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I. R. Higgins

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CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section

MECHANICAL FEATURES OF THE HIGGINS CONTINUOUS ION EXCHANGE COLUMN

I. R. Higgins

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0.0 ABSTRACT

The accessory equipment for the Higgins continuous ion-exchange contactor is described. Photographs of various systems used during the development work are shown.

1.0 INTRODUCTION

This report* presents the details of the equipment used in constructing a continuous ion-exchange contactor. The mechanical accessories recommended are those that have been found satisfactory, and are not necessarily the best that could be found since detailed engineering studies have not been made. The recommendations are based on three years of development work and testing and several thousand hours of reliable column operation.

The principles of the contactor design and operation were presented at an AIChE meeting in December, 1953.** Briefly, the advantages of a continuous moving-bed system as compared with a fixed bed are lower costs due to lower resin inventory, reduced chemical consumption, greater separation and concentration efficiency, and higher uniformity of product. Advantageous features of the Higgins continuous contactor are: The HTU and HETS*** are minimized because the resin particles

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* A revision of an ORNL internal memorandum, CF-54-3-90, March, 1954.

** I. R. Higgins and J. T. Roberts, "A Countercurrent Solid-Liquid Contactor for Continuous Ion Exchange," Chem. Eng. Symposium Series, 50(14): 87-92 (1954).

*** "Height of a transfer unit" and "height equivalent to a theoretical stage."

maintain their positions relative to each other. Resin flow is accomplished in a gentle and easily controlled manner. The aqueous flow rate is not limited because of solution density or the possibility of fluidization of the resin, as is the case in many systems where the solutions flow upward and the resin falls by gravity. The mechanical accessories are almost entirely standard equipment. An application for a patent on this contacting device has been filed.

The author wishes to acknowledge the assistance received in the studies from other members of the Chemical Technology Division, particularly, W. E. Shockley who provided the nontechnical assistance throughout the development program, J. T. Roberts who was largely responsible for designing the ion exchange experiments which proved the advantages and efficiency of the contactor, T. A. Arehart who designed the first completely automatic model, C. W. Hancher who designed and operated the large-scale models, and T. M. Gayle of the Instrumentation and Controls Division who designed the instrument controls and suggested some of the mechanical accessories.

2.0 CONSTRUCTION AND OPERATION OF THE CONTACTOR

The contactor is constructed of individual fixed bed sections which are connected to form a single resin loop. Each section is an independent unit with feed and take-off distributors and is isolated from its neighbors with a valve or water seal. There is a column section for each operational step in the process, namely, sorption, wash, and elution.

The resin is moved within the sections and from one section to another as a dense bed, much like the sliding of a piston in a cylinder. The movement is brought about by applying a sudden hydraulic pressure drop across the bed. Valving and timing are such that the resin by-passes the pump that produces the pressure drop and goes into a two-compartment

reservoir, so that it never comes in contact with the pump.

The overall operation is intermittent (see Fig. 1). Liquids are pumped for several minutes through the dense resin bed; then liquid flow is automatically shut off for a few seconds while the resin is moved one increment countercurrently to the solution flow. The solution flow is then resumed, and the cycle is repeated. During the solution pumping period the resin is locked in place by a valve, which is opened during the resin pumping period. A second valve is closed during the resin pumping period so that pressure is applied to the resin bed but the resin itself is isolated from the pump. Inlet and outlet solution flows are shut off or by-passed during the resin movement period to prevent jamming or fluidizing of the resin bed. The liquid contained in the bed is sufficient to lubricate it but not so much as to induce fluidization during the resin-movement period.

Column arrangements vary depending on the application, for example, whether the columns are being used for recovery and concentration (Fig. 2a) of a given element or for separation of two elements (Fig. 2b).

3.0 DISTRIBUTORS

The solution distributors probably affect the efficiency of operation of the contactor more than any other portion of the equipment. Unless the feed is distributed evenly over the top of the resin before it flows down the column, channeling may occur in the resin bed. Horizontal distribution of the liquid becomes a greater problem as the diameter of the column increases. A wide surface area is also required for solution take-off, but the problem is less acute at the take-off point than at the feed point. Once the feed solutions are introduced homogeneously, they travel down the column with a square wave front and are withdrawn from the column across the entire lower surface.

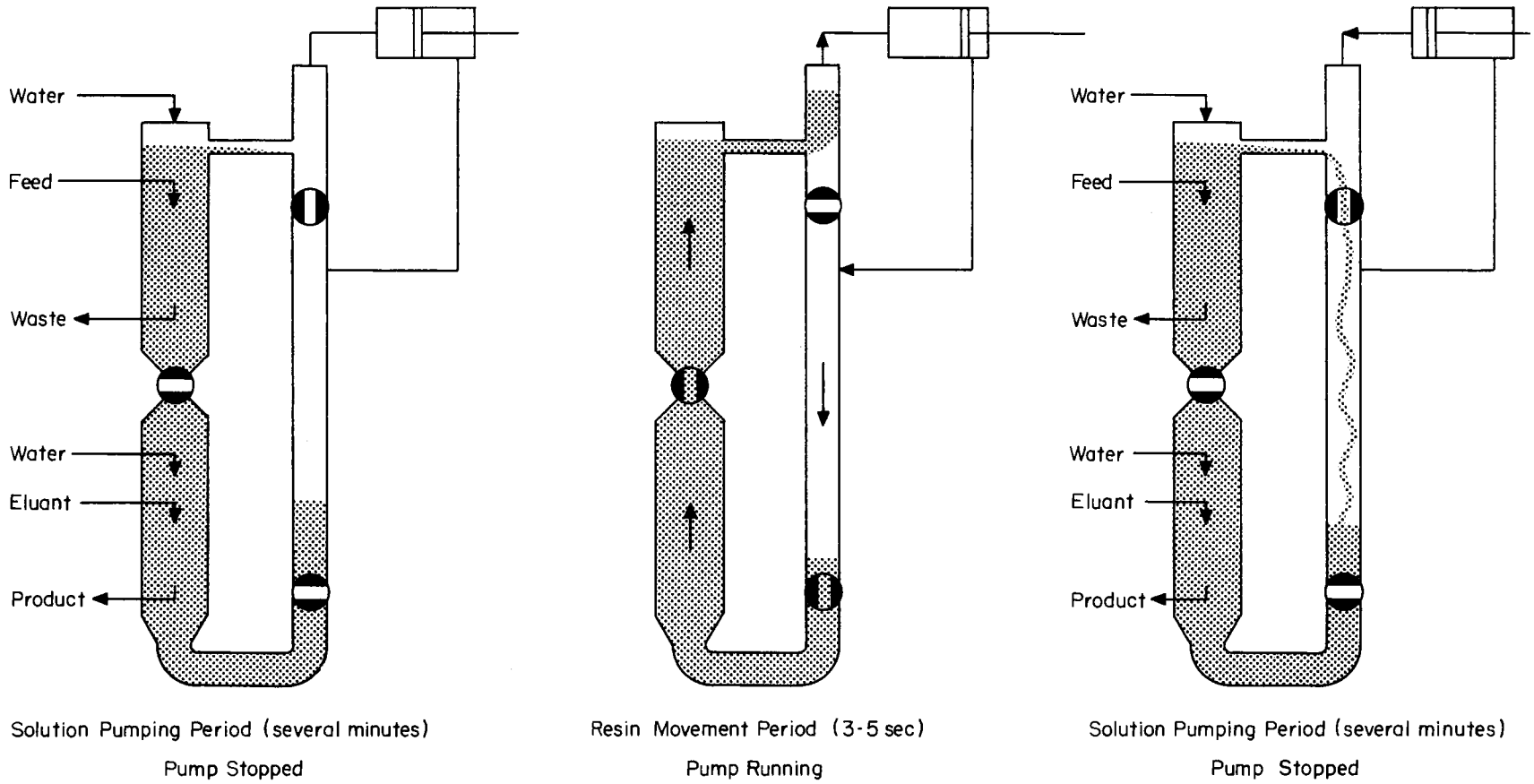

Fig. 1. Schematic Diagram of Higgins Continuous Ion Exchange Contactor, Showing Mechanism of Countercurrent Solid-Liquid Flow.

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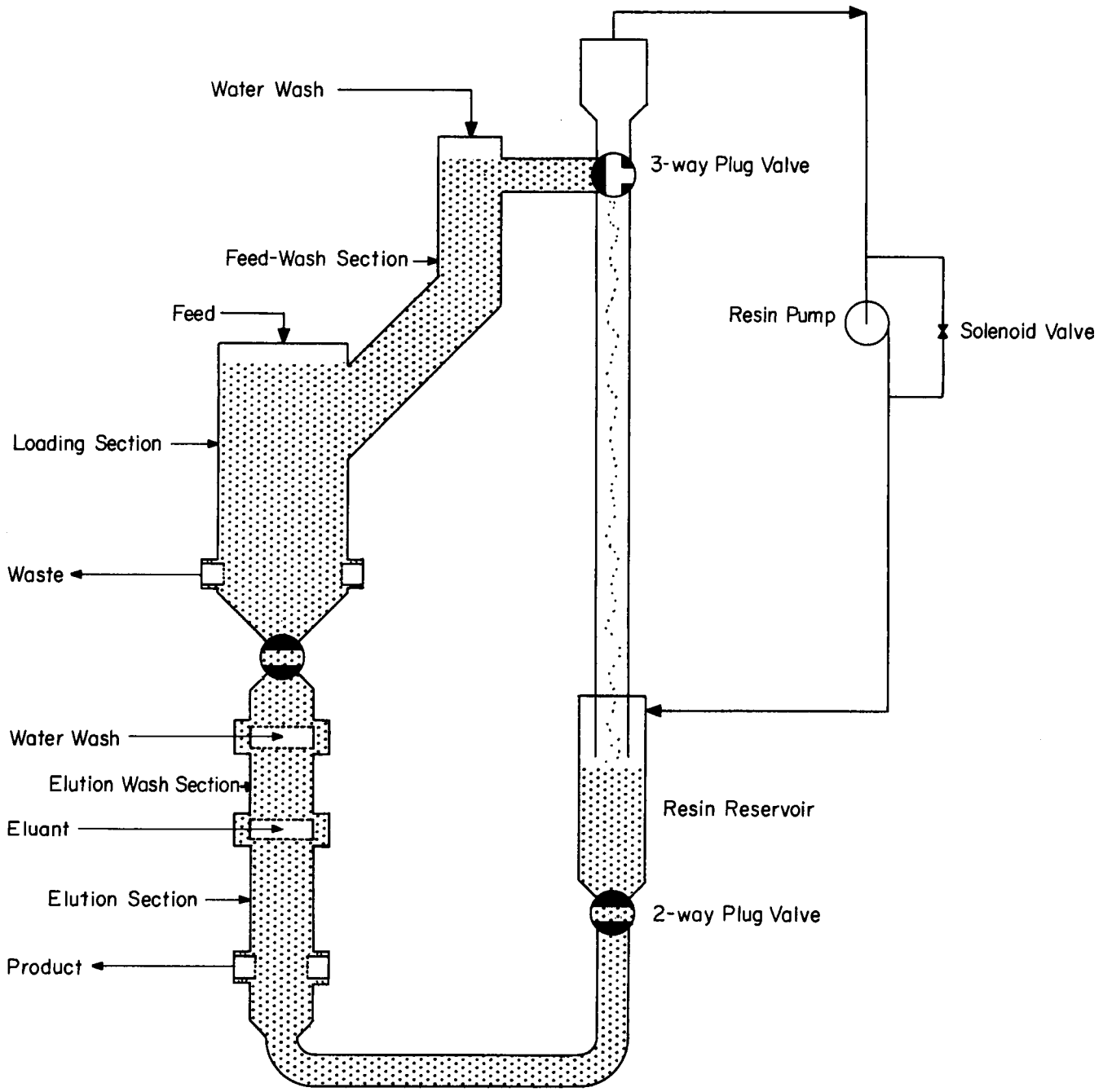

Fig. 2a. Schematic Diagram of Recovery-Concentration Contactor.

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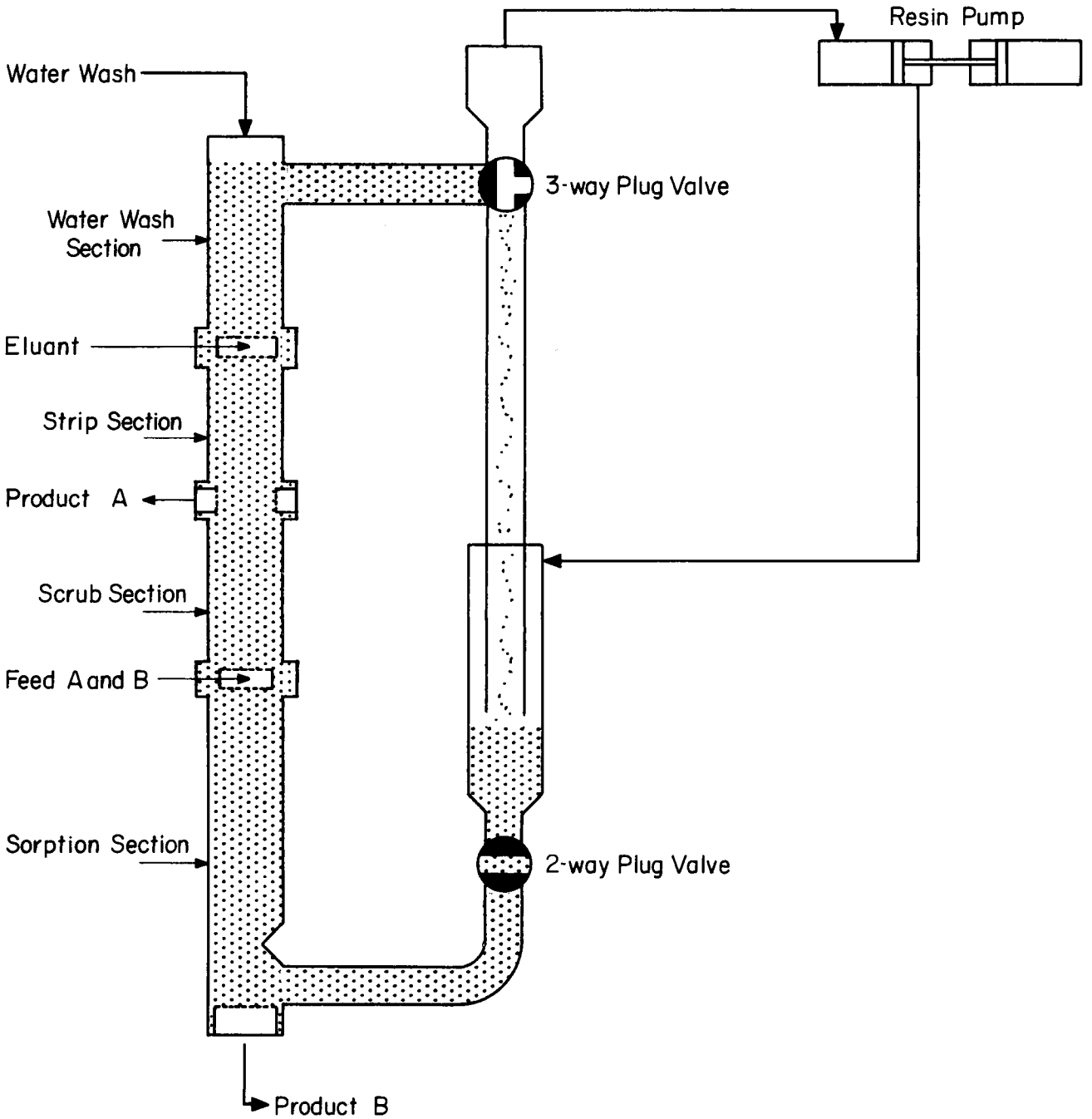

Fig. 2b. Schematic Diagram of Separation Contactor.

The most successful method of accomplishing uniform lateral distribution of the feed solution is to provide a liquid pocket in the bed at the point of solution introduction. Three feed distributors which provide such a pocket are discussed here: the tee, bell, and pipe types.

3.1 Feed Distributors

Tee Type. The tee distributor is the simplest of the three types because it consists of a standard pipe tee and has a liquid pocket and a free resin path across the full area of the column. Feed solution enters through the top of the straight-through section and wash solution through the side-arm port. Resin moves up through the bottom straight-through section and leaves through the side arm opening, into the adjoining column section. However, in this type of distributor the wash solution entering the side arm tends to move down the side of the resin column and does not mix efficiently with the feed stream. Therefore, the use of a tee distributor is limited to cases in which efficient mixing of aqueous flows is not necessary.

In some systems the liquid pocket tends to fill up with resin and becomes ineffective. In these cases the pocket can be maintained with a screen (see Fig. 3a).

Bell Type. In the bell type distributor the feed enters the side of a liquid pocket formed by a section of pipe the same diameter as the column, screened at the top and bottom. The column is expanded around this pocket to provide a path for resin flow. The wash solution enters the pocket through the top screen and mixes efficiently with the feed. Consequently, this type of feed distributor is used where the wash stream affects the distribution during sorption and efficient washing is essential, as differentiated from the tee type. For example, the bell distributor is used where the wash stream is a complexing agent or

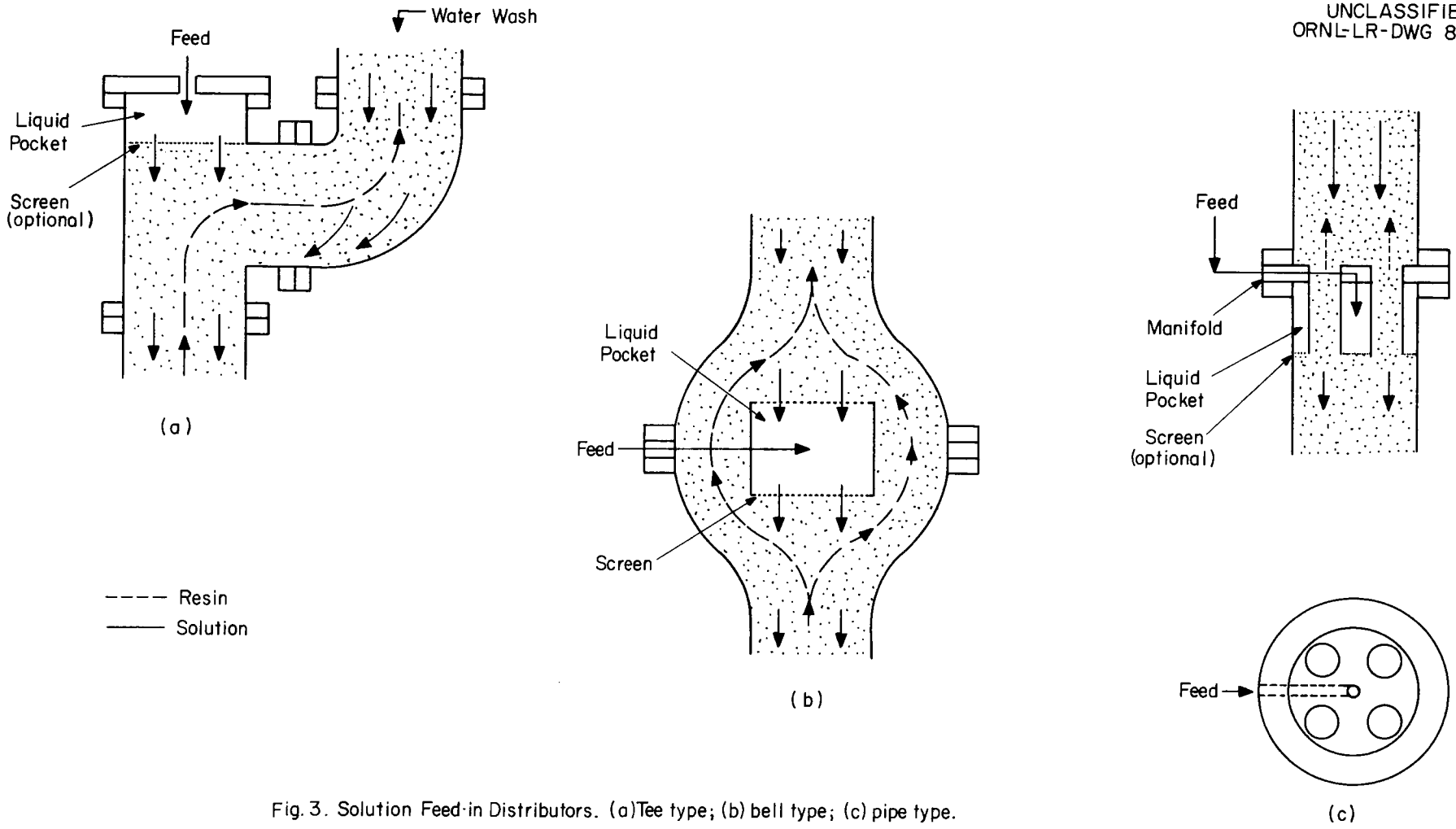

Fig. 3. Solution Feed-in Distributors. (a)Tee type; (b) bell type; (c) pipe type.

in cases in which the concentration of the feed solution is relatively important to the ion exchange process. The disadvantage of the bell type distributor is the added cost on scale-up due to the added complexity and bulkiness of the unit (see Fig. 3b). For columns smaller than about 6 in. diameter a standard bell reducer may be used to provide the resin path around the liquid pocket.

Pipe Type. The pipe type distributor divides the cross-sectional area of the column between a solution stream going down around the outside of a bundle of pipes and the resin going up through the pipes. The feed solution enters a manifold plate and is distributed to a liquid pocket, which is enclosed on the top by the manifold plate and on the sides by resin riser pipes. The wash solution passes down through the riser pipes and mixes efficiently with the feed in the top portion of the lower resin bed. The pipe type offers more resistance to resin flow than the bell type in small columns and is recommended for columns larger than 12 in. diameter. It does not require an expanded section of column, and consequently should be a cheaper unit for large-scale applications. In some applications, however, the liquid pocket fills up with resin and a screen can be used (see Fig. 3c).

3.2 Take-Off Distributors

Take-off distributors are not a problem. Once solutions have been introduced properly, they travel down the column with a square wave front. Quantitative stream removal is not a problem and occurs automatically. The chief concern with the take-off point is to provide as large an area as possible in order to minimize channeling effects. As with the feed distributor, the simplest mechanical arrangement is some modification of a tee (see Fig. 4). Type a has been used most frequently with resin entering vertically from the bottom and solution leaving horizontally from a wide area. Type b has the advantage of a full area take-off with the solution flowing in only one direction. Some resin

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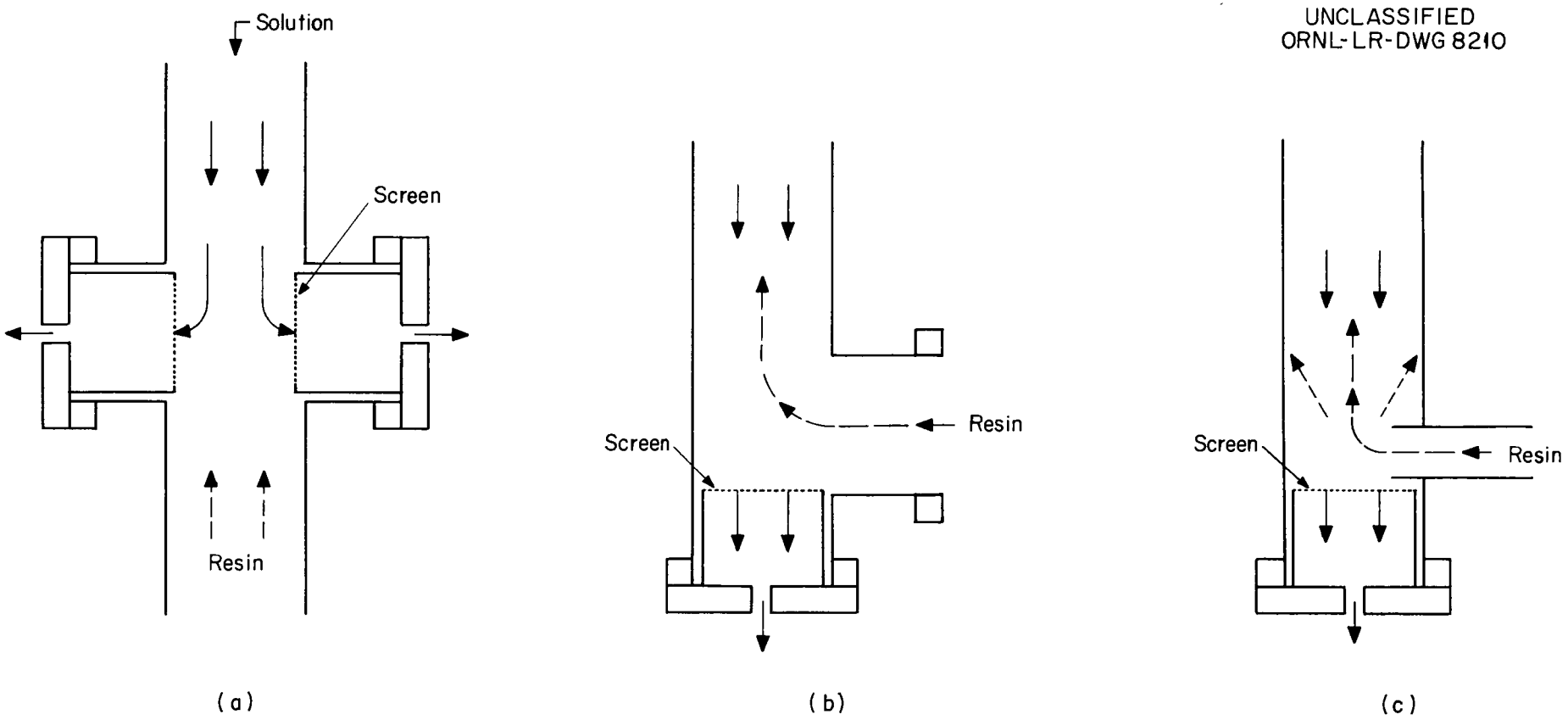

---- Resin
—— Solution

Fig. 4. Tee Type Take-off Distributor. (a) Double tee; (b) single tee; (c) single tee with center resin inlet.

channeling has been noted, however, when the resin enters horizontally. This is corrected by bringing the resin into the center of the column, as in type c. Type c is believed to be the cheapest form for larger applications.

4.0 PUMPS

4.1 Feed Pumps

A critical discussion of feed pumps is not necessary because almost any type may be used—centrifugal, gear, diaphragm, etc. There are two different methods of contactor operation which affect the choice of pump. For some chemical systems a positive displacement type pump may eliminate the necessity of many control instruments. The desired flow rates are predetermined, and the pumps must be capable of maintaining these rates. If instrument control is used to maintain the desired chemical conditions in the contactor, the tolerance on metering ability of the pumps may be relaxed. Usually there is a cost advantage in eliminating an instrument by the use of a metering pump.

4.2 Resin-Moving Pumps

The resin is moved in the Higgins contactor by a sudden application of pressure from a pump in the liquid medium. Since the resin is moved by a liquid surge from a remote pump, many types of positive displacement pumps are satisfactory. A higher capacity pump is required for resin pumping than for solution pumping. Rates on the solution pumps are controlled by the ion exchange rates, but this limitation does not apply to the resin pump. On the contrary, fast resin movement is desired to minimize the time of solution stoppage. Depending on the size and resistance of the column, from 10 to 50 psi is required for resin movement.

Both reciprocating and rotary pumps, including air-motor-driven bellows and double-acting hydraulic piston, diaphragm, and gear pumps, have been used satisfactorily. A rotor type pump need run in only one direction and therefore is less complex. Resin which has been displaced falls from the upper to the lower reservoir by gravity after the resin pumping period. A by-pass valve around the pump increases the resin fall by allowing free passage of liquid (see Fig. 5a). When a reciprocating pump is used, the first stroke moves the resin around the resin loop and the return stroke forces resin from the upper to the lower reservoir (see Fig. 5b).

5.0 VALVES

5.1 Solution Valves

The function of the solution valves is to stop all flow in and out of the resin loop during resin movement. If liquid is allowed to enter the column while the resin is moving, a section of the bed may be fluidized. The contained liquid in the bed provides lubrication during resin movement, and if liquid is allowed to leave the column, the resin will not move. A valve may be eliminated if the feed pump runs at a slow rate and is of the positive displacement type. To avoid continually stopping and starting pump motors, three-way valves should be used. Almost any type of solution valve may be used that is sufficiently corrosion-resistant and reliable; plug, solenoid, and diaphragm valves have been used. Since all the solution valves open and close at the same time, they may be ganged together in a manifold operated by one pilot valve.

5.2 Resin Valves

Valves are used in the resin circuit to isolate and direct pressures

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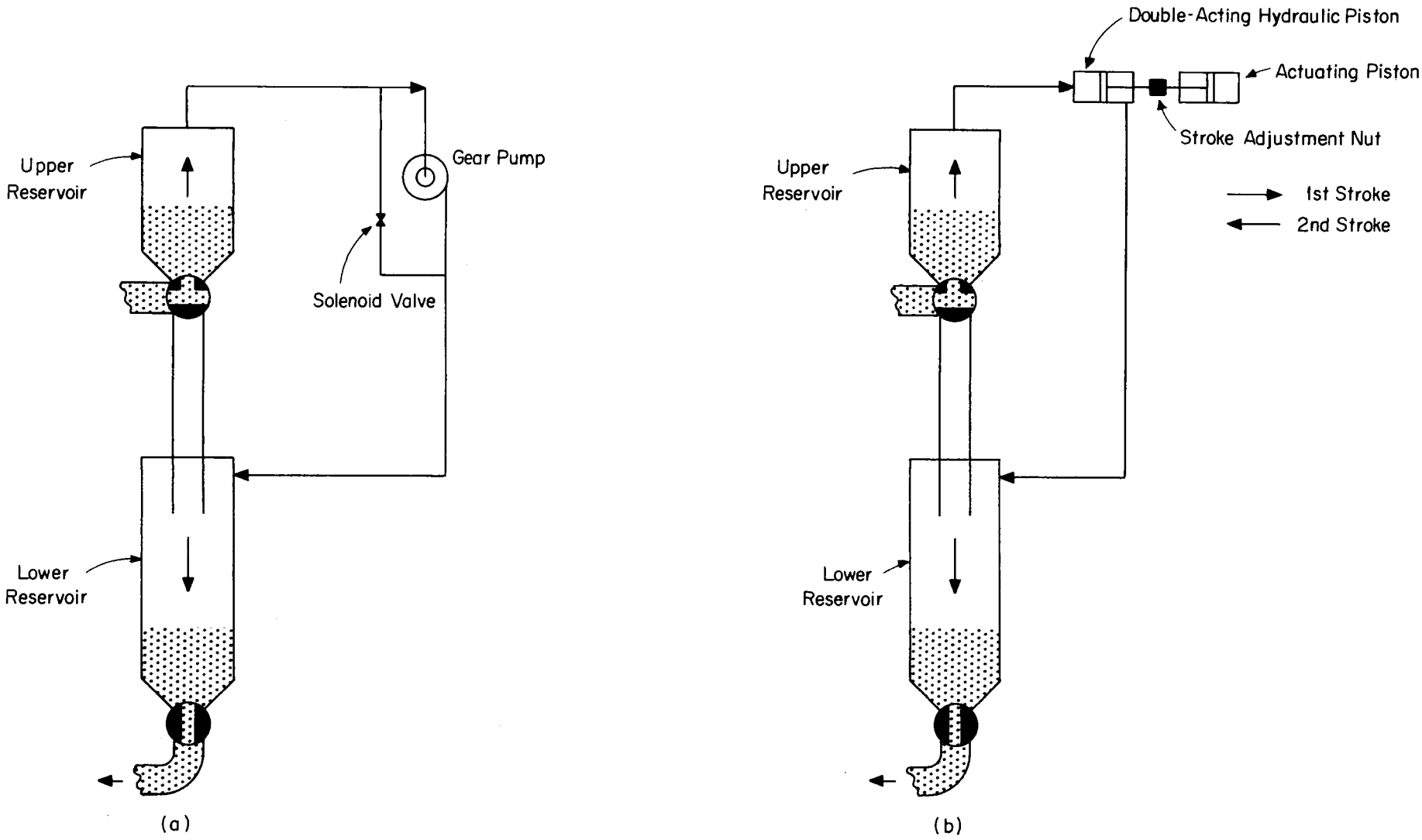


Fig. 5. Resin Pumps. (a) Rotor type; (b) reciprocating type.

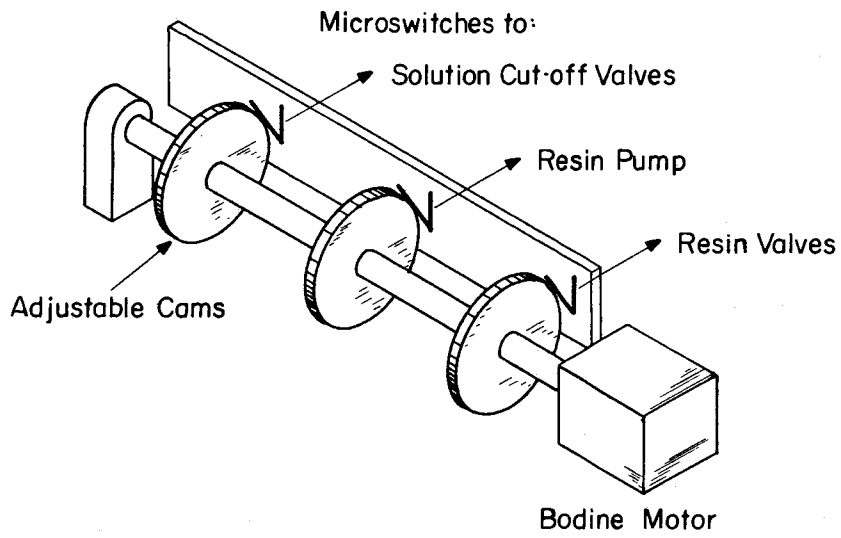
throughout the contactor to give positive performance without the need of close pressure or flow control. Plug-cock valves are favored since they are simple in construction, have a wide straight-through port, close with a shear type action which does not degrade the resin, and are available in large sizes for use with larger equipment. Valves with diameters 1/4 to 1/7 of the diameters of the column sections have been used satisfactorily. These ratios are intended as guides and do not necessarily represent a limit.

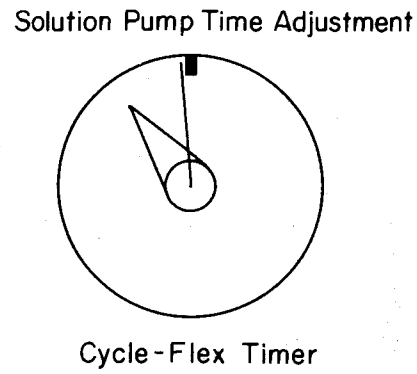
Corrosion-resistant bronze and stainless steel valves have been used. However, since the resin valves are usually located in a water-washed section of the column where corrosion is less severe, lubricated semisteel valves are preferred since they are cheaper and harder and therefore more erosion resistant.

6.0 INSTRUMENT CONTROL

The essential instrumentation for a continuous ion-exchange column is a sequence timer to synchronize the valve changes with the resin and solution flow periods. One type of timing device consists of a set of cams and microswitches to signal the sequence and duration of the various steps in a complete cycle. An adjustable cam is used for each valve change (Fig. 6a). In a simpler timer, the cams are eliminated. The valves receive a signal at the end and the beginning of the solution pumping period. A Hayden timer is included to provide a delay between valve changes and the start of the resin pump (see Fig. 6b). If a rotary type resin pump is used, a clock is required to determine resin pumping time. If the resin flow is not regulated by an instrument which indicates the equilibrium conditions in the column, a clock is used to control the solution pumping time.

Instruments for controlling interfaces or exchange zones in each column section should be included wherever practical or in situations





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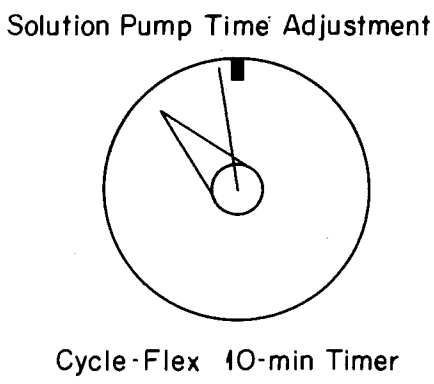

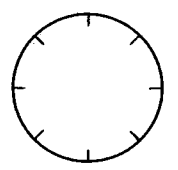

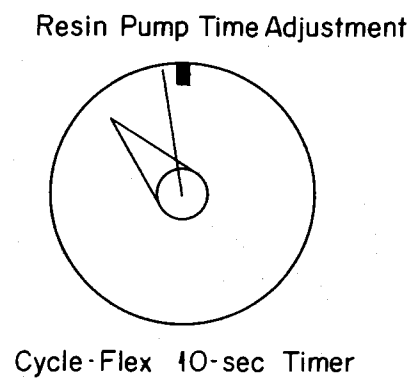

(b)

Fig. 6. Two Types of Sequence Timers. (a) Type with cam; (b) type with no cams.

where control by predetermined flows is difficult or unsatisfactory. Various properties of the resin in the exchange zone may be used to signal its position, for example, its conductivity, density, pH, refractivity, or radioactivity. Conductivity has been used frequently because of the simple instruments required.

The most frequent use of a conductivity instrument has been to eliminate void volume and slip water when maximum concentration of product is desired. For example, when water-washed loaded resin enters an elution section, it includes the water in the resin voids plus water that has traveled faster than the resin during the resin-pumping period. (Slip water is usually 2-4 void volumes.) The difference in conductivity between this incoming water and the eluant solution normally surrounding the resin in the elution section is transmitted to a set of solenoid valves, one bleeding the conducting solution and the other the water. The incoming water is thus discarded (see Fig. 7).

The difference in conductivity between a salt and its corresponding acid in a cation-hydrogen exchange has been used to control resin flow. The change in conductivity at the exchange zone controls the resin flow so that the position of the exchange zone is fixed in the column. This system is preferred to that of presetting the solution and resin flows because prediction of the position of the exchange zone, which cannot always be determined accurately, is unnecessary.

7.0 OVERALL CONTACTOR DESIGN

Major features of the contactor have been pointed out in detail. Certain general features should also be considered before a contactor is assembled.

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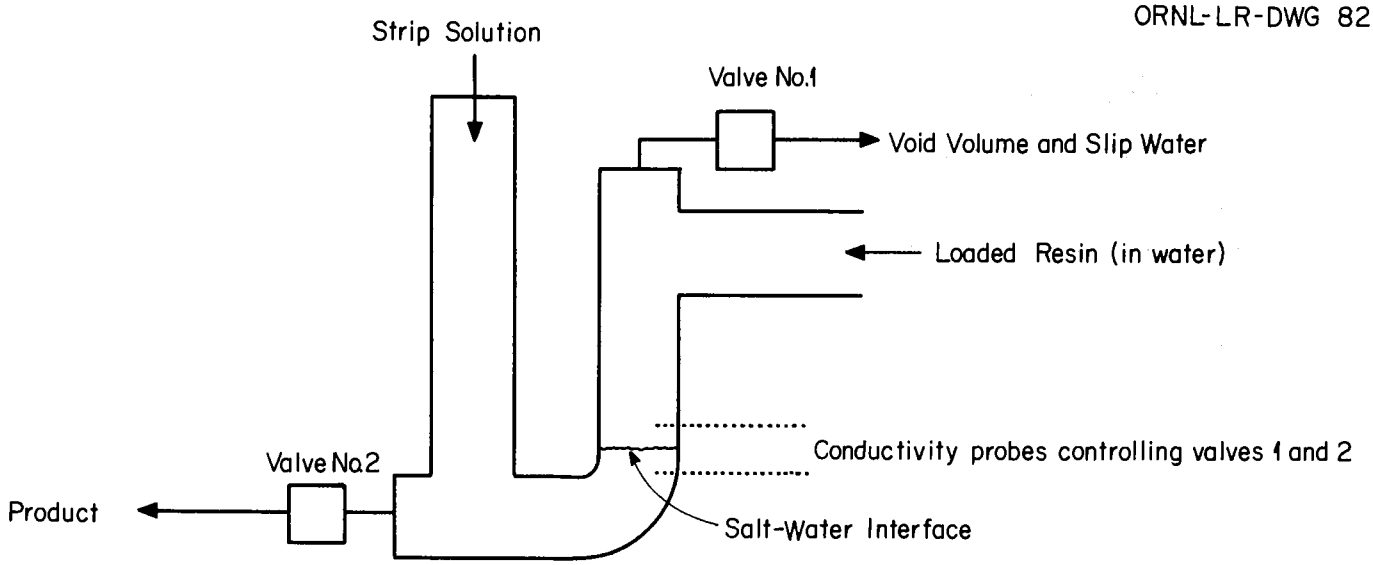
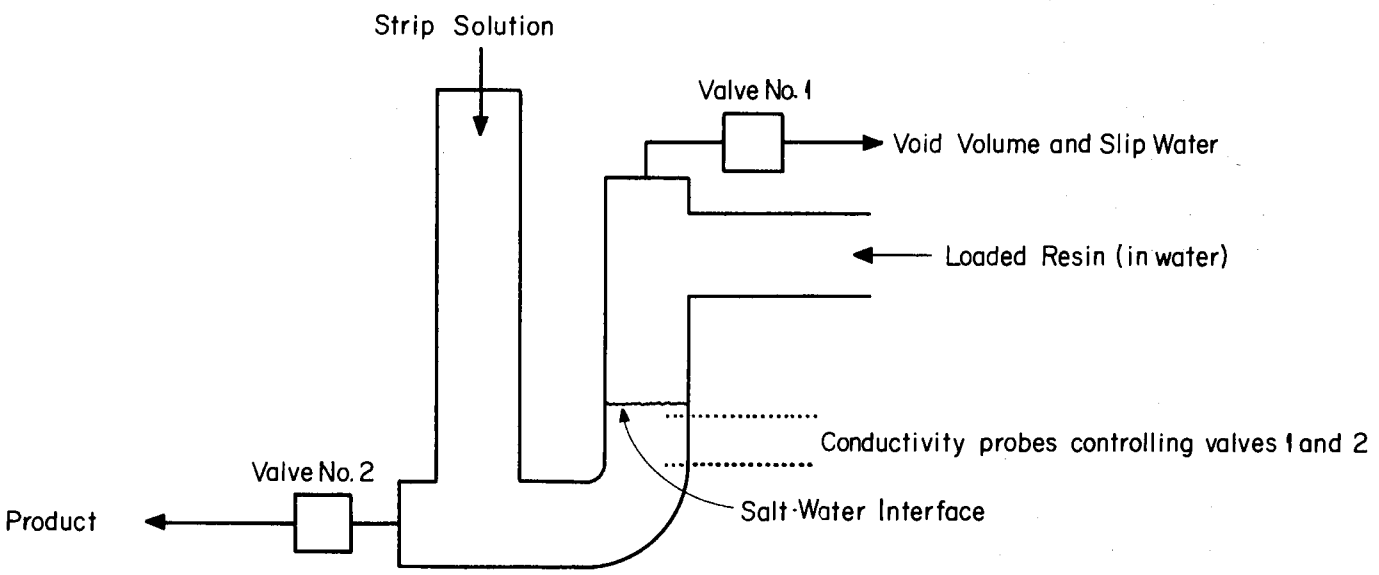


Just after resin movement; valve 1 open, valve 2 closed



Later, valve 1 closed, valve 2 open

Fig. 7. Conductivity Type Water Eliminator.

7.1 Laboratory-Size Contactors

Glass pipe is very convenient and provides easy observation of resin movement and color changes in laboratory-size columns. If short pipe sections separated by thick (1/4-in.) rubber or neoprene gaskets are used, samples can be taken from the flowing stream with hypodermic needles inserted through the gaskets.

The resin path should be as streamlined as practical. The smaller the column diameter, the higher the relative friction of resin flow. Plug valves should be as large as the column, at least up through 1-in.-diameter columns, to decrease friction. One-inch plug valves are relatively inexpensive. Small air or hydraulic pistons can easily be used to drive valves. When the columns are made of glass the resin valves must be mounted rigidly to a frame so that the glass pipe will not be under strain. This is not a problem with an all-metal column.

It is desirable to have low-voltage solenoid and pilot valves to simplify wiring and reduce the hazard of electric shock in situations where conducting solutions are frequently spilled.

Plastic which can be welded was found to be a convenient construction material for making special distributors and connections. It has the advantage of being easy to work and more corrosion resistant than metals.

7.2 Scale-Up Features

As the contactor is scaled-up, several features that were insignificant on the laboratory scale become important. In general, the variety of accessories is not so great for the larger contactor and the best suited type of accessory depends on the particular application. Since less pressure drop is required to move the resin in the larger contactors, a number of economies can be achieved in sizing connecting pipes and valves and in choosing distributors as outlined below.

The liquid pocket type of feed distributor may be used, but the liquid in the pocket must be kept homogeneous by agitation or manifold distribution of liquid. The tee type distributor scales up most easily. On scale-up, the pipe type gains in efficiency as compared to the bell type because no expanded section is required and its pressure drop-to-resin flow ratio decreases. The features of the two may be combined by passing the resin both through and around the liquid pocket. The pipe type has the advantage that resin and liquid are mixed more efficiently; the bell type has the advantage that solutions mix more intimately.

For very large columns the side take-off distributors will waste column height. Bottom take-off with center introduction of resin would permit simpler construction and reduce channeling tendencies because the solution does not change its direction of flow (Fig. 4, type c). As far as resin flow is concerned, the contactor resin path should be scaled up proportionally. For very large exchange sections it is undesirable to have correspondingly large connecting pipe. In this case the size of the connecting pipe is reduced to the point where resin channeling becomes prohibitive (see Fig. 8).

The upper limit on practical resin valve size is related to the connecting pipe size, not the exchange section size. The plug valve may be the same size as the connecting pipe or reduced in diameter until the resin pump pressure is prohibitive or the resin movement is seriously retarded (see Fig. 8).

As the contactor size increases it becomes more desirable to use the rotary type resin pump, rather than the reciprocating, because a greater capacity is available in a smaller piece of equipment.

Pumps can be made of corrosion-resistant material or of the cheaper mild steel. Pump corrosion and wear can be decreased by using oil as a displacement fluid and maintaining an oil-water interface in a reservoir. In this case only oil comes in contact with the pump.

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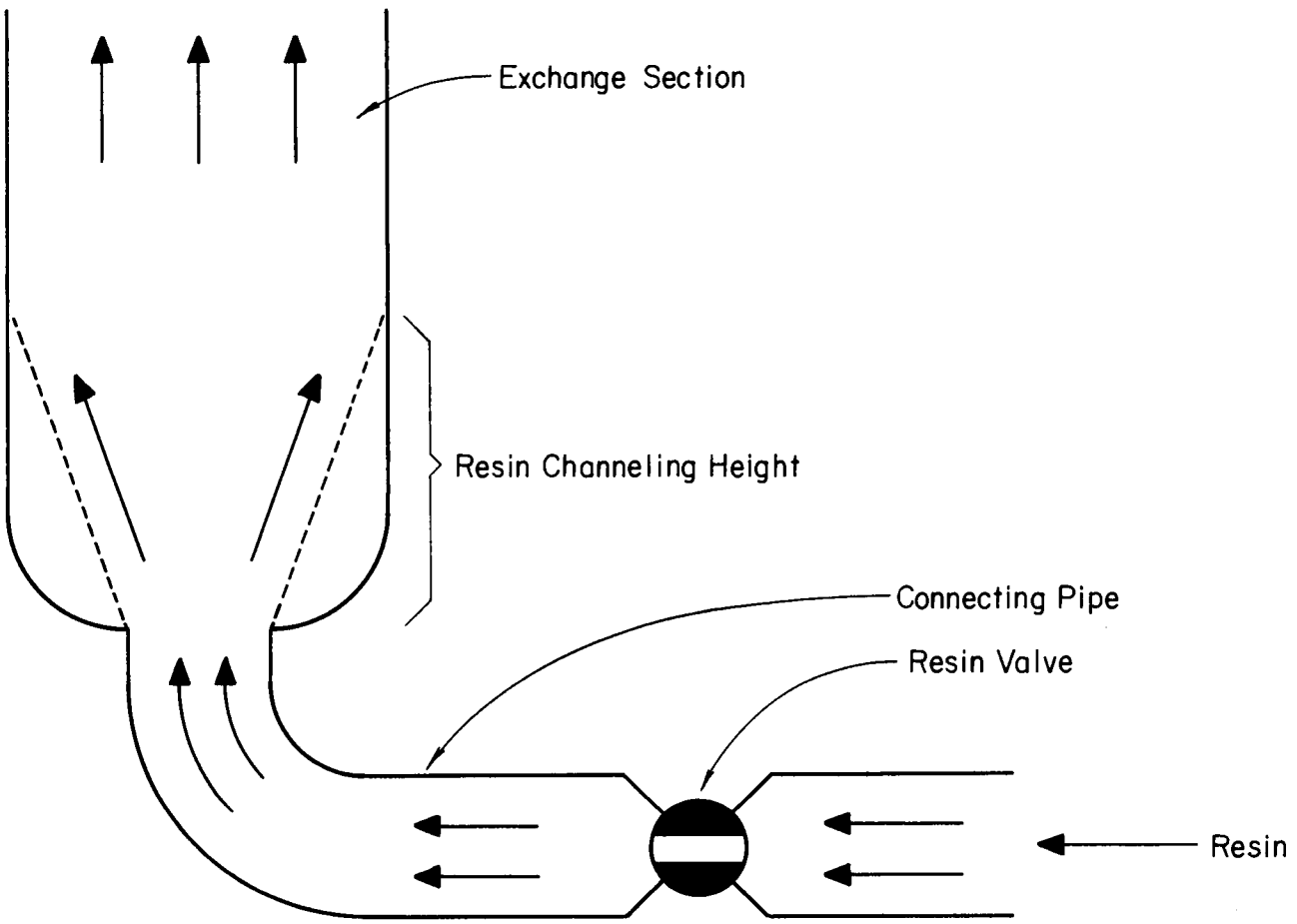

Resin channeling is proportional to $\frac{\text{Exchange Section}}{\text{Connecting Pipe}}$

Plug valve size is governed by permissible resin pump pressure and permissible slippage during resin movement

Fig. 8. Scale-Up Considerations of Connecting Pipe and Resin Valves.

8.0 PHOTOGRAPHS OF VARIOUS EXPERIMENTAL MODELS

A group of photographs of various experimental models is included to indicate the general appearance of the equipment at different stages of the development work.

The first contactor studied, a hand-operated model, is shown in Fig. 9. The column diameter was 0.5 in. Flow ratios were predetermined and solution flow was by gravity. The resin was moved by applying a vacuum to the top of the feed tanks. Among the experiments performed on this column were separation of barium and strontium, of sodium and potassium, of cobalt and nickel, and of uranium and cesium. The chemical efficiency of this model was greater than that of a fixed bed using the same amount of resin and the same chemical conditions.

Figures 10a and b are front and back views of the first completely automatic contactor, which had 3/4-in.-diameter columns. The two views were taken at different times, and the column arrangements differed slightly. The solution was pumped by a Sigma motor pump, the resin by a double bellows. Plug valves were opened and closed with a Bodine motor drive. A tee solution distributor was used in some of the runs with this model.

A later model, 3/4 in. in diameter, is shown partially installed in Fig. 11. Piston-operated valves, which were found to be much faster than motor-driven valves, and a piston resin pump were used in this model.

Figure 12 shows a 2-in.-diameter model in which the sorption and elution sections were placed side by side and connected by a smaller pipe. A tee distributor was used, in which a liquid pocket may be seen. The resin was moved by two double-acting pistons placed end to end with a stroke adjustment on the shaft. One piston acted as a hydraulic pump and the other as the air-motor drive. All the solution cut-off valves were air-operated diaphragm valves, ganged for simpler control by a pilot valve.

A 2-in.-diameter column in which a bell type distributor was used is shown in Fig. 13. The void and slip water eliminator is also shown. In the sorption section of this column, the solution flows upward and the resin downward rather than the reverse.

The bottom of a model in which a 6-in.-diameter sorption section and a 4-in.-diameter elution section were used is shown in Fig. 14a. Moyno and Eco solution pumps were used. The resin was moved with a mild steel gear pump. The top of the same model is shown in Fig. 14b.

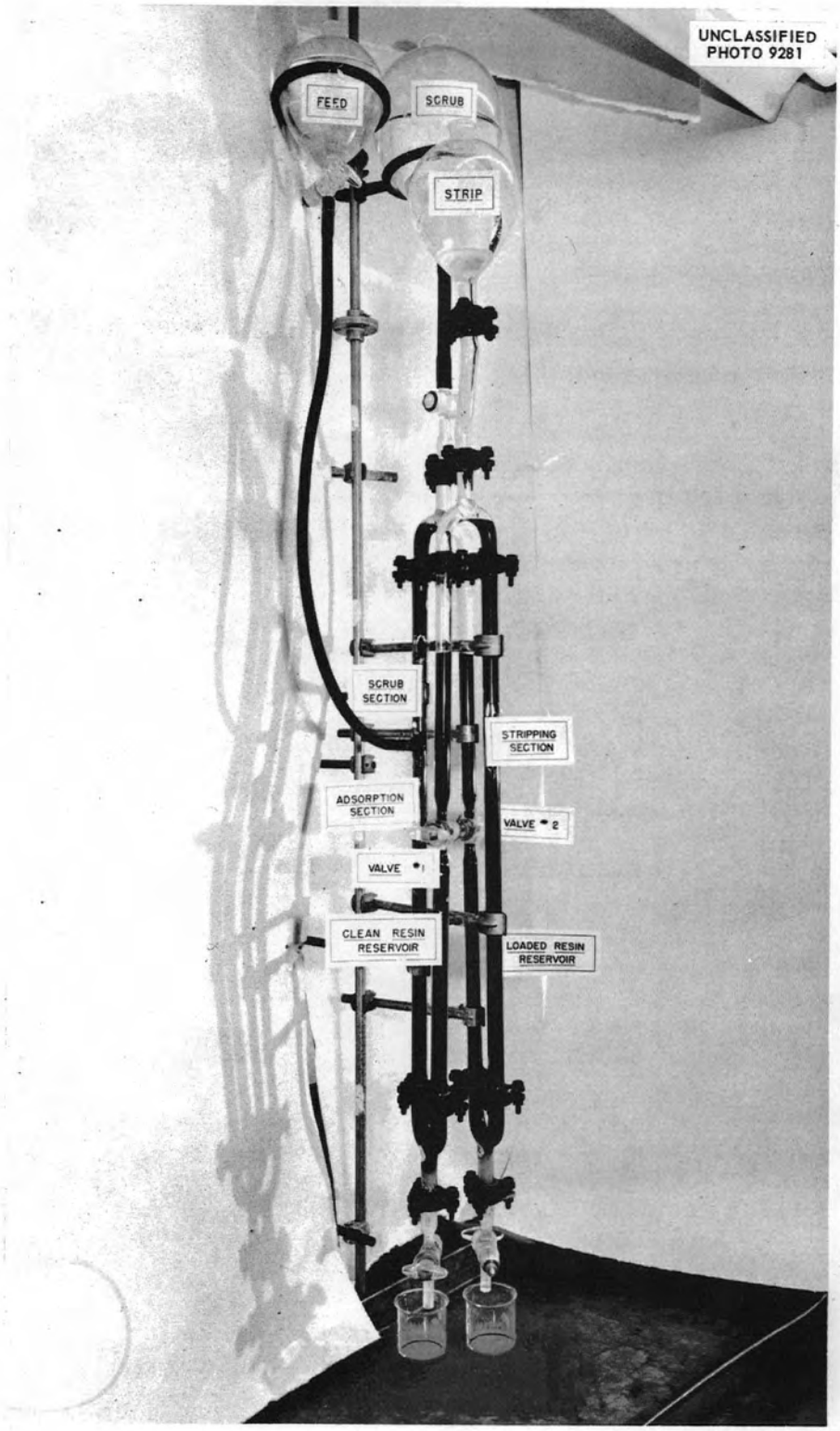

Fig. 9. First Contactor Built, a Hand-Operated Model with 0.5-in.-dia Columns.

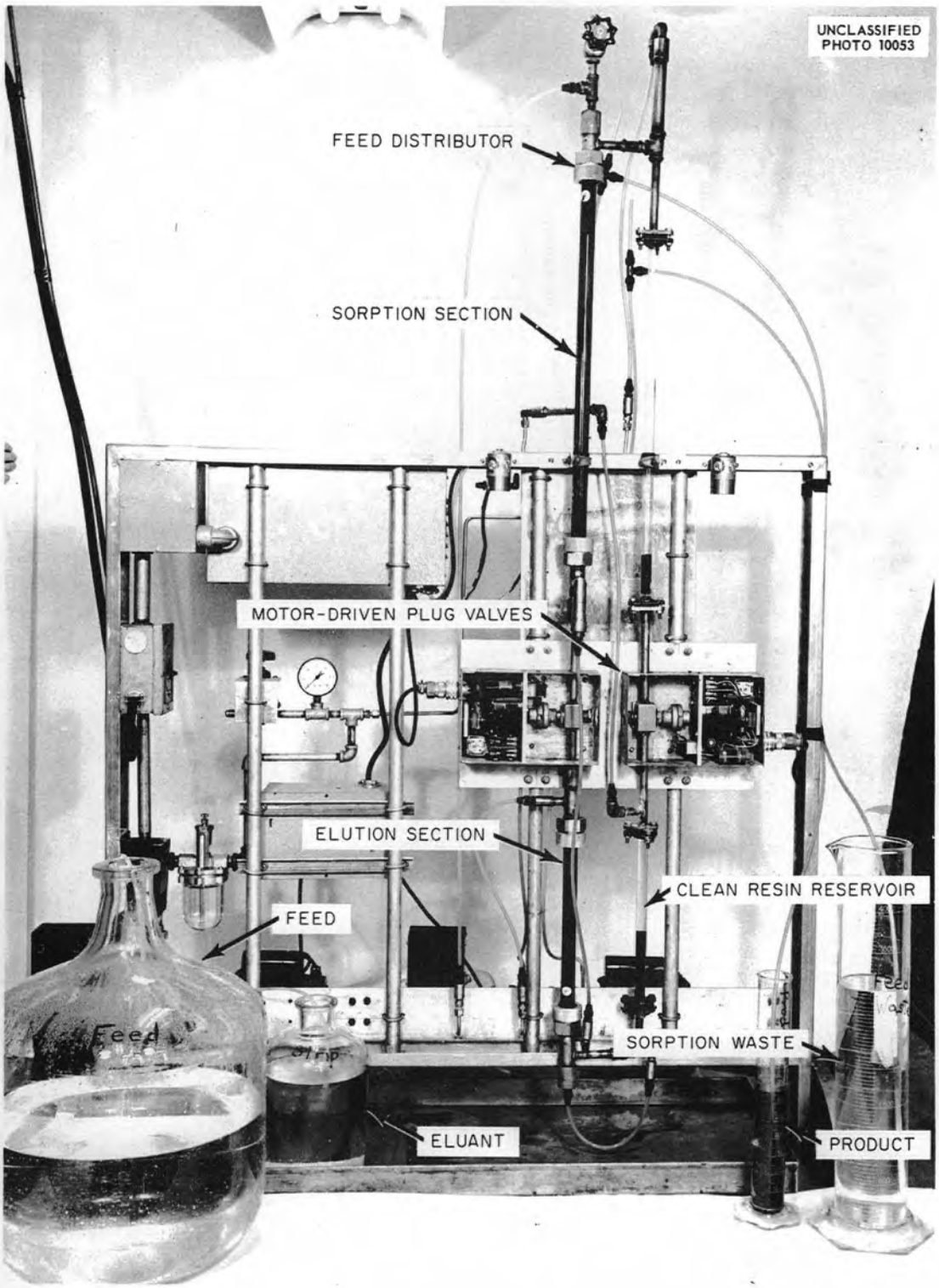

Fig. 10a. Front View of First Completely Automatic Contactor; Columns Were 3/4-in.-dia.

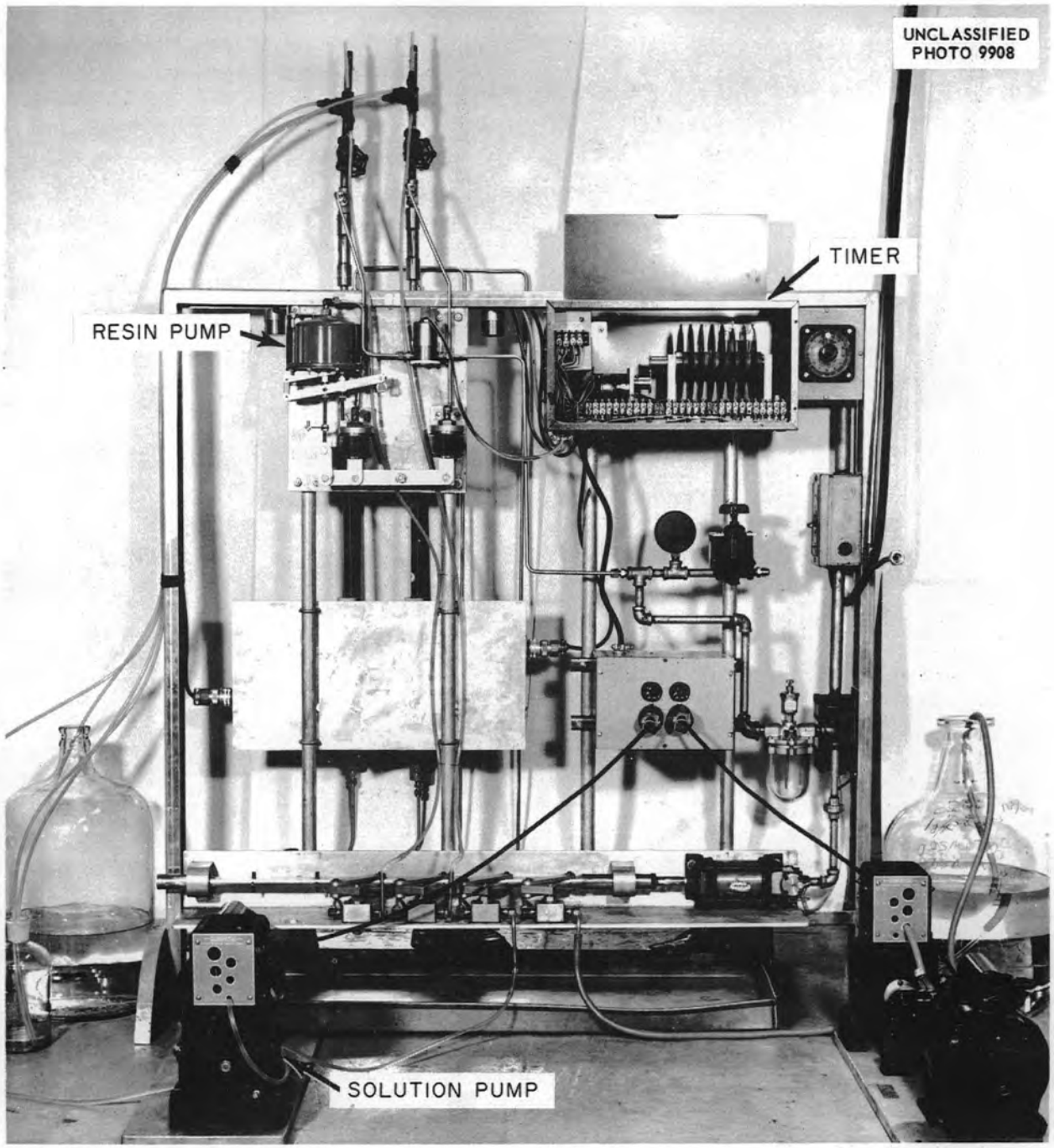

Fig. 10b. Back View of Contactor Shown in Fig. 10a (taken at a different time, with slightly different column arrangements).

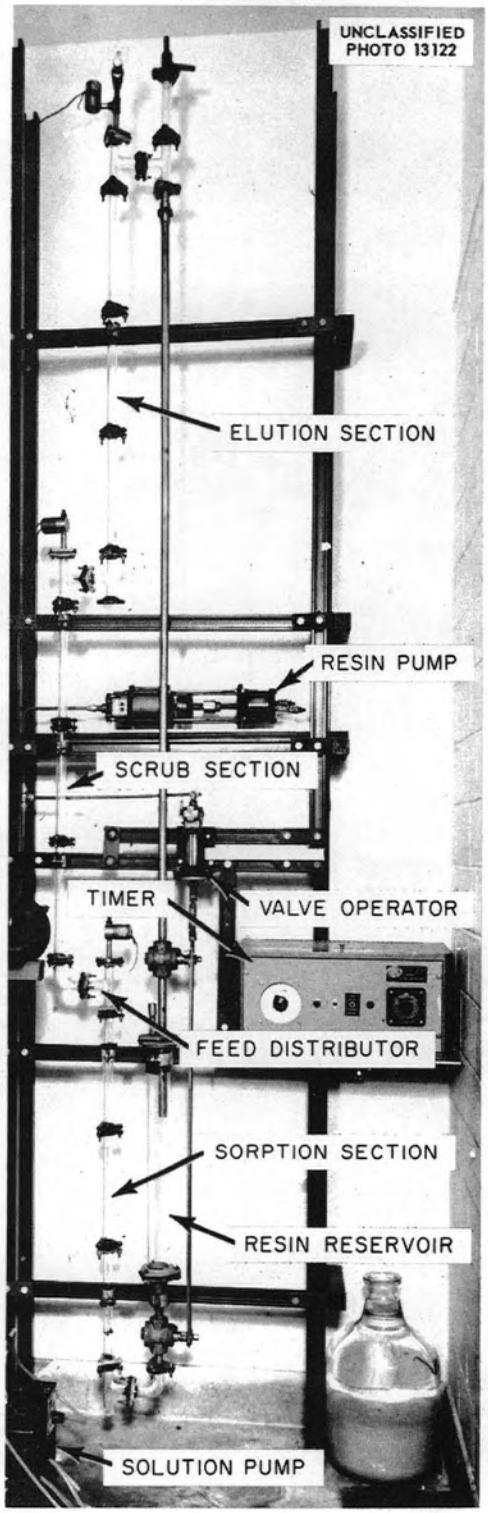

Fig. 11. Partially Installed 3/4-in.-dia Column Contactor.

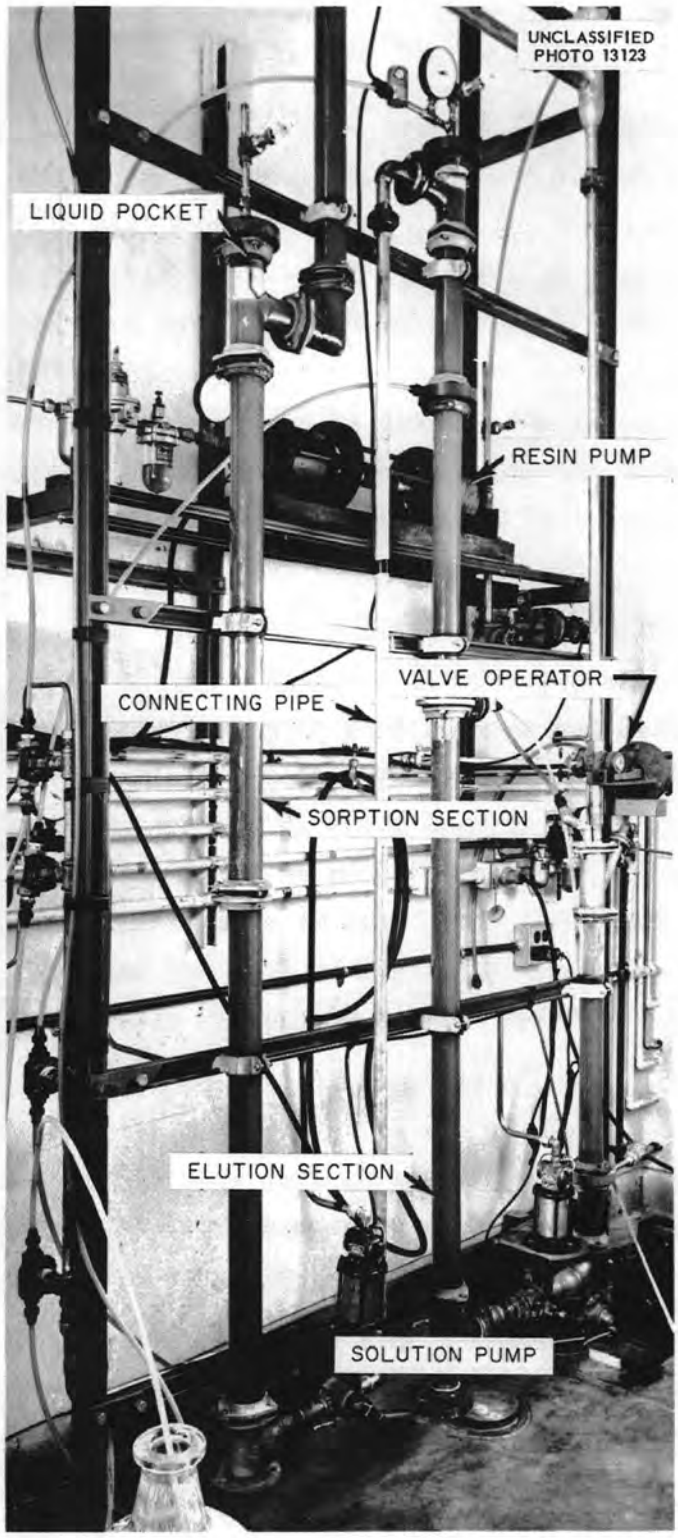

Fig. 12. Model in Which Sorption and Elution Sections of the Column (2-in.-dia) Were Placed Side by Side and Connected by a Smaller Pipe. Tee Type Solution Distributor Was Used.

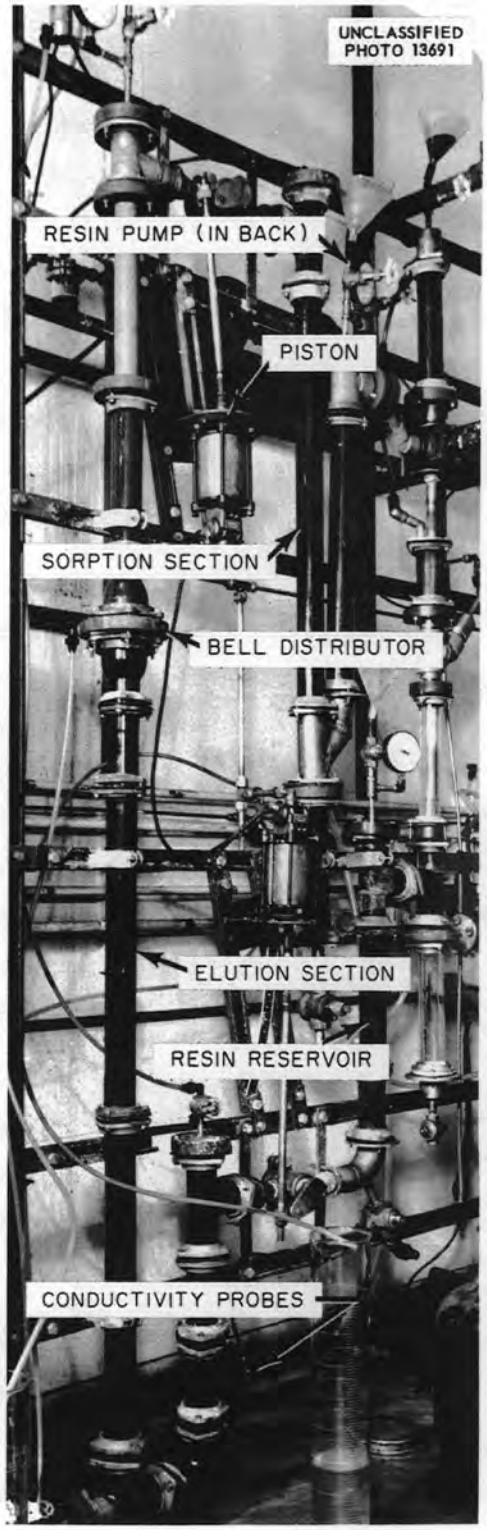

Fig. 13. Model, 2-in.-dia Columns, in Which a Bell-Type Distributor Was Used.

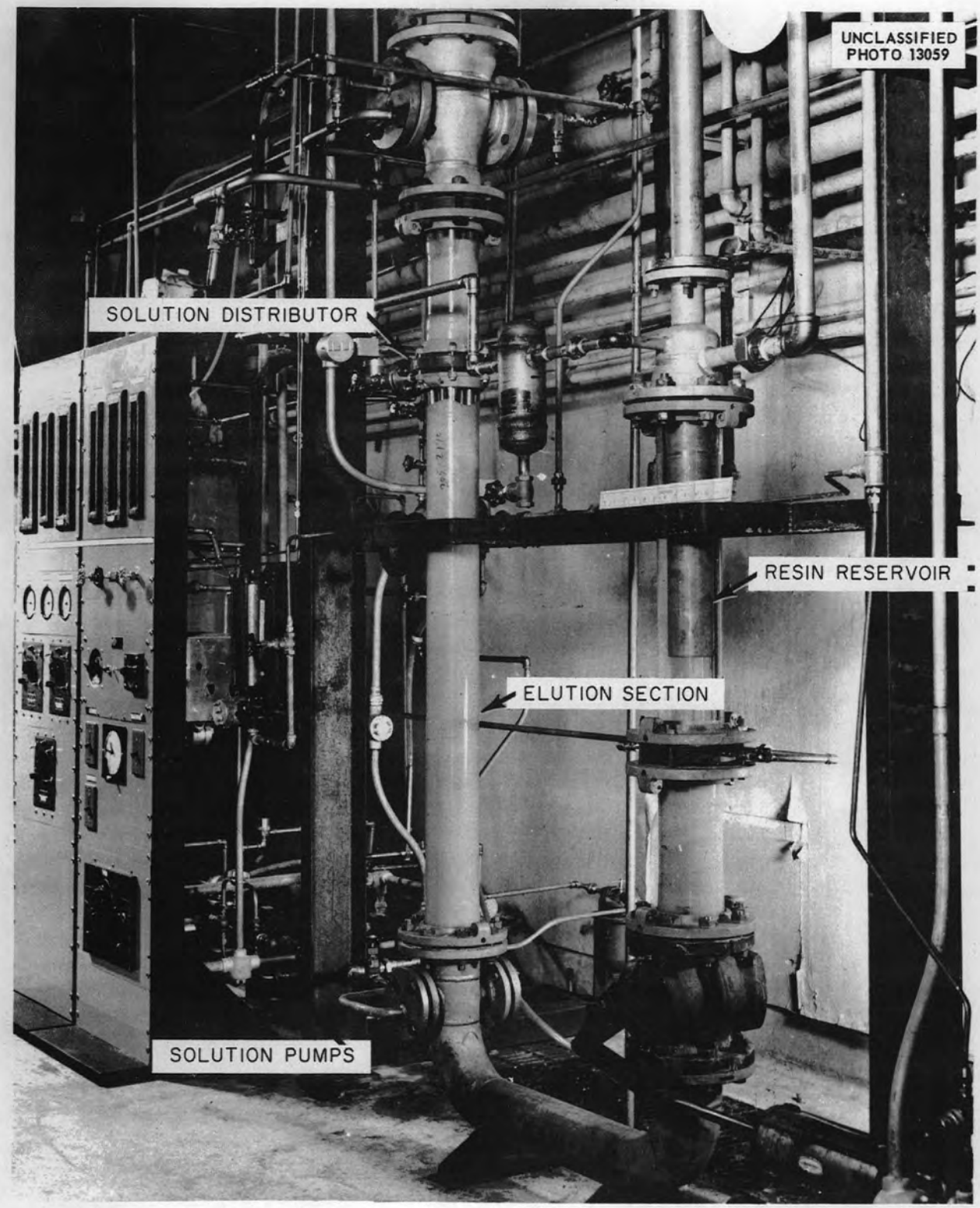

Fig. 14a. Bottom Portion of Model with 6-in.-dia Sorption Section and 4-in.-dia Elution Section.

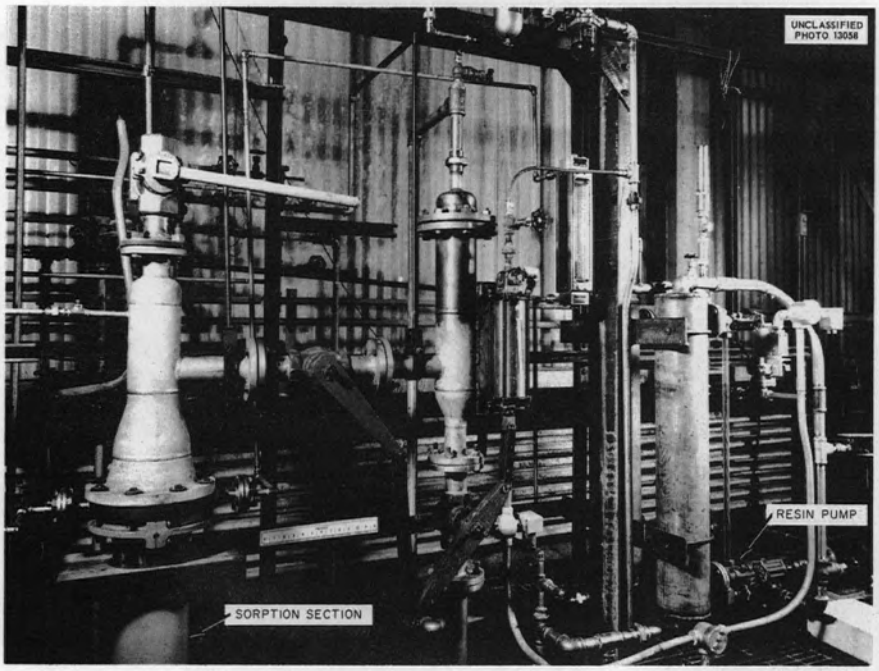

Fig. 14b. Top Portion of Model Shown in Fig. 14a.