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**M. D. Thaxter, H. P. Cantelow, and C. Burk**

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## OFF-GAS TREATMENT IN BERKELEY ENCLOSURES

by

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In the past ten years quantities of alpha-, beta-, gamma- and neutron-emitting isotopes handled in enclosures have increased from microcuries to kilocuries. Contaminated-atmosphere problems have increased accordingly. Developments at Berkeley are reviewed with particular emphasis on recent equipment, viz: a multiple-purpose gas scrubber and a total-capture system for slug-dissolver off gas.

## OFF-GAS TREATMENT IN BERKELEY ENCLOSURES

### A. Introduction

More than ten years ago it became apparent to astute radiochemical workers that personnel risks and area contaminations arising from bench and fume-hood processing of radioisotopes could be economically obviated by the use of total enclosures. An early application became known as the "Berkeley" enclosure. Some persons, unfamiliar with the versatility developed with these enclosures, even today erroneously think of them solely as dry boxes, or alpha boxes, or gloved boxes. Actually, of course, they are also routinely employed in multicurie wet chemical or physical operations. In either case remote manipulation, massive shielding, and other appurtenances may be required.

Throughout the development of the Berkeley enclosure an important but seldom emphasized aspect has been establishment of ventilation and air cleaning adequate both to the contained processes and to the radiological realities.

The first ventilation concept, basic and essentially simple, which runs throughout the various systems, is the maintenance of a continuous pressure differential such that the interior of the enclosure is not less than about 3/8 inch water gauge below atmospheric pressure. This is to prevent out-breathing of activity. It is readily accomplished by the installation of a suitable exhaustor at the off-gas stack (preferably at the terminus).

The second concept, or goal, which has never been nor will be 100% satisfactory, is decontamination of the exhaust prior to its discharge to the atmosphere. As a logical corollary, the search must go forward for an acceptable ultimate disposal not involving discharge to the atmosphere.

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This paper illustrates two recent applications of developmental work in air cleaning and ultimate disposal of off gases at Berkeley against a backdrop of past practice.

## B. Chronological Development of Off-Gas Treatment in Berkeley Enclosures

The early Berkeley enclosures, about 1947, were essentially gloved boxes in which processed activity seldom exceeded a millicurie. Ventilation and air cleaning were simple, viz: a crude air-inlet filter of the air-conditioning-equipment type, an exhaust filter of laboratory glass wool, and an exhauster discharging to air via a convenient fume hood. The air turnover was about 20 seconds, for example 15 cubic feet per minute through a 5-cubic-foot enclosure. Air and swipe samples occasionally revealed activity downstream. In 1949 the exhaust filter was replaced with a CWS 6 unit, resulting in higher efficiency for particulate decontamination at a cost of less throughput. Increasing levels of activity led to higher enclosure contaminations. A close-capture fume hood was installed and the air path rerouted through a more efficient entrance filter. The employment of stronger reagents threatened the CWS 6 filter, and an exhaust prefilter was added. All these reduced the throughput, thus dictating an exhauster of higher capacity to reestablish the 20-second turnover rate. By 1953 we thus had the essentially simple system shown in Fig. 1, which may be termed a high-throughput ventilated enclosure. These enclosures are in current use and deemed useful, in good hands, to house activity levels to about 10 millicuries. Exceptional operators can stretch this to 0.1 curie with some risk to contamination control.

Before 1953 processing requirements had risen to substantially more than a curie per enclosure in several instances. Some of these Berkeley enclosures were operating behind shielding walls with remote tong manipulation. Evidence of unacceptable levels of radioactive material downstream from CWS filters came to hand. Much of this was particulate in nature. It appeared we had passed a practical safe operating limit for general chemical work under high-throughput ventilation design, in view of the finite efficiency of the CWS 6 filter as an air cleaner.

We elected to revise the ventilation design -- to get higher cleaning efficiency -- by markedly reducing the flow rate of the contaminated off gases. This would enhance the capture efficiency of the cleanup filter by increasing the residency time of the contaminants, thus permitting Brownian motion to bring about contact between particle and filter. For this purpose our basic enclosure was designed substantially leaktight. A number of problems immediately arose because of the lack of throughput air. For example, heat transfer became very poor -- requiring refrigeration; humidity became high -- requiring dewatering; gaseous reactants formed smokes and impaired visibility -- requiring specialized removal. In recognition of these, our low-leak system was designed to have two major attributes: (1) a recirculation system operating at 15 cfm, continuously processing the enclosure atmosphere; and (2) an exhaust system cleaning the total net off gases (i. e., the leak plus the product gases) at 0.02 cfm. Figure 2 illustrates a typical setup wherein three enclosures behind shielding at the right are served by three recirculating units, themselves enclosed, outside the shield at the left.

It must honestly be stated that this is not a trouble-free system, it is not cheap, and it requires supervision. But it is an improvement, and we can give a few illustrative numbers. A 1956 operation involved the chemical reduction of ten pile-irradiated slugs, containing about  $2 \times 10^{15}$  dpm of alpha emitters associated with more than 1000 curies of beta-gamma emitters requiring more than 6 inches of lead shielding. The total off gas, including leaks, was 0.02 cfm. The process air-cleaning equipment included a condenser, a refrigerated scrubber, a roughing filter, and two layers of final prefilters of fine glass, and terminated in two CWS #6 filters in series. Downstream from these were two continuously operating membrane filters, in series, through each of which the entire off gas was drawn. The original purpose of these was to sample the effluent, with the hope they would also afford additional decontamination. The hope was realized when, during one period, the first membrane filter picked up  $1 \times 10^8$  dpm of alpha emitters in less than 24 hours. The second such unit showed 200 dpm. Parenthetically we observe these membrane filters were usable because the off-gas volume was small.

We are convinced that had this low-leak system not existed for this operation, substantially more than  $10^8$  dpm of alpha emitters would have gone out the stack daily.

### C. Recent Equipment Developments

Having now gone once over lightly the trends in off-gas handling in Berkeley enclosures I have the pleasure to report two most ingenious developments by my colleagues, H. P. Cantelow and C. Burk.

Earlier I mentioned the many problems that arose when the low-leak design basis was adopted. Prominent among these problems were the circulating acid gases, enhanced humidity, and smoke formation. H. P. Cantelow's solution to these follows:

The low-leak system used at UCRL for handling the off gas from high-level operations in Berkeley enclosures is shown in Fig. 3. Since the only source of fresh air for the system is from leaks -- about 0.02 cfm -- a recirculating loop is provided for controlling the atmosphere in each enclosure. Within this loop is a refrigerated scrubber, which prevents corrosion by removing acid vapors and excessive moisture and prevents smoke by also removing ammonia. Fifteen cfm of air withdrawn from the 30-ft<sup>3</sup> enclosure is thus treated and returned. The scrubber consists of a wetted saran fiber bed housed in a lucite box, as shown in Fig. 4.

The selection of a scrubber liquor to perform all three of these functions presented a problem. A simple caustic solution was quite unsatisfactory, since it was ineffective in removing ammonia. In experiments with a trial enclosure no visible smoke was generated if the air concentrations of both ammonia and hydrochloric acid were less than 10 parts per million. It was estimated that in a typical run the scrubber would be called upon to remove about 1 mole of hydrochloric acid and 0.1 mole of ammonia per liter of scrubber liquor. Vapor pressure curves show that adequate removal of ammonia may be obtained if the scrubber liquor pH is maintained below 7. (See Fig. 5.) For control of acid corrosion a minimum pH of 2 was arbitrarily set.

The possibility of using a buffered scrubber liquor appeared to be a promising course to pursue. A phosphate solution was chosen because of buffer action within the desired range and good solubility characteristics. Figure 6 shows the titration curve of a 1.0-molar phosphate solution. A solution containing 1 mole monobasic potassium phosphate and 0.5 mole sodium hydroxide per liter was selected for general use. This solution has a pH of 6.3 and has a "capacity" to absorb 0.25 mole of  $\text{NH}_3$  or 0.7 mole of  $\text{HCl}$  per liter without exceeding the desired pH limits, and thus satisfactorily met the requirements. On occasions when a greater proportion of ammonia might be expected, as in the caustic-nitrate dissolution of aluminum slugs, less sodium hydroxide is added to the buffer solution.

If the pH reaches the limit of 2 during a run, caustic may be added to the solution to bring the pH back up to 6. This can usually be done twice without excessive precipitation of solids. After that the scrubber solution must be replaced. In our experience a single charge of 17 liters of scrubber liquor has been sufficient to last for a 2- to 3-month run.

Humidity control has been simply and effectively obtained by cooling the scrubbing liquor. The liquor reservoir is maintained at  $8^\circ$ - $10^\circ$  C by circulating water at  $4^\circ$  C through a small stainless steel coil in the scrubber reservoir. This has proved adequate to prevent condensation on surfaces and to eliminate any fogging of the windows.

This multiple-purpose scrubber system has been successfully used for ten different shielded Berkeley enclosures during the past year and a half. Control of corrosion and humidity has been quite adequate and no smoke problems have been encountered, even though smoke constituted a serious hazard in a prior run of similar nature in which the scrubber liquor was a caustic solution. There have been no operational difficulties, nor have there been any failures of materials or equipment. On some occasions a build-up of solids on the face of the scrubber wet pad has occurred, but not to the extent of obstructing the flow of air through the scrubber. The system has required very little attention other than an occasional check of the pH of the scrubbing liquor.

The other development we wish to report is our initial assemblage, demonstrated through pilot runs, for total capture of radioactive gases (as well as liquid and solid contaminant aerosols) arising from the dissolving of pile-irradiated slugs. Mr. Burk's report may, we hope, forecast the ultimate goal of elimination of an exhaust stack discharging pollutants to the air.

Our first approach to the problem of controlling the radioactive gases produced during the dissolution of a pile-irradiated slug was to consider the use of a charcoal trap on the exhaust line of the chemistry enclosure. A literature search was made (see bibliography), and charcoal traps were built based on a composite of various reports. The traps were copper U tubes, 2 inches in diameter, containing about 2 lb of Cenco C-1543 charcoal in a bed 30 inches long. The charcoal was activated by evacuation for several hours while being heated with a 100-watt tape heater. The traps were incorporated into the exhaust system of a group of enclosures housing the equipment used for the dissolution and chemical processing of some pile-irradiated transuranic slugs. One trap removed about 90% of the gaseous activity from the exhaust



air from the dissolution of 5 slugs. The activity was measured by ion chambers before and after the trap. The rare gas concentrations were about  $10^{-4}\%$  Kr and  $5 \times 10^{-6}\%$  Xe. The net flow rate was about 0.013 cfm in the trap. Changes of pressure in the system--e. g., due to tong operation--made large changes in the instantaneous flow rate, or even reversed the direction of flow.

We felt that there were several serious disadvantages to using a charcoal trap in this way. In view of the trend toward higher activity levels, the escape of 10% of the noble gas activity might soon be intolerable. Constant watch must be kept on the charcoal trap to avoid breakthrough, especially if the rate of gas production varies. Under shutdown conditions, the gaseous activity tends to equalize throughout the trap by desorption and readsorption, and eventually come out of the exhaust end. In view of these faults, and the undesirability of contaminating the box atmosphere, it was decided to develop a method of sequestering off gases before they could mix with the atmosphere of the box.

Since the slug-dissolution process consists of dissolving the aluminum slug in a caustic nitrate solution, the gas of greatest volume is ammonia, associated with a variable amount of water vapor released by the heat of reaction. The amount of fission gases is a negligible percentage of the volume of the products of dissolution. Calculations indicate that more than 98% of the Kr and Xe are in the gaseous phase and thus can be removed.

Tests were made to determine the adsorptive capacity of charcoal for air. A chamber containing charcoal was evacuated for periods of from 1 to 48 hours, with and without heating to various temperatures up to  $600^{\circ}$  C. The chamber was then connected to a rotameter, and air was allowed to enter at various rates. Activated coconut charcoals such as Cenco-1543, Fischer 5-685, or BC Adsorbite, evacuated at room temperature, would adsorb seven to eight times their nominal volume of air. When the evacuated chamber was at  $-77^{\circ}$  C the charcoal would adsorb 40 to 45 times its nominal volume of air. The adsorptive capacity was not appreciably affected by heating during evacuation. A sample of wood charcoal tested at room temperature would adsorb only four to 4.5 times its nominal volume of air.

An experimental dissolution apparatus was made, consisting of a Pyrex reaction vessel cooled by water in an immersion coil and an air-cooled reflux condenser connected to an evacuated charcoal chamber through a needle valve. Sufficient water cooling was used to keep the reaction rate low enough so that the gas produced would not cause foaming and production of mist. Near the end of the dissolution, hot water was put through the coil in order to dissolve the last bit of the aluminum. The negative pressure over the liquid was at first manually regulated by the needle valve to about 5 inches of mercury. Since the rate of gas evolution varied widely, it was impracticable to use manual control. A Cartesian manostat was installed, and was capable of holding the desired negative pressure to within 0.25 inch of mercury.

Since a vessel with a cooling coil would offer difficulties in recovery of very small amounts of valuable product, it was decided to use a water jacket for temperature control. The reaction rate is approximately exponential with temperature, therefore in order to have a reasonably fast reaction rate, it is necessary to have an efficient, low-lag cooling system. This was accomplished by use of a low-volume water jacket with a tangential inlet.

A complete pilot-plant assembly, diagrammed in Fig. 7, was made and operated with nonradioactive dummy slugs. It consisted of a covered reaction vessel of about 1.5 liter total capacity and with a water jacket in which the water level is at the level of the liquid in the vessel, an air-cooled reflux condenser, a laboratory Cartesian manostat, an evacuated 800-ml container of activated charcoal, and a heater for the jacket water. Instrumentation consisted of a thermometer in the solution, a manometer on the reaction vessel, and a compound gauge on the charcoal vessel. The reaction vessel had a coned bottom for ease in removing the contents at the end of the run. The charcoal was activated by evacuation for 2 to 4 hours at room temperature. During the run the charcoal vessel was maintained at  $-77^{\circ}\text{C}$  by a dry ice-n-butanol bath.

One cycle consisted of the following operations: The 20-gram slug of 2S aluminum was placed in 350 ml of 3.6 M NaOH and 2.6 M  $\text{NaNO}_3$ . The vessel was covered, the valve to the evacuated charcoal vessel was opened, and the vacuum in the dissolver adjusted to 5 inches of mercury by means of the manostat. The exothermic reaction gradually raised the temperature of the solution. At  $60^{\circ}$  to  $70^{\circ}\text{C}$  the cooling water was started, and the reaction temperature was controlled at  $80^{\circ}$  to  $90^{\circ}$ . When the evolution of gas stopped, or the solution temperature dropped below  $75^{\circ}$ , the jacket water was heated to raise the solution temperature to  $90^{\circ}$ , dissolving any residual aluminum. A typical run is plotted in Fig. 8.

The gases from one slug reduced the vacuum in the charcoal vessel from 30 inches to about 27 inches of mercury. When the charcoal was warmed to room temperature, the remaining vacuum was about 15 inches of mercury. There is a large safety factor in the amount of charcoal used. Tests show that if the charcoal at  $-77^{\circ}\text{C}$  was allowed to come to equilibrium with air and the vessel was sealed, the pressure at room temperature would be about 150 psig.

Designs are now being made for a closed dissolver system for a run using 5-gram pile-irradiated slugs. This is a scale-down factor of  $1/4$ , and is expected to offer no difficulties. Only the dissolver vessel itself will be in the enclosure. All the accessory apparatus outside the enclosure will be either nonradioactive or at less than atmospheric pressure. A series of charcoal vessels, one for each slug plus some extras, will be manifolded together in such a way that they can be disconnected without the release of activity. The vessels of gas adsorbed on charcoal may then be disposed of as solid waste, or used as sources of radioactive gas.

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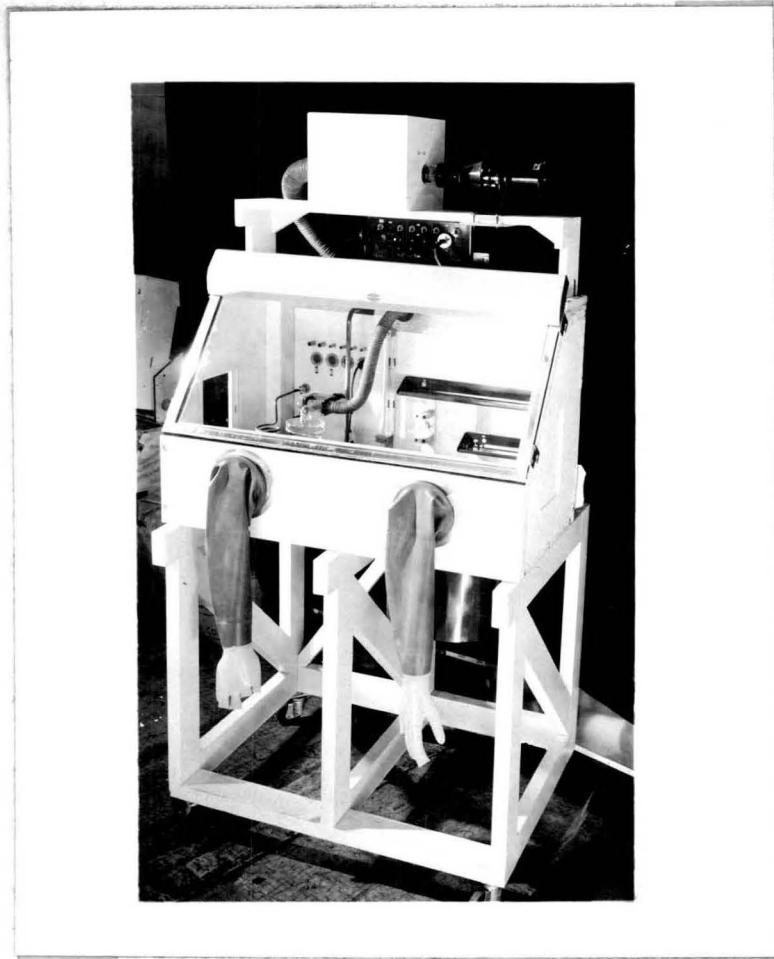


Fig. 1



Fig. 2

# OFF-GAS PROCESSING FLOW SHEET

EXHAUST SECTION

RECIRCULATING SECTION

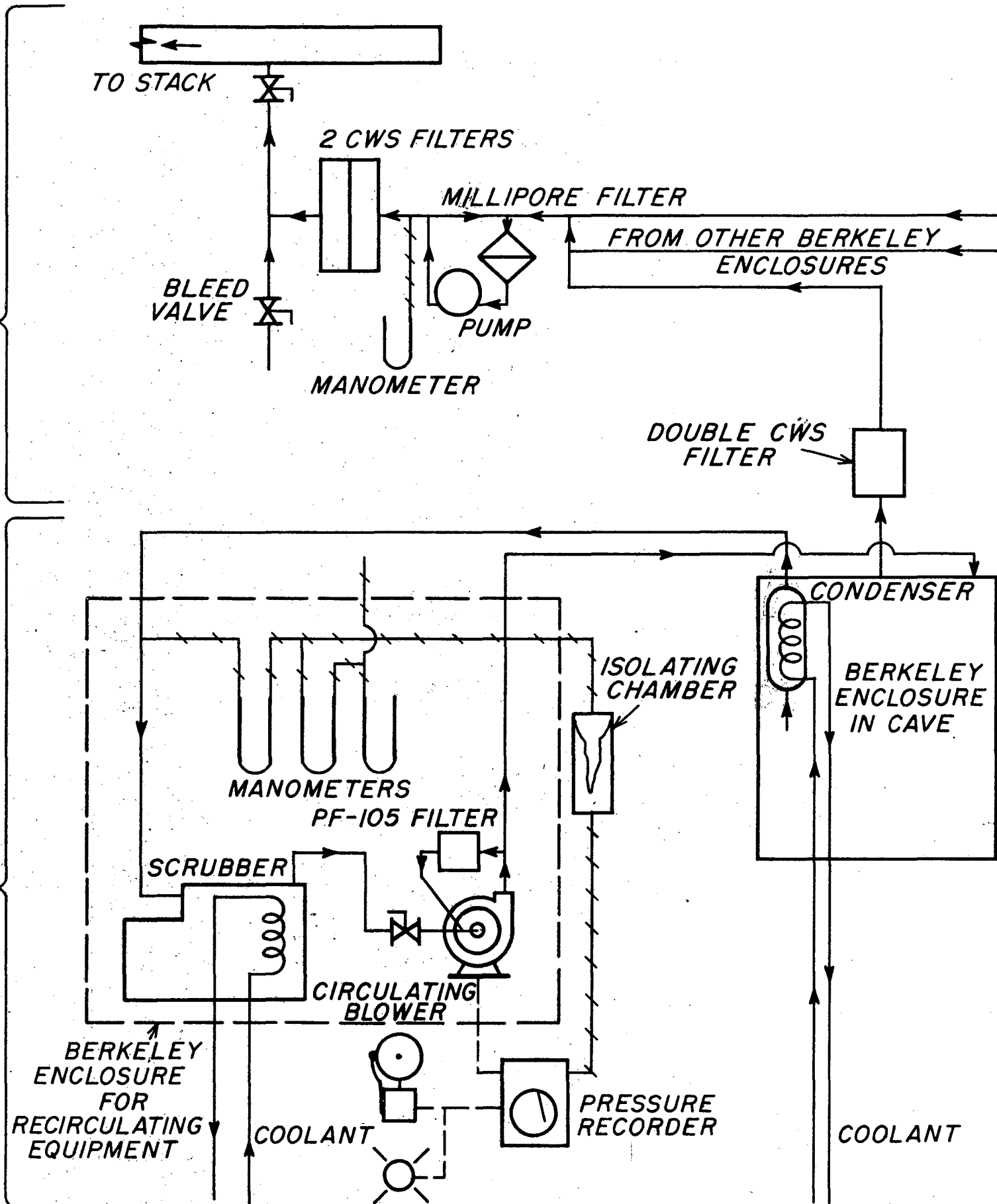


FIG.3

OFF - GAS PROCESSING EQUIPMENT  
SCRUBBER

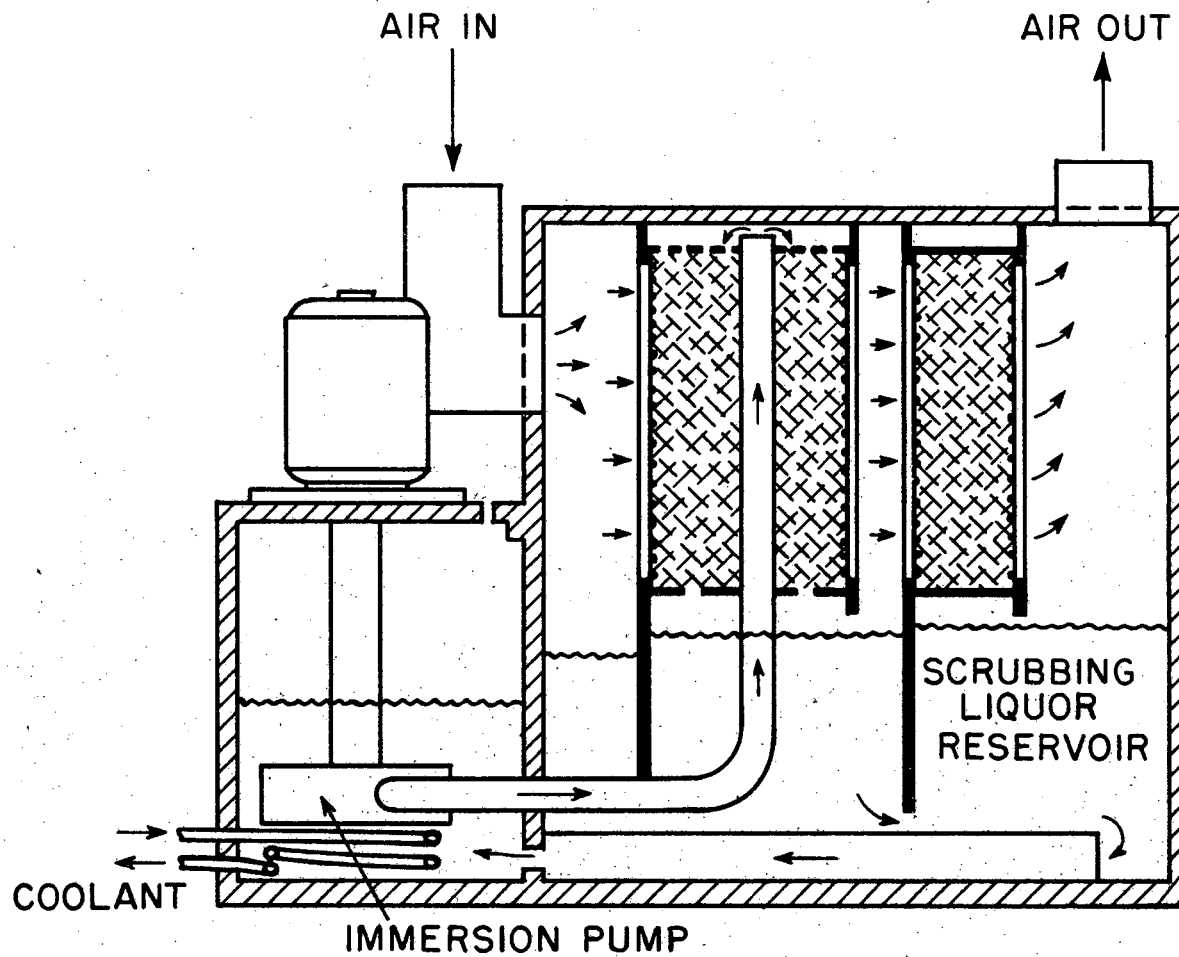


FIG. 4

59

# ESTIMATED HCl & NH<sub>3</sub> VAPOR PRESSURES AS A FUNCTION OF pH

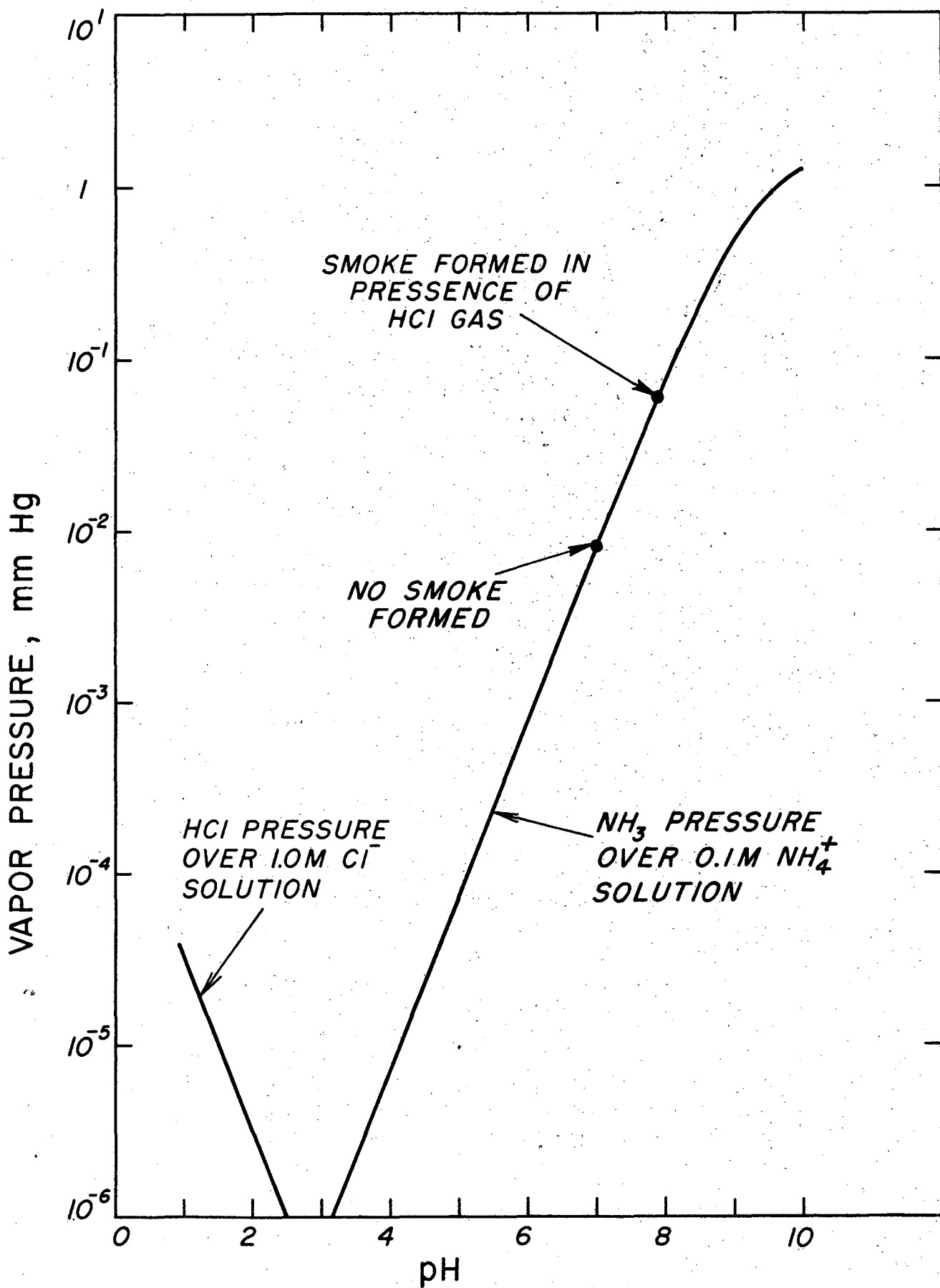


FIG. 5



OFF-GAS PROCESSING  
TITRATION CURVE OF PHOSPHATE SOLUTION  
(USED AS SCRUBBING LIQUOR)

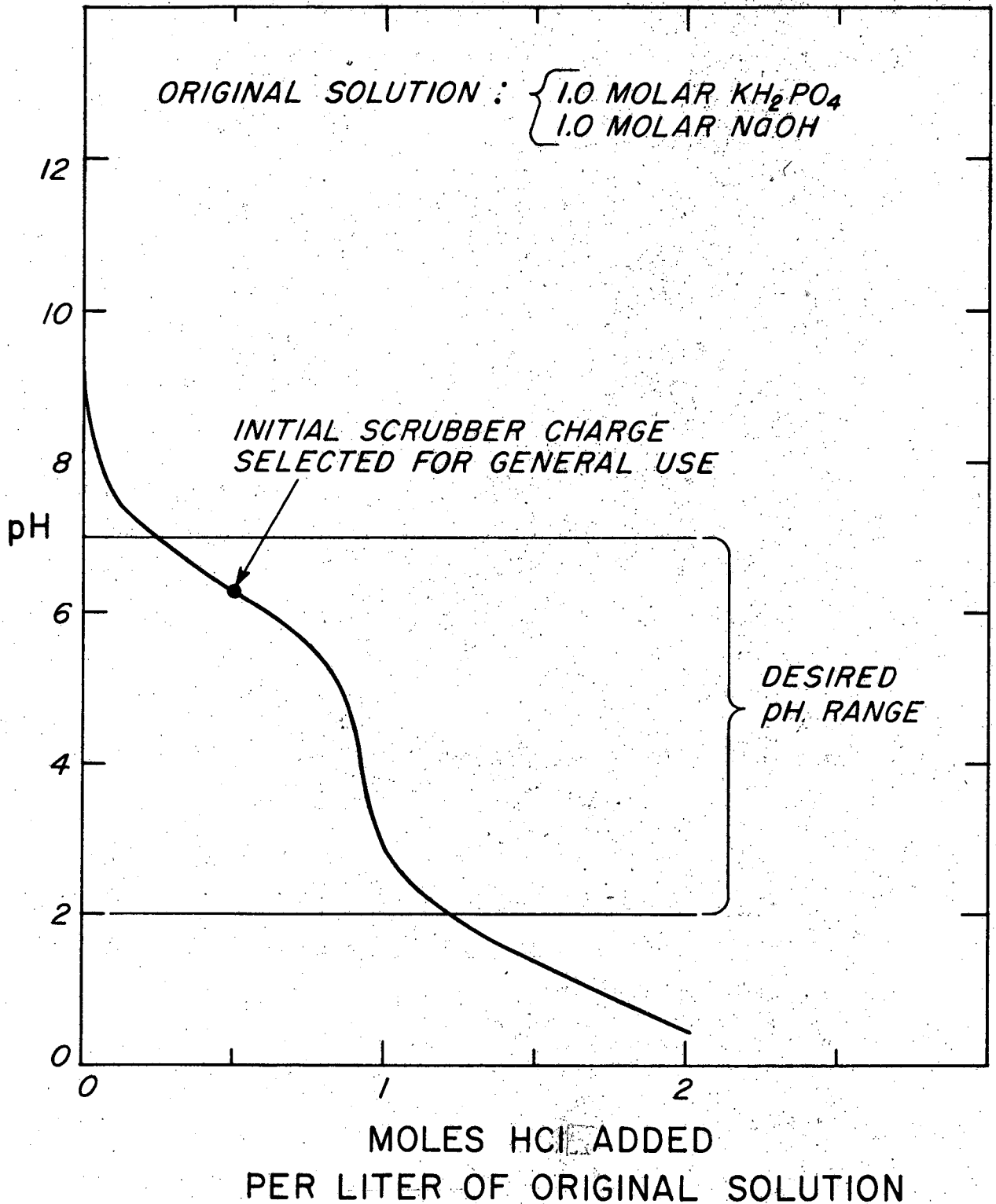


FIG. 6

DIAGRAM OF PILOT PLANT ASSEMBLY  
OF CLOSED DISSOLVER SYSTEM FOR DISSOLUTION  
OF ALUMINUM SLUGS

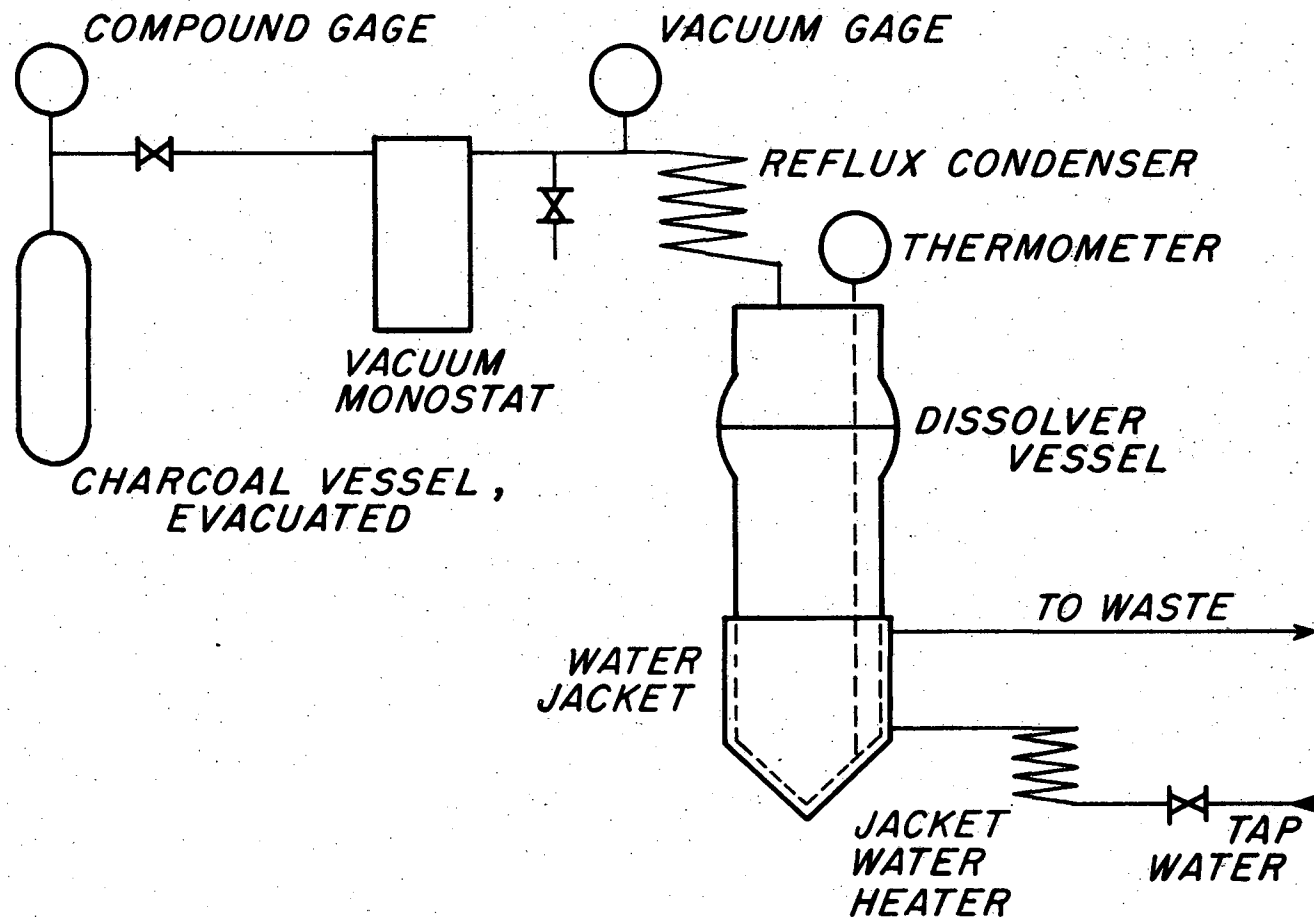


FIG. 7

TIME - TEMPERATURE CURVE OF DISSOLUTION  
OF 20g ALUMINUM SLUG IN 350ml  
NaOH - NaNO<sub>3</sub> SOLUTION IN CLOSED DISSOLVER

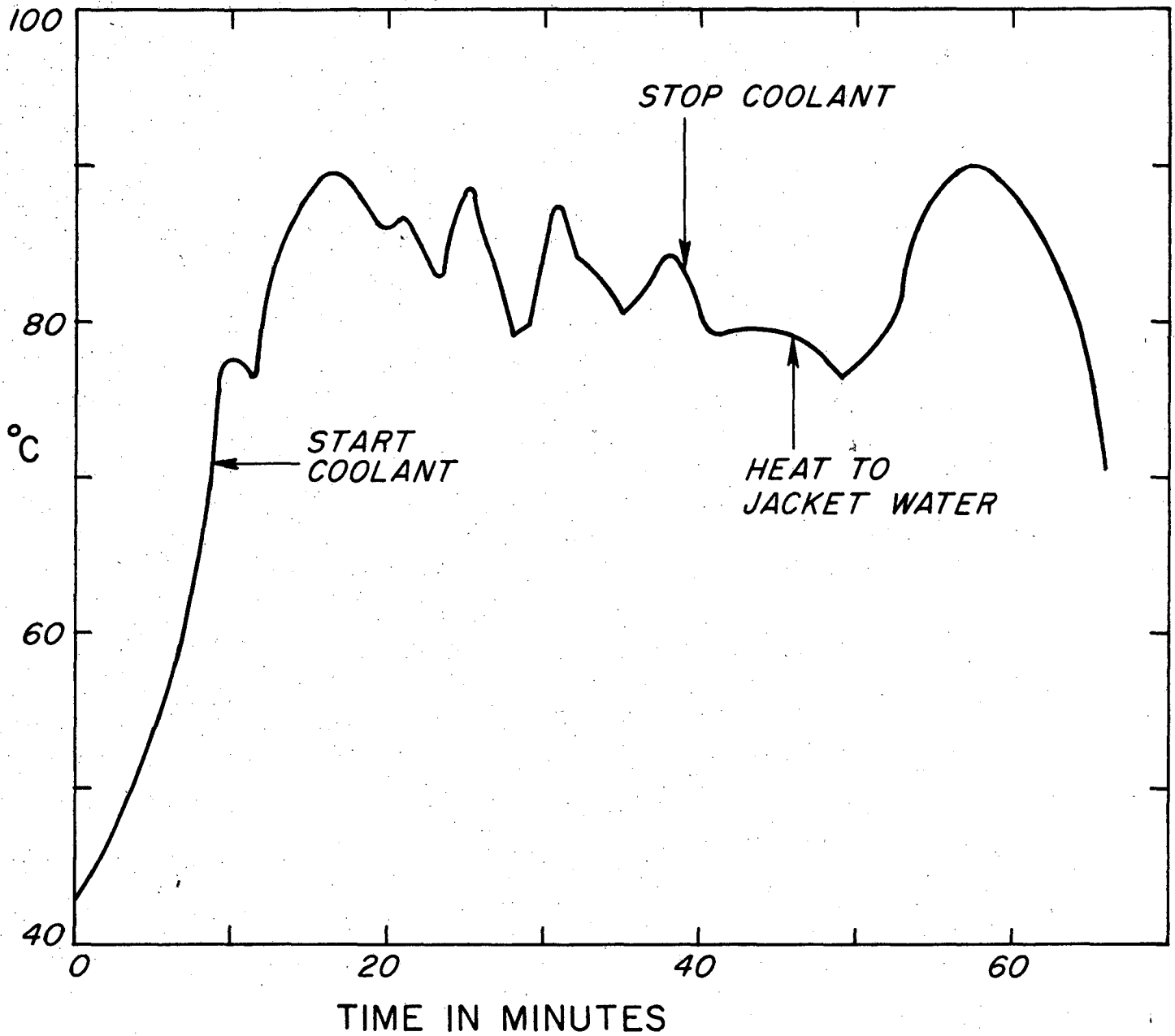


FIG. 8