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TREATMENT OF RADIOACTIVE WASTE STREAMS AT THE IDAHO
CHEMICAL PROCESSING PLANT BY EVAPORATION AND ION EXCHANGE

by

D. W. Rhodes
G. E. Lohse

Idaho Nuclear Corporation
Chemical Programs Division
Idaho Falls, Idaho

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INTRODUCTION

The Idaho Chemical Processing Plant (ICPP) is an AEC-owned facility operated by Idaho Nuclear Corporation at the National Reactor Testing Station (NRTS) for the recovery of fissile materials, principally U-235. This facility is located about 50 miles west of Idaho Falls, Idaho. The irradiated fuels are prepared for solvent extraction of the uranium by several methods including (a) dissolution in mineral acids (nitric, sulfuric, and hydrofluoric); (b) electrolytic dissolution in nitric acid; and (c) in the near future, dissolution of ash (following burning of combustible fuel in a fluidized bed burner) with mineral acids. During the fuel reprocessing, high- and intermediate-level radioactive wastes containing greater than 99% of the fission products, and low-level wastes containing only a fraction of 1% of the fission products, are generated. The high-level wastes are stored for an interim period in cooled 300,000 gallon stainless steel tanks (Type 316 ELC, 304L, and 348) contained within buried concrete vaults. Eventually, these wastes are converted to a granular solid by fluidized bed calcination, and stored in stainless steel bins which are also contained in buried concrete vaults^[1]. Using this technique, nearly all of the radionuclides eventually are stored as a part of the granular solids.

The low- and intermediate-level wastes containing small quantities of radionuclides are processed to minimize the volume of waste that has to be stored, and also to reduce to the lowest practicable level, the amount of radionuclides released to the environment. The intent of this report is to describe the system and methods used for treating these low- and intermediate-level wastes at the ICPP.

WASTE COLLECTION AND TREATMENT

All of the potentially radioactive waste generated at the ICPP, except the high-level waste, is received in an evaporator waste collection tank. This amounts to about 1,200,000 gallons of liquid waste annually and includes waste from laboratory sinks and safety shower drains, cooling water from a uranium product denitrator, process sampler drains, cell floor drains, equipment decontamination, and some wastes from the test reactor areas at the NRTS. A simplified schematic diagram of the liquid waste collection system at the ICPP is shown in Figure 1.

Evaporation-Equipment and Operation

The liquid waste that is received in the waste collection tank is transferred to a thermosyphon-type evaporator; a schematic diagram of the evaporator and associated equipment is shown in Figure 2. This evaporator has an external heat exchanger of 520-ft² heat-exchange area and is rated at 6×10^6 Btu/hr heat duty. All of the equipment is constructed of stainless steel Type 347. Vapor from the flash tank of the evaporator passes into the bottom of a 6-ft-diameter entrainment separator equipped with four bubble-cap trays, where the vapor is scrubbed with a small stream of fresh water. All equipment is designed to operate from high vacuum to about atmospheric pressure. Normally, it operates at about 2 inches of water vacuum. Two 5000-gallon condensate

catch tanks equipped with a remote sampling apparatus receive the condensate. Routinely, the evaporator processes 500 gal/hr of waste continuously, but the waste load ordinarily does not require full time operation.

The waste collected in the waste evaporator collection tank usually contains low concentrations of common chemicals such as nitric acid, sodium hydroxide, potassium permanganate, oxalic acid, aluminum nitrate, zirconyl nitrate, and sodium carbonate. Other chemicals are used occasionally. The waste is usually acidic, but on occasion, may be basic. In addition to the chemicals, radionuclides are present in concentrations up to about 200 $\mu\text{Ci/ml}$. The results of a test using the contents of one evaporator feed tank are shown in Table I.

TABLE I

OPERATING TEST OF ICPP EVAPORATOR USING RADIOACTIVE FEED SOLUTIONS

Measurement	Test Result
Volume of Feed (gal)	14,730
Volume Concentration Factor	21.2
Gross β Activity in Feed ($\mu\text{Ci/ml}$)	185
Gross β Activity in Condensate ($\mu\text{Ci/ml}$)	3.6×10^{-3}
Decontamination Factor (feed/condensate)	5.4×10^4
Decontamination Factor (condensate/concentrate)	1×10^6

These results demonstrate that an appreciable decontamination factor was obtained for gross radionuclides. A high decontamination factor for gross radionuclides is an important characteristic of evaporation compared to other treatments such as flocculation-sedimentation or ion exchange, which are often

selective for specific radionuclides. After sampling and analysis, the condensate is pumped to an ion exchange system for additional decontamination.

Ion Exchange System - Equipment and Operation

The ion exchange system consists of two ion exchange columns 2.0 ft diameter by 3.0 ft high, each containing 6 cubic ft of resin. A schematic diagram of the system is shown in Figure 3. The resin tanks are stainless steel Type 347, and the resin is retained in the tanks with 50-mesh stainless steel screens placed above and below the bed. The columns can be operated either in series or in parallel. The resin is a macroreticular resin, Rohm & Haas Amberlite-200. This type of resin has an open structure and has excellent physical characteristics. It is made with tougher copolymers than standard gel-type resins and has more than 20% divinylbenzene by weight in its copolymer. For this reason, it provides good resistance to attrition and oxidative attack. It has particularly high affinity for strontium-90^[2], which is one of the main requirements for decontaminating evaporator condensate at the ICPP, and functions well at low pH values.

The columns are operated downflow and regenerated upflow. Sodium nitrate solution is used as the regenerant; the expended regenerating solution is routed to the waste evaporator feed tank for eventual calcination.

In an operating test, batches of evaporator condensate were sampled and analyzed prior to and after treatment in the ion exchange system. Analysis of 32 batches of condensate (influent) prior to treatment over a three-month period had the radiochemical composition shown in Table II.

TABLE II
RADIOCHEMICAL COMPOSITION OF EVAPORATOR CONDENSATE

Radionuclide	No. of Samples	Concentration, $\mu\text{Ci/ml} \times 10^{-4}$		
		Avg	Minimum	Maximum
Gross β	32	11.00	1.20	48.00
Cs-137	30	1.50	0.23	4.50
Sr-90	32	1.40	0.26	8.90
Ru-106	28	0.68	0.17	1.70
Ce-144	26	2.00	0.14	13.00
ZrNb-95	18	0.42	0.07	0.75
Sb-125	3	<0.28	--	--
pH	32	2.08	1.20	2.61

The average concentrations (Table II) for all radionuclides in the influent except Sr-90 were below drinking water requirements for occupational release [3]. In actual practice, the effluent from the evaporator ion exchange columns at the ICPP mixes with nonradioactive cooling water in the ratio one volume of condensate to greater than 100 volumes of cooling water before it is released. This reduces the concentrations to values below drinking water requirements for the general population. Thus, the incentive for removal of radionuclides from the condensate is based on a philosophy of removing radionuclides to the "lowest practicable level" rather than a need to meet specific release limits.

The average concentration for Sr-90 in the influent was about 14 times greater than the requirements for occupational releases. Thus, Sr-90 is the principal contaminant that needs to be removed by the ion exchange system.

The purpose of the ion exchange system is to reduce the long-lived radionuclides, Sr-90 and Cs-137, to as low a level as practicable with particular emphasis on the removal of Sr-90. The results of the test run are shown in Figures 4, 5, and 6.

These results indicate good removal of Cs-137, Sr-90, and Ce-144 for about 3000 bed volumes. Beyond this point, breakthrough of Cs-137 was evident with 100% breakthrough occurring at about 5000 bed volumes. Strontium breakthrough began at about 4000 bed volumes, but was still only about 1% at 6000 bed volumes (~270,000 gal). Removal of Ce-144 was essentially complete throughout the test.

Very little Ru-106 or ZrNb-95 were removed by the ion exchange resin. Ruthenium chemistry in waste processing is extremely complex and is known to be intimately related to nitrogen-oxygen chemistry, which also is complex. Both the nitrate and nitro complexes of ruthenium are present usually in equilibrium in waste solutions^[4]. Zirconium and niobium form complexes, also, usually anionic complexes, in process solutions. However, because of the relatively short half-lives (<1 year) and low biological toxicity of Ru-106 and ZrNb-95, and their moderate-to-low concentration in evaporator condensate, no additional attempt was made to remove these radionuclides.

The breakthrough data indicate that the need for regeneration will be determined by Cs-137. This is a convenient phenomenon, because (a) Cs-137 is a gamma emitter, while Sr-90 is a beta emitter; thus, Cs-137 can be used readily to determine breakthrough by appropriate instrumentation, and (b) the permissible level for Cs-137 in drinking water is about 70 times greater than Sr-90, so a higher breakthrough value for Cs-137 is permissible. Based on the test run data, if regeneration is determined by Cs-137 leakage, removal of Sr-90 will be adequate when the run is terminated.

Effect of pH

The possibility that the pH of the condensate might be higher in some batches of condensate exists, because alkaline decontamination solutions are used occasionally. To determine the effect of higher pH on the performance of the ion exchange columns, equilibrium-type experiments were run using condensate to which sodium hydroxide had been added to raise the pH. The results of this study are shown in Figure 7. Increasing the pH apparently improves the removal of Cs-137 slightly and improves the removal of Sr-90 markedly. Thus, a less acidic condensate will be beneficial rather than detrimental.

The possibility of deliberately raising the pH to increase the capacity of the resin for Sr-90 does not appear to be feasible for the current ion exchange system. Another ion exchanger more specific for Cs-137 would need to be added to the system; otherwise, Cs-137, which is not greatly affected by increasing the pH, would still be the "controlling" radionuclide.

CONCLUSIONS

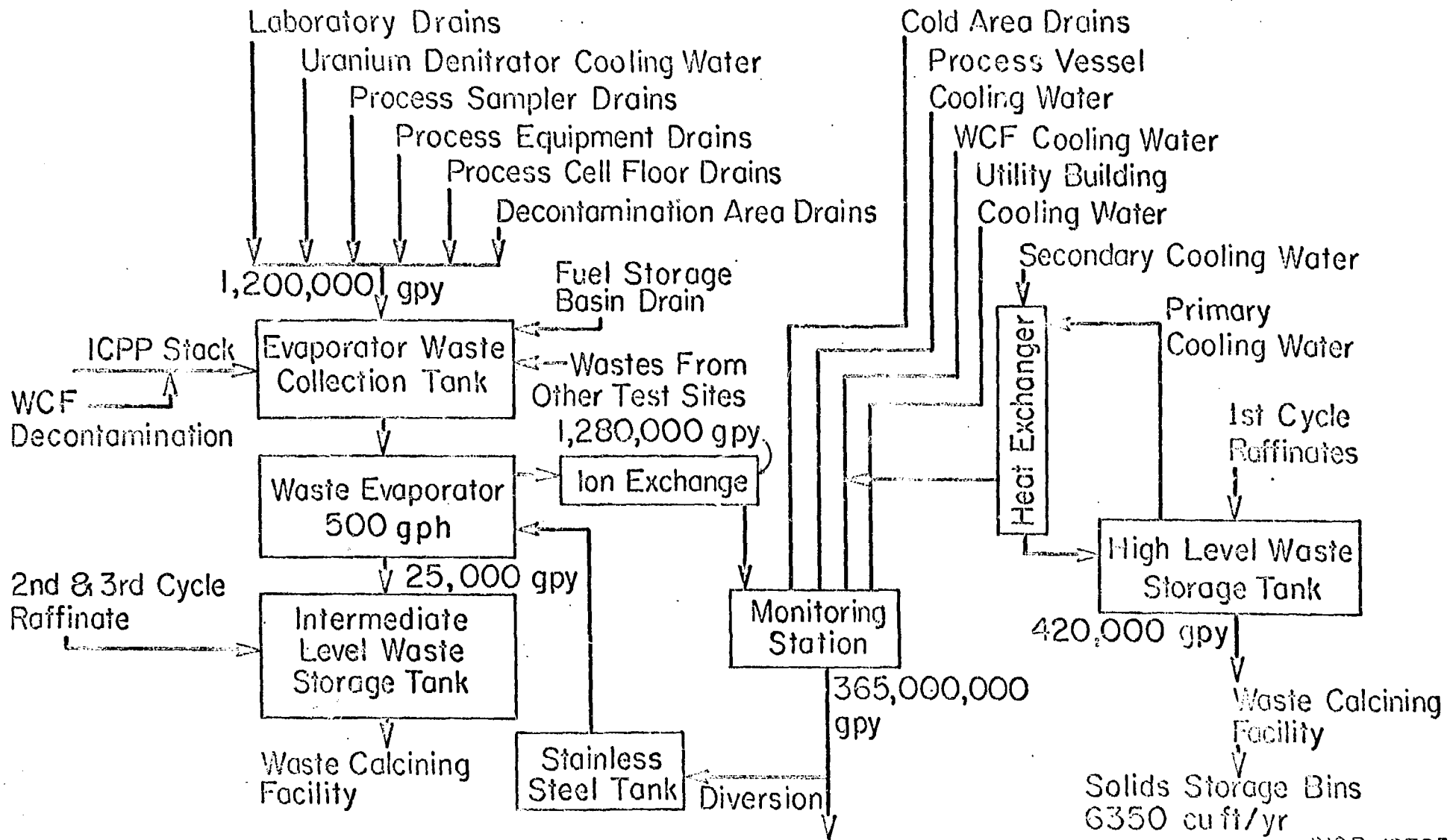
Cs-137 and Sr-90 can be removed from ICPP acidic evaporator condensate by ion exchange on Amberlite-200 ion exchange resin. However, neither Ru-106 nor ZrNb-95 are removed from ICPP evaporator condensate by the ion exchange system. This is due largely to the formation of complex or anionic species of Ru-106 and ZrNb-95 which do not react with the cation resin.

The need for regeneration in the ICPP system can be determined readily by monitoring for Cs-137 in the column effluent with reasonably high assurance that Sr-90 is being removed adequately.

At the ICPP, the annual input of Sr-90 to the plant is about 1×10^6 Ci in irradiated fuel elements. Most of this eventually is stored in calcined solids, but some finds its way to the intermediate- and low-level wastes. With an evaporation and ion exchange system for final cleanup of these wastes, less than 5×10^{-4} Ci of Sr-90 is released in the approximately 365,000,000 gallons of water returned annually to the ground water table.

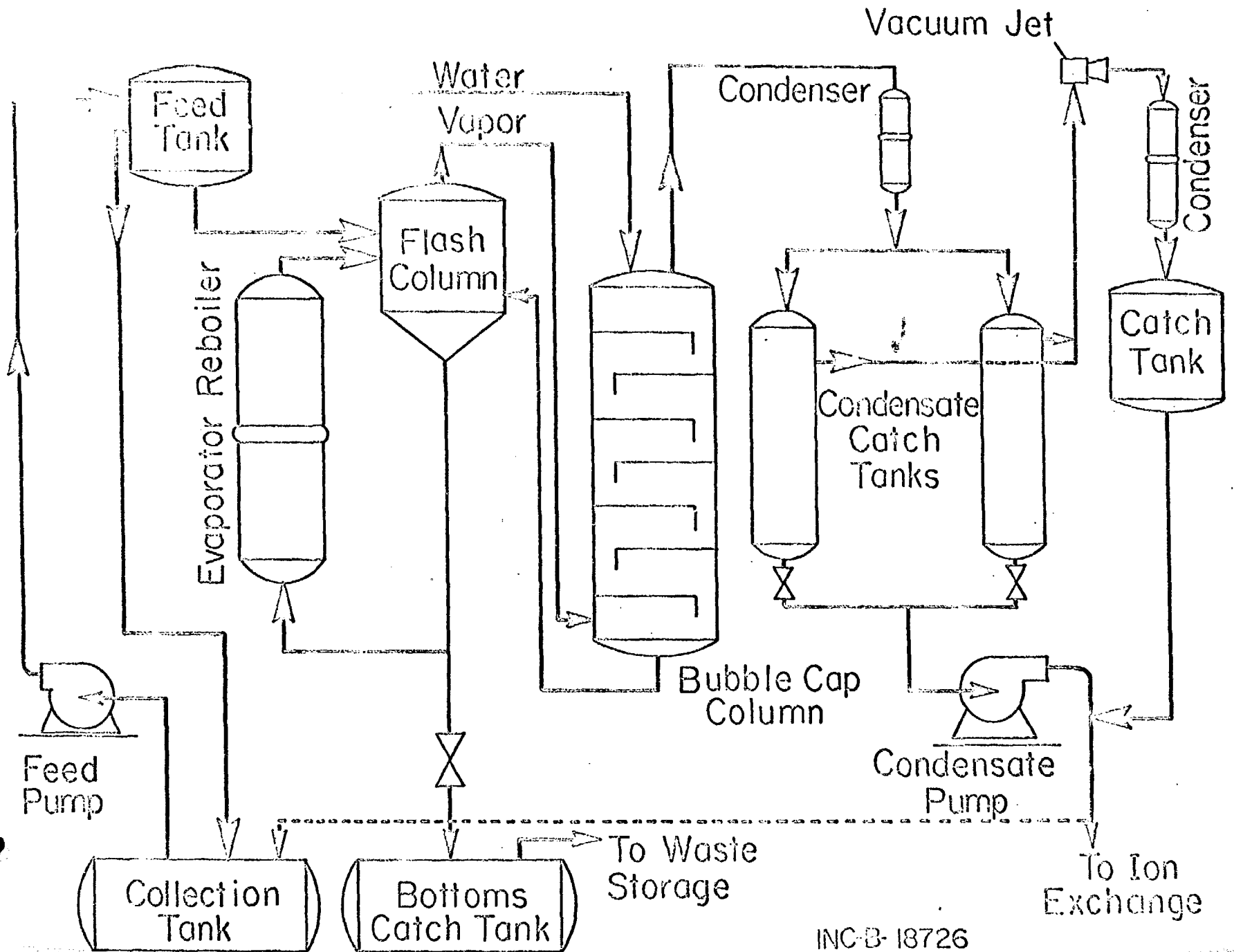
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2. B. W. Mercer, Ion Exchange Equilibria of Trace Cesium-137 and Strontium-85 in Multicomponent Systems, BNWL-SA-1173 (March 1967).
3. Rules and Regulations of the USAEC, Standards for Protection Against Radiation, Title 10 CFR Part 20, Appendix B, published in the Federal Register December 10, 1969.
4. J. M. Fletcher et al, J. Inorg. Nucl. Chem., 1 (1955) pp 378-401.



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Fig. 1 Liquid waste collection systems at the ICPP.



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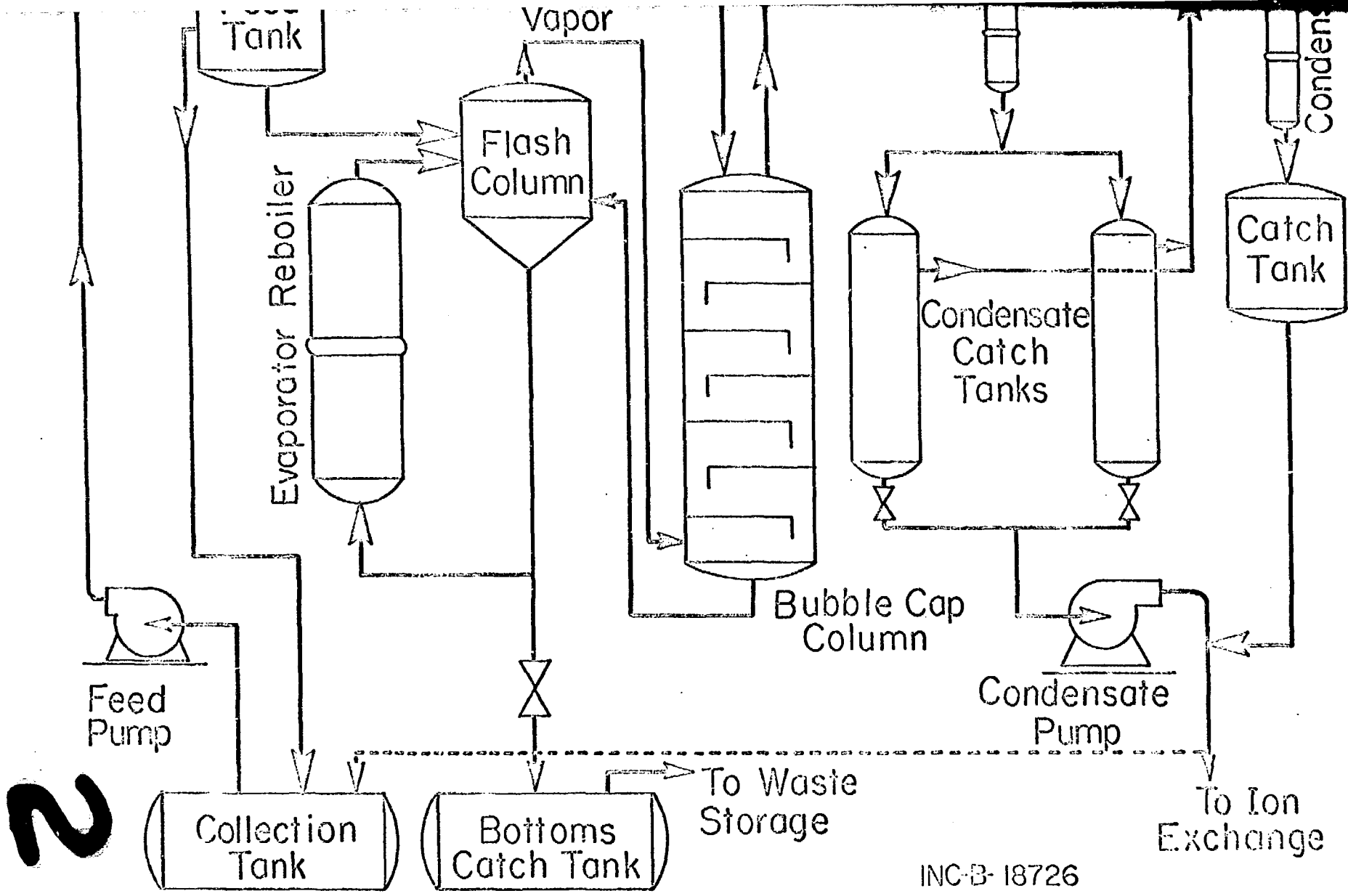


Fig. 2 Continuous waste-evaporation system.

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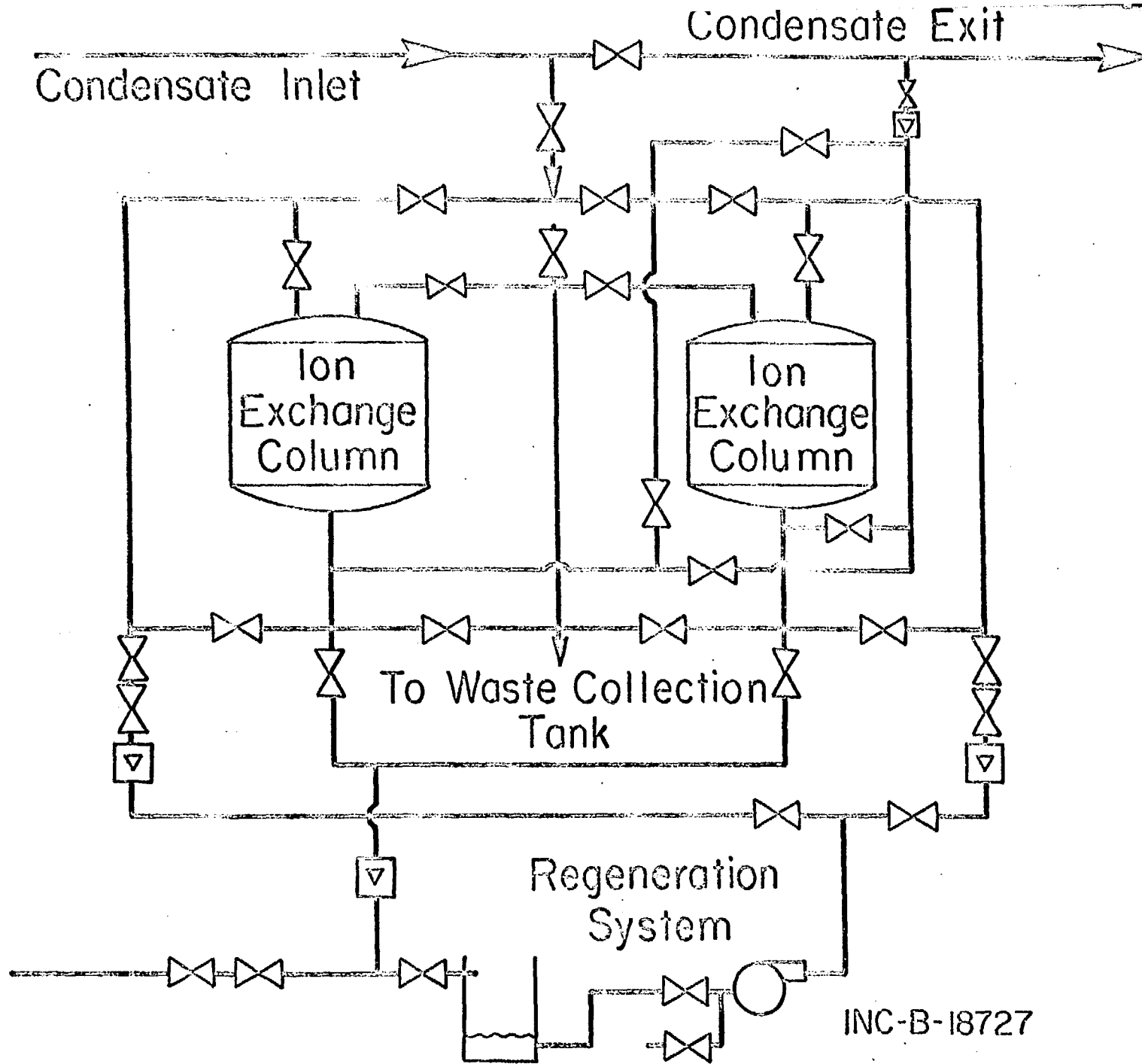


Fig. 3 CPP-Condensate Decontamination scheme.

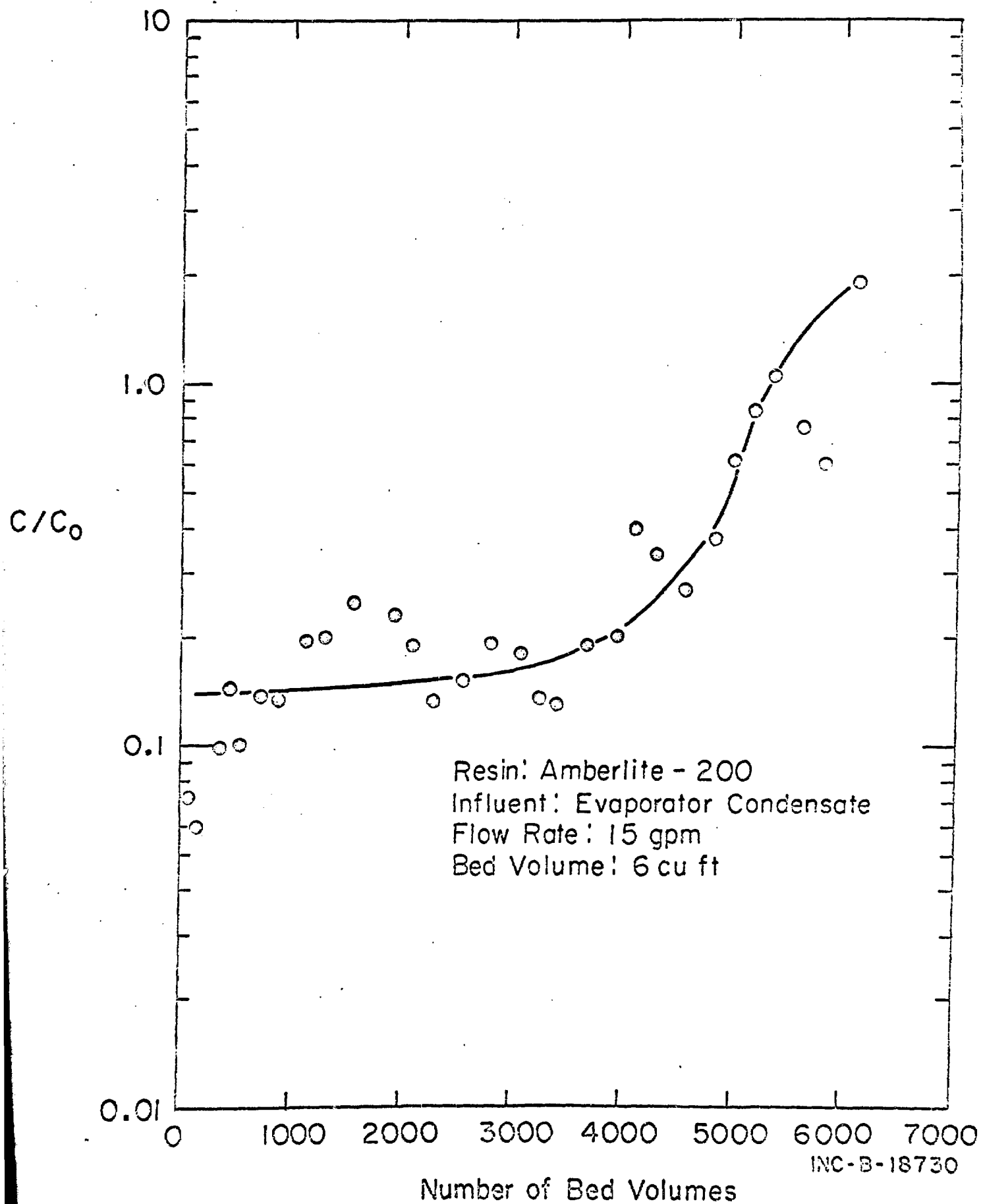


Fig. 4 Breakthrough curve for total beta emitters in evaporator condensate.

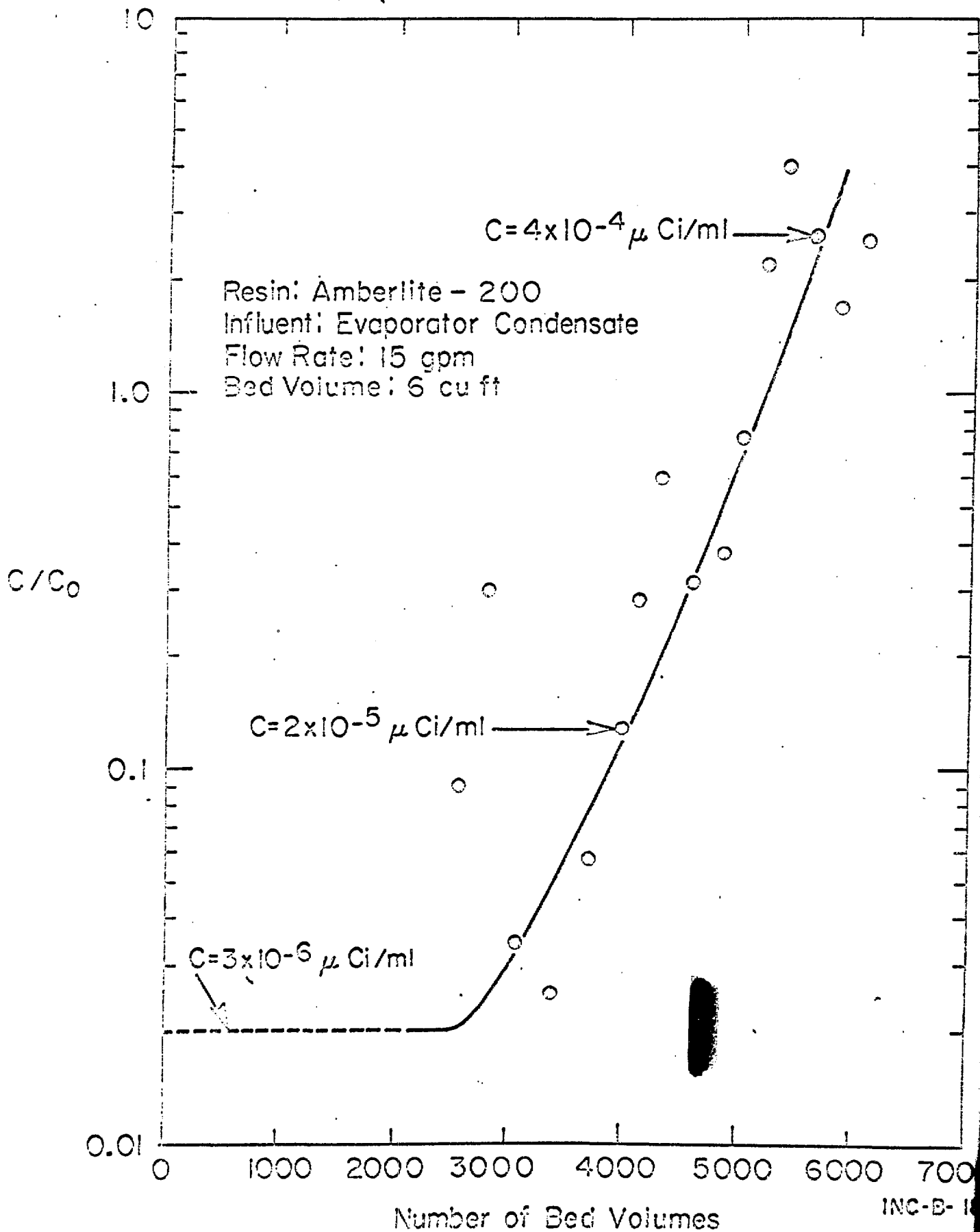


Fig. 5 Breakthrough curve for Cs-137 in evaporator condensate.

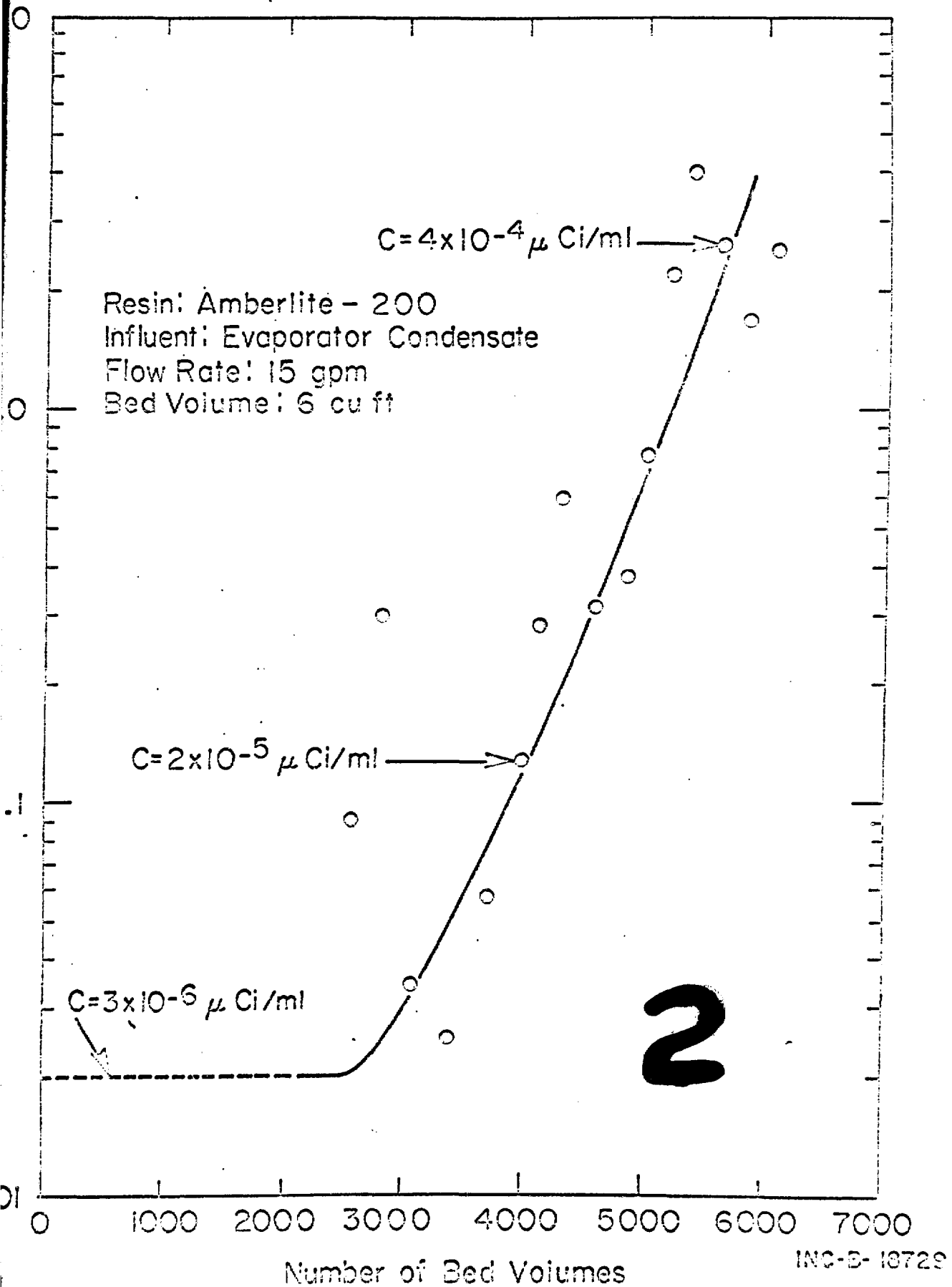


Fig. 5 Breakthrough curve for Cs-137 in evaporator condensate.

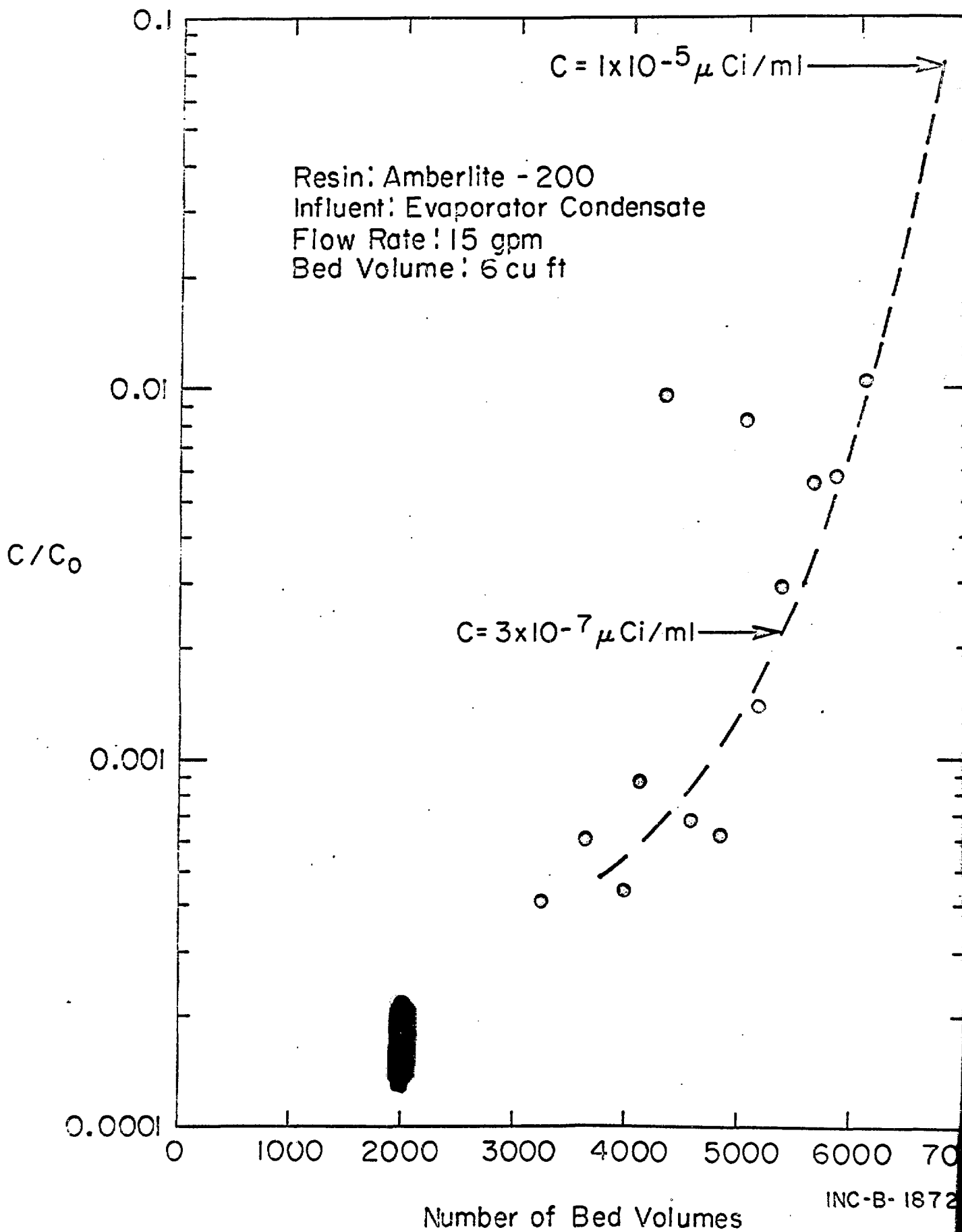


Fig. 6 Breakthrough curve for Sr-90 in evaporator condensate.

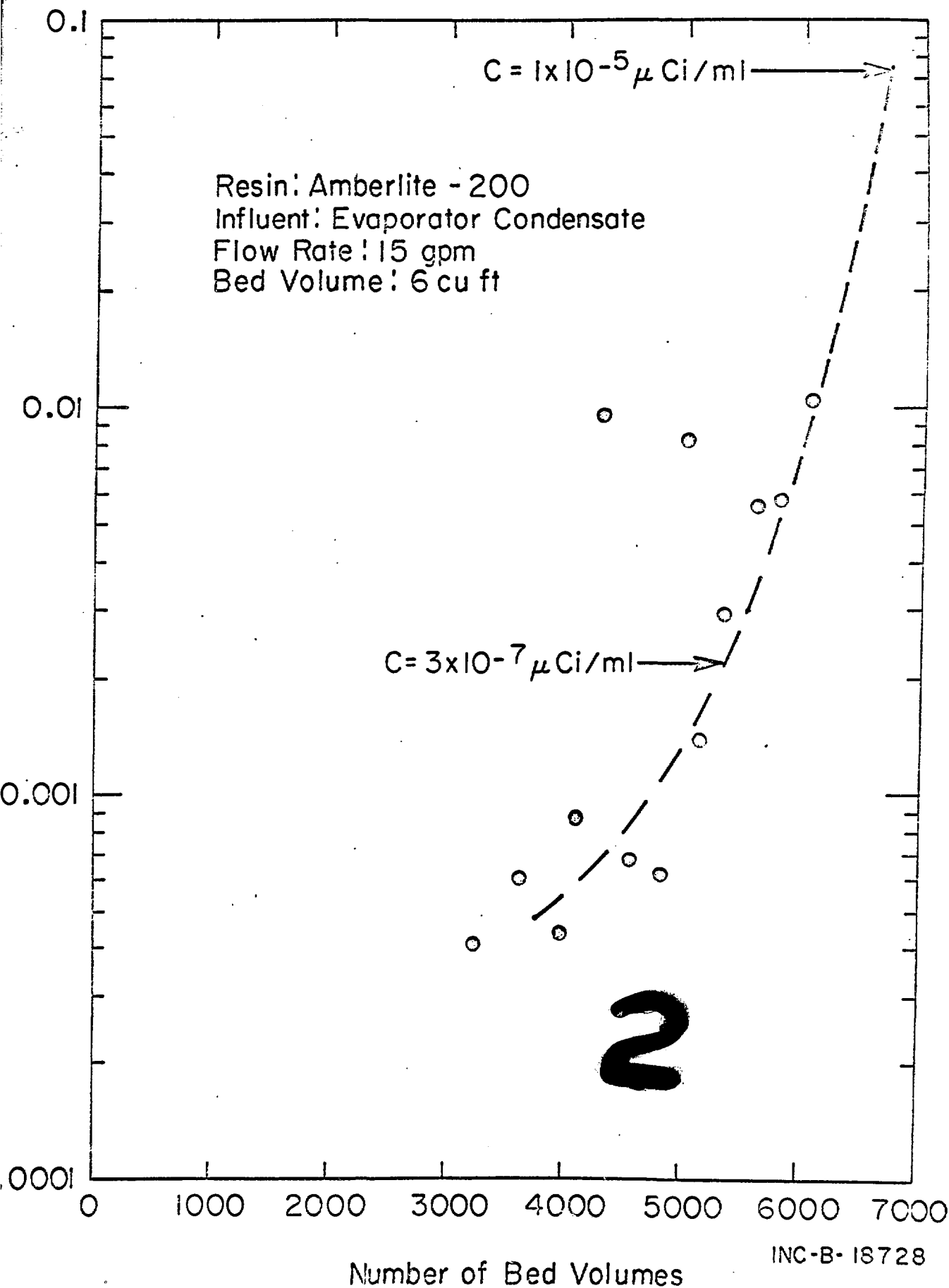


Fig. 6 Breakthrough curve for Sr-90 in evaporator condensate.

Distribution Coefficient (Kd)

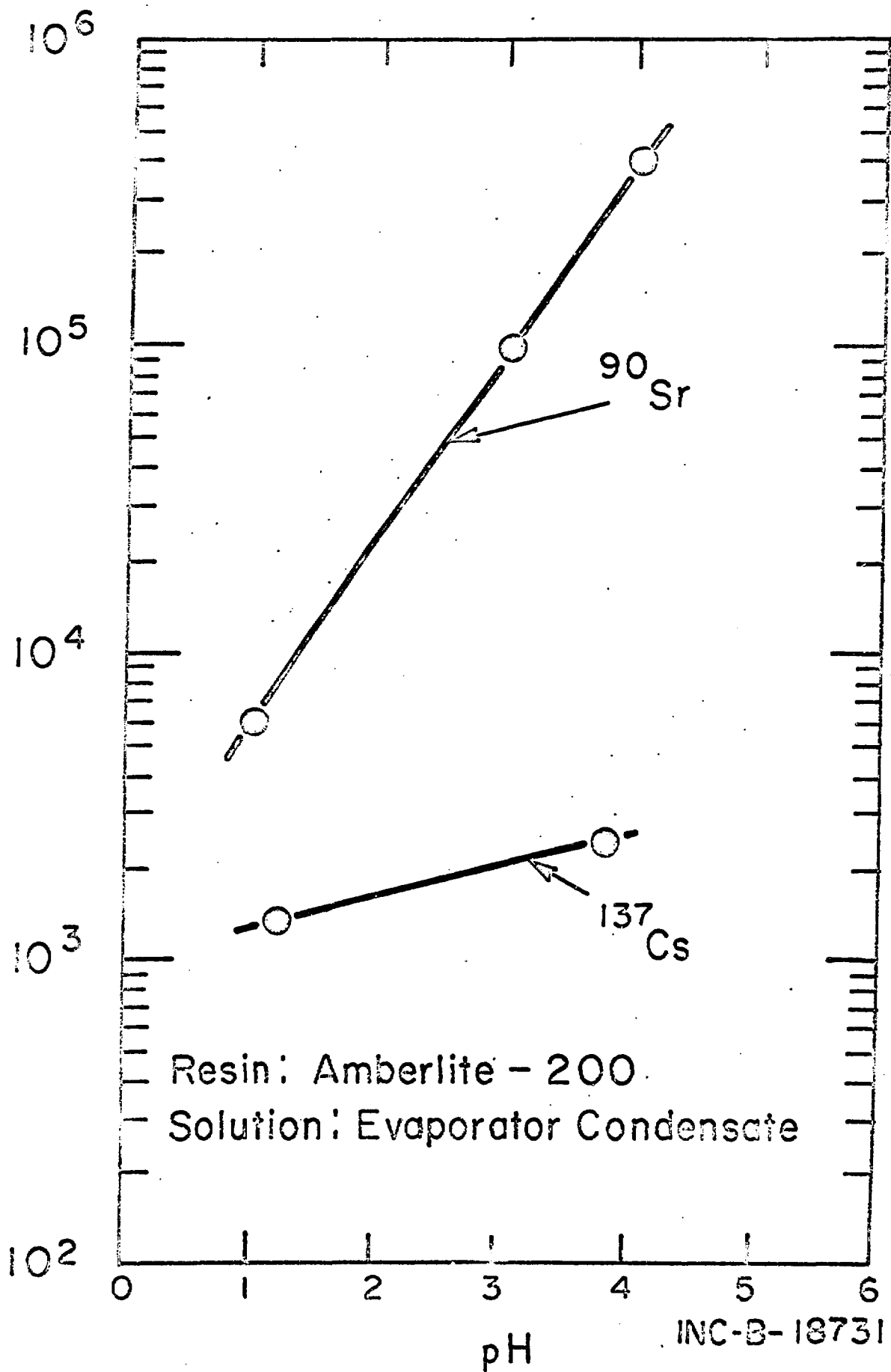


Fig. 7 Effect of pH on cesium and strontium distribution coefficients.