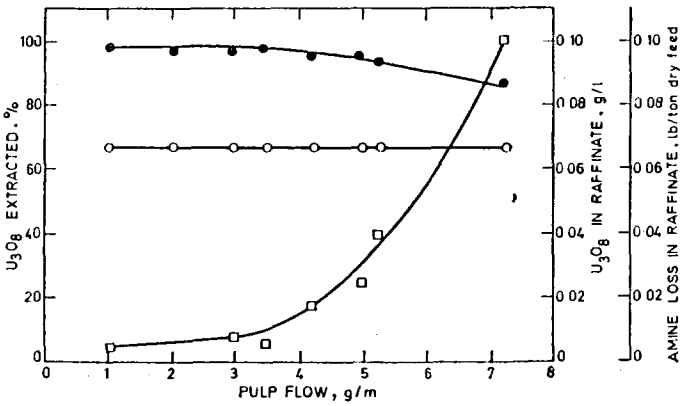

Fig. 4 *Effect of pulp flow.* ● *uranium extraction;* ○ *amine loss;* □ *uranium in raffinate; pulp density 30 per cent solids*

The use of concentrated solvents in a solvent-in-pulp system is not desirable since the loss of solvent to the raffinate is greatly increased. At the Mines Branch the effect of increasing the solvent concentration from 0·1M to 0·25M in systems using primary, secondary or tertiary amines was investigated, as well as a system using di-(2-ethylhexyl) phosphoric acid. The results were similar to those obtained by Eldorado for the tertiary amine – that is, the solvent losses increased by at least a factor of 10 with an increase in solvent concentration from 0·1 to 0·25M.

A comparison of solvent losses of Eldorado's solvent-in-pulp process with solvent losses from a liquid-liquid

Table I

Comparison of amine losses in raffinate using various contact systems for extraction of uranium

Amine loss	*Mixer-settlers Liquid-liquid*	*Rotary film contactor Solvent-in-pulp*	*Needle columns Solvent-in-pulp*	*Pulsed-plate column Solvent-in-pulp*
lb/1000 gal. feed solution	0·22	System forms stable emulsions, and amine losses high due to adsorption on gangue	0·32	0·13
lb/ton feed solution or slurry	0·038		0·05	0·02
lb/ton dry feed	0·026		0·16	0·067

mixer-settler operation[2] shows 0·22 lb amine/1000 gal. feed using the liquid-liquid system, and 0·13 lb amine/1000 gal. feed solution using the solvent-in-pulp system. Amine losses in the mixer-settler circuit after column solvent extraction-in-pulp were found to be in the order of 0·0001 lb/1000 gal. of feed. Organic matter entrained in the filtered graphite was recovered by repulping in kerosene and recycling this organic repulp as make-up to the circuit where necessary.

Other solvent-in-pulp systems have been proposed, such as one using the rotary disc contactor[3] and another[4] using fine needles to disperse the organic phase up through a hollow column. A comparison of the solvent losses in these systems is shown in Table I.

Solvent loss measurements were made at the Mines Branch on the solids and solution in a slurry extraction system for the extraction of uranium from bacteria-leached solutions using a tertiary amine. Pilot plant tests using a 2in diameter column showed that 90 per cent of the total solvent loss was caused by absorption on to solids particles. Only 10 per cent of the total solvent loss was present in the aqueous solution. A summary is given in Table II.

Table II

Summary of solvent-in-pulp pilot plant on uranium recovery from bacterial leach slurry

Feed 0·25g U_3O_8/l
7·2g Fe/l
Pulse 36/min, 2in amplitude
Solvent 2·5 *v/o* 'Alamine 336'+5 *v/o* isodecanol
Flows Aqueous – 900ml/min
Solvent – 100ml/min

		Amine losses					
Per cent solids	*s.g. of solids*	*In solution (g/l slurry)*	*On solids (g/l slurry)*	*Total (g/l slurry)*	*lb/1000 gal. slurry*	*lb/ton feed slurry*	*lb/ton dry feed*
25	1·185	0·0017	0·017	0·0187	0·187	0·0325	0·13
35	1.270	0·002	0·023	0·025	0·25	0·0394	0·13
40	1·320	0·002	0·030	0·032	0·32	0·0485	0·21

Crud formation during solvent-in-pulp extraction

Quite frequently during extraction, factors such as fineness of grind, preferential flotation of ore particles such as pyrite or graphite, excessive agitation, insufficient or no emulsion inhibitor, or presence of chemicals added during the leaching, may cause stable emulsions or 'cruds'. These cruds that are encountered can very frequently be broken down, either during the extraction, if they are in small quantity, by non-wetting materials such as 'Teflon'; or later by allowing the crud to rise in the column and overflow with the extract and then filtering to recover the extract.

In one pilot plant operation the amount of crud overflowing with the extract was about 100 lb/ton dry feed. After filtering through a plate-and-frame press, the amine loss on an unwashed cake amounted to 0·28 lb/ton dry feed. Repulping in kerosene reduced the loss to 0·05 lb/ton dry feed. Subsequently, a minor change in the operation of the pulse column reduced the amount of crud filtered to 5 lb/ton dry feed. The amine loss in this unwashed filter cake was only 0·025 lb/ton dry feed. Thus, it can be concluded that although crud may still form after many changes in the mode of operation of the column, the solvent losses can be kept low, recovered by washing with kerosene, and subsequently recycling to the solvent make-up tank.

Recent Mines Branch solvent-in-pulp studies on ore slurries

After the successful work on the 10in diameter column further tests using a 2in diameter laboratory column were undertaken to study the several possible variables affecting good solvent-in-pulp extraction operation. The parameters considered included screen size; specific gravity of the ore; definition of the pulse conditions with emphasis on the pulse wave type; different organic and metal extraction systems; acidic or basic conditions and type of plate material. The 2in glass diameter pulsed column was chosen for the investigation because of its flexibility, low volume requirements and transparency. As specific extraction systems were being studied, the particular variables required were inserted into the several programmes, as opposed to taking all the parameters and making the study on one extraction system.

Studies on uranium/thorium/rare earth pulps

The recovery of uranium, thorium and rare earths in pulse columns was one of the problems studied. The slurry feed, at 32 per cent solids and pH1·65, contained: 0·77g U/1; 1·03g Th/1; 0·092g Y/1; 7·28g Fe^{+3}/1, plus varying quantities of rare earths.

This feed was derived from a bacterial leach of an Elliot Lake ore carried out in the summer of 1969, and originally uranium had been extracted by a solvent-in-pulp process at that time. For this series of tests, the uranium content was brought back to near the original concentration by the addition of a solution of uranyl sulphate. The solids in this slurry, (92 per cent−325 mesh) were much finer than normally encountered in uranium circuits.

Generally it was found that the throughput of the solvent-in-pulp system was a little lower than was used with clarified solutions, say about 10 to 15 per cent. The various pulse conditions noted for each system were selected to best suit the particular system. The pulse conditions in Table III were the optimum for each system.

Although this leach slurry contained only trace amounts

Table III

Sequential extraction

2in diameter column; 32 per cent solids pH 1·65
0·77g U/l; 1·03g Th/l; 0·092g Y/l; 7·28g Fe^{+3}/l

		Pulse conditions				*Raffinate*		*Extraction*	
Metal Extracted	*Extractant*	*Phase ratio A/O*	*Total flow (ml/min)*	*Frequency (pulses/min)*	*Amplitude (in)*	*U (g/l)*	*Th (g/l)*	*U (%)*	*Th (%)*
U	5 *v/o* 'Alamine 336' +5 *v/o* isodecanol	6/1	900	62	1	0·0008		99·9	
Th	5 *v/o* 'Primene JM-T' + 5 per cent isodecanol	3/1	800	44	1		0·005		99·5
Rare earths	3·3 per cent D2EHPA +5 per cent tributyl phosphate	10/1	700	50	1				

of the rare earths the physical characteristics of extraction using D2EHPA was studied on the raffinate from thorium extraction. Physically the solvent-in-pulp operation using D2EHPA was good. No chemical data were taken on this system because of the unrepresentative concentrations of the rare earths.

Unfortunately no solvent loss values were determined from these runs since the slurry had been previously used. However, it can be safely assumed that the losses would be no higher than previously reported, that is, in the range of 0·1 lb amine/ton of dry feed. No suitable analytical method has been developed for the determination of the losses of the alkylphosphate, but since this organophosphate is known to be less soluble than amines in aqueous solutions, the losses for this system can be expected to be at least as low as the amine system, judging by the appearance of the raffinate.

From these pilot plant studies it is a reasonable assumption that a leach slurry can be processed by the solvent-in-pulp technique for the step recovery of several metals. The use of an alkylphosphate after an amine appears feasible, and any entrained solvent from one extraction system appears to have no chemical or physical effect on the subsequent system. The mesh size of the feed material does not appear critical. One of the advantages of the sieve-plate pulse column is that it provides a number of theoretical stages of contact, as well as the gentle action required for mass transfer in a pulp system.

Separation of copper from cobalt, nickel and iron

Previous work showed that copper could be effectively extracted and separated from Co, Ni, and Fe, in the pH range 0·8-2·0, using a mixed solvent consisting of 10 per cent LIX 63+10 per cent D2EHPA.[5] The results of a solvent extraction-in-pulp operation, in a 2in diameter pulse column, are shown in Table IV, indicating excellent discrimination between the copper and the cobalt and nickel.[6]

In subsequent work at the Mines Branch, a sulphuric acid pressure leach of a nickel ore (s.g. 3·7) resulted in a leach slurry which was impossible to filter in a reasonable time. Therefore, solvent extraction-in-pulp was looked to as a possible solution to the problem, using the Eldorado process of the mixed extractant previously described. Because of the difficulty in filtering, tests were conducted in a small, 6ft high pulsed column to determine whether the metals could be sequentially recovered from such a slurry. Copper was extracted first because the pH of the slurry (1·15) was suitable. The mixed organic system of 10 per cent LIX 63+5 per cent D2EHPA was used for the separation of copper (3·53g/l) in the presence of cobalt (0·53g/l), nickel (17·8g/l), and iron (10·2g/l), from a 20 per cent leach slurry. The flow ratio of organic to aqueous was 800/400ml/min, with a pulse frequency of 46/min at a 2in amplitude.

Table IV

Conditions and results of copper extraction from leached pulp in 2in diameter pulse column

Conditions	Total flow	8gal./h		
	A/O	1·0		
	Pulse amplitude	3/4in		
	Pulse frequency	52		
	Initial pH	2·5		
	Terminal pH	2·2		
	NH_4OH added	1 per cent of aqueous flow as 1·2M NH_4OH		
Results		*Cu*	*Ni*	*Co*
	Pulp feed analysis (g/l)	6·4	2·8	5·1
	Organic	6·5	0·014	0·017
	Raffinate	0·04		

Although the column used for these tests was very short, having insufficient theoretical stages for complete copper extraction, the system worked well as regards solvent entrainment losses. The extract was stripped with 15 *v/o* H_2SO_4 at 60°C, and showed 60 per cent copper extraction, with no indication of extracted nickel, cobalt, or iron. However, a small amount of fine solids floated with the extract, which was identified as chalcopyrite. This selectivity of ore floating with the extract could possibly be made use of in certain systems, and recovered from the extract for further processing by a filtration stage.

The raffinate, after removing the copper at pH1·15, was neutralised with $CaCO_3$ to pH5. Subsequent attempts to extract cobalt and nickel from such a leach slurry containing precipitated ferric hydroxide, using 30 per cent D2EHPA,[7] failed. The problem was extremely fine iron precipitate floating over with the extract. Because of the fine chemical precipitate, the rising organic phase bubbles were coated, and consequently the extraction of cobalt and nickel was depressed. Further work in this area is definitely warranted in order to achieve a successful operation under such slurry conditions. Tests to be performed include use of surface active agents to affect flocculation, wetting of the particles, and so on.

There were no visible losses of D2EHPA and LIX-63 to the slurry raffinate. Although no analyses for these solvents in the raffinate were carried out, D2EHPA losses are reported in the literature to be less than about 20 p.p.m.,[8] and LIX-63 losses, reported by Swanson and Agers,[9] are also sufficiently small to make the use of this solvent economically feasible.

Evaluation of equipment for solvent-in-pulp processing

In Table V the results of applying various types of liquid-liquid contactors to the recovery of metals from leach slurries are noted. The mixer-settlers, mixing columns, centrifuges and one type of Graesser contactor have been tried and found unsuitable for processing slurries. It is concluded that, of the equipment tested by the author and by others for the recovery of metals from leach slurries, the sieve-plate pulse column and the grid packed column of Byerlee[11] is the only equipment amenable. The main reasons for this are the simplicity of design, no moving parts, use of gravity to move the solids, and gentle agitation without extreme mixing as is encountered in other extraction equipment. This method of achieving mixing and therefore mass transfer of the metals enables extraction to take place from slurry systems as well as from clarified solutions.

Table V

Comparison of equipment for solvent-in-pulp processing

Equipment type	*Results and observations*
Mixer-settlers	Formation of stable emulsions
Centrifuges (Podbielniak)	Holes plug as well as formation of stable emulsions
Horizontal contactor (Graesser)	Ore particles not transported – no wall clearance
Agitated columns (Mixco)	Can tolerate in the range up to 10 per cent solids, but a tendency to form emulsions
Sieve plate pulse columns	Operated satisfactorily up to 40 per cent solids with low solvent losses

Tests were performed on a 2ft long, 4in diameter Graesser contactor, using a leach slurry containing 90 per cent – 325 mesh solids obtained from sulphuric acid leaching of a copper concentrate. The slowest operating speed was 10 r.p.m. using the sprockets supplied with the equipment, but from studies of a liquid-liquid system, this speed appeared reasonable for a solvent-in-pulp operation with respect to possible emulsion tendencies. However, on feeding a slurry containing 30 per cent solids into the contactor containing a clear acid solution, the contactor had to be quickly shut down because the tolerance between the wall and the cup mechanism was not sufficient to pass even these fine solids. The pulp density in the contactor at time of shut down was less than 10 per cent solids. The process would not work with the contactor tilted or in the level position. It can only be concluded that, although this contactor is suitable for liquid-liquid processing, a major redesign would be necessary before pulp could be treated.

Solvent-in-pulp costs and comparison with other systems

In Table VI some estimated costs for the recovery of uranium by ion exchange or solvent extraction, from either clarified solutions or slurries, are given. The basis for comparison is for an Elliot Lake mill with an ore grading 0·1 per cent U_3O_8, produced at a rate of 3000 ton/d. The values for liquid-solids separation and sand/slimes separation were reported recently by Smith and Garrett.[10] The cost of solvent extraction from filtered and clarified solutions is estimated to be slightly cheaper than using ion exchange by approximately 2·5 cent/lb U_3O_8. However, in extraction from slurries, the solvent extraction route has a cost advantage of 8·2 cent/lb U_3O_8, with an estimated 20 cent/lb U_3O_8 compared to 28·2 cent/lb U_3O_8 for ion exchange. Also, the cost of solvent extraction from clarified solutions is approximately twice the cost of solvent extraction from slurries. With such a cost differential a mill could well afford extra processing costs of the slurry (such as flocculation, addition of depressants, surface active agents, and so on), small extra solvent losses, and possible necessity of filtration of the extract, in order to decrease processing costs. In the example shown, a possible cost incurred by filtering the extract through a plate and frame press has been included.

Table VI

Comparison of total costs of ion exchange and solvent extraction from clarified solutions or slurries
(Elliot Lake Ore, 0·1 per cent U_3O_8)

	Cost/lb U_3O_8 (cent)			
	Clarified solutions		*Slurries*	
	IX	*SX*	*IX*	*SX*
Liquid-solids separation				
Operating	13·0	13·0		
Equipment	14·0	14·0		
Sand/Slimes separation				
Operating			5·0	
Equipment			6·0	
Solvent filtration				0·5
Resin or solvent loss	1·0	3·0	1·5	4·5
Elution or stripping				
Reagents	5·7	1·2	5·7	1·2
Depreciation of equipment and instrumentation	7·0	7·0	7·0	10·8
Maintenance and labour	2·5	2·5	3·0	3·0
Totals	43·2	40·7	28·2	20·0

10 year straight line depreciation of about $630,000 for the IX or SX plants. For SX in pulp, depreciation of about $980,000. The capital cost multiplied by 2 to give approximate installed cost. Plate and frame for solvent – 2 @ $36,000

Conclusions

A process has been developed to recover metal values from acid slurries thus eliminating costly filtration. Throughput rates achieved in the pilot plant operation using a 10in diameter pulse column were 13·5 ton/d dry solids. The amine loss was 0·07-0·10 lb/ton dry feed and the uranium recovery was 98 per cent. Additional work on other slurry systems has substantiated this earlier work. Although other equipment has been proposed for recovery of uranium from slurries, it is believed that the advantages of using sieve plate pulse columns are of great interest, not only in the uranium field but in many other metallurgical operations.

The main advantage of this process is the elimination of

the capital and operating costs of filtration. Throughput capacity has not been seriously affected by the pulp density in the range of 30-40 per cent solids, and high solvent losses, which have been reported in other solvent-in-pulp systems, were not experienced. By use of solvent-in-pulp extraction the processing of slurries containing even low grade metal values becomes an important aspect in metal recovery plants.

References

1 Ritcey, G. M., Joe, E. G. & Ashbrook, A. W., 'Solvent-in-pulp extraction of uranium from acid leach slurries', *Trans. AIME*, 1967, **238**, 330-334

2 Tremblay, R. & Bramwell, P., 'Eldorado's solvent extraction plant at Port Radium, NW.T.', *C.I.M.M. Trans.*, 1959, **62**, 44

3 North, A. A. & Wells, R. A., 'Solvent extraction of uranium from slurries by means of a rotary-film contactor', *Trans. Inst. Min. and Met.*, 1964, **74**, 463

4 Boutin, P. & Melvanin, F. W., 'Method and apparatus for the extraction of minerals', Canadian Patent 756,991, April 1967

5 Ashbrook, A. W., Ritcey, G. M. & Joe, E. G., 'Mixed extractant and process for separating copper from cobalt and nickel in acid medium', US Patent 3,455,680, July 1969

6 Joe, E. G., Ritcey, G. M. & Ashbrook, A. W., 'The Eldorado process for liquid ion exchange-in-pulp of uranium and copper', *Journal of Metals*, January 1966, 18

7 Ritcey, G. M. & Ashbrook, A. W., 'Separation of cobalt and nickel by liquid-liquid extraction from acid solutions', U.S. Patent 3,399,055, August 1968

8 Blake, C. A., Brown, K. B. & Coleman, C. F., ORNL, 1955, 1903

9 Swanson, R. R. & Agers, D. W., 'A new reagent for the extraction of copper', 1964, *AIME*

10 Smith, S. E. & Garrett, K. H., 'UK developments in uranium ore processing with potential for commercial application', presented at the October 1970 meeting of the Canadian Uranium Producers, Ottawa, Ontario

11 Byerlee, H. W., 'The solvent extraction of uranium from slurries', presented at the 1969 Annual Conference of the Aus. I.M.M., Sydney, New South Wales