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UNIVERSITY OF MICHIGAN

ANN ARBOR

CHELATION SOLVENT - EXTRACTION OF STRONTIUM, BARIUM
AND CESIUM FROM GROSS FISSION PRODUCTS

PROJECT D-109

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I ABSTRACT

A process is presented for the extraction of strontium, barium, and cesium from solutions of gross fission products. Basis for the process is a chelation-solvent extraction using thenoyltrifluoroacetone (TTA) dissolved in methylisobutylketone (MIBK).

The process is supported by data from sixty-six laboratory equilibrations. These data are presented in plots of distribution ratio versus pH as well as in tabular form. Radioactive tracers were used in determining the distribution ratios and the procedures used in making these runs are given.

A flowsheet is presented illustrating how this data combined with data from other sources can be used in the process to extract the long-lived fission products from gross fission waste solutions that are derived from plants processing spent nuclear fuels. The purpose of this process is to provide a means for preparing pure Cs-137 and Sr-90 for industrial uses at low cost and to permit discarding of the remaining fission products after a storage period of approximately six years.

II INTRODUCTION

One of the major problems in using nuclear energy for industrial purposes lies in disposing of radioactive fission products. At the present time these products are stored in large tanks as waste solutions from the chemical processing plants. Storage of these solutions is costing the Atomic Energy Commission at various sites from \$0.60 to \$3.00 per gallon. While storage costs accumulate, industry and academic institutions need radioactive materials as sources of beta and gamma irradiation. Such irradiation can be used to promote chemical reactions, sterilize foods and medical supplies, in thickness gages and instrument applications, and for industrial X-ray applications. The two most valuable isotopes for industrial use would be the long-life fission products of Cs-137 and Sr-90 which have half-lives of 37 and 20 years respectively. The removal of these two isotopes from gross fission products would serve two purposes: It would permit the discard of processing plant wastes after a storage time of five to six years; and, if the isotope removal process were economical, it would permit the sale of long-lived isotopes at a price well below the present one. There is also the possibility that, if processing charges could be reduced considerably, industrial interests could be so stimulated that fission products would be transformed from hazardous waste material to valuable commodities.

A review of possible processes for extracting Cs-137 and Sr-90 by conventional chemical precipitations, filtrations, or centrifugation steps indicated that removal would be incomplete and processing costs would be excessive. Likewise, methods of carrying Cs and Sr down as ferrocyanide precipitates appear to give incomplete recovery, although processing costs are reasonable. References on chelation-solvent extraction separations of the alkaline earth and alkali metals indicated that such an approach might be reasonable (1) (2) (3) (4) (5). Similar solvent extraction techniques have been successfully worked out for separating fissionable material from gross fission products, so these methods were considered worth investigating.

Funds were made available from the Engineering Research Institute and investigations were initiated in October, 1954. The Fission Products Laboratory furnished laboratory space to carry out the project.

III EXPERIMENTAL PROCEDURE AND DATAA. Run Procedures

An aqueous solution of nitrate salts was contacted with an organic solution of TTA. Sodium hydroxide solution was used to adjust pH. To determine the distribution ratio, a small amount of radioactive tracer solution was mixed with the non-radioactive aqueous solution. The two phases were then mixed by shaking until the pH of the aqueous phase was constant, indicating equilibrium had been reached. The phases were then separated by centrifuging, and the radioactivity of each phase was determined. From the latter measurements, the distribution ratio was calculated, the concentration of metal in each phase being proportional to the concentration of the radioactive isotope of that metal in each phase.

Runs were made with only one tracer present per run at various concentrations of the TTA and salts, different excesses of TTA over the stoichiometric amount for 100% chelation, and with combinations of two or more metals.

Runs were also made to determine solubility of the metal chelate in various solvents. Barium-TTA was made, crystallized from the organic phase, and portions of it placed in various solvents such as benzene, petroleum ether, heptane, methyl-isobutyl ketone (MIBK). The chelate dissolved readily only in MIBK. This was verified in a later run in which no extraction occurred when benzene was substituted for MIBK as the solvent for TTA.

The probable error in the counting and sampling methods was measured. Various size aliquots of the aqueous phase of one run were taken and evaporated onto discs and counted. The probable error was calculated for each size aliquot. This was done to determine the effects of activity magnitude on the overall counting accuracy.

The run procedure followed in general is illustrated by Run #52. An aqueous feed solution was prepared which was 0.069 molar with respect to $\text{Sr}(\text{NO}_3)_2$, 0.0342 molar with respect to $\text{Ba}(\text{NO}_3)_2$ and 0.1070 molar with respect to CsCl . Of this solution, 18 milliliters were mixed with 15 milliliters 0.8175 molar TTA in MIBK and 2.0 milliliters of Sr-89 was added. The tracer was approximately 10^{-6} molar with respect to Sr^{+2} , and it had an activity of about 10 millicuries per liter. The mixture was shaken by hand for two minutes and its pH taken using a Beckman Model H-2 pH meter equipped with glass and calomel electrodes. This was repeated until the pH was constant for two consecutive readings. Then 5 milliliters of each were put in centrifuged tubes and centrifuged to separate the phases. Appropriate size aliquots ranging from 25 to 1 milliliters were withdrawn from each phase and evaporated under an infra-red heat lamp onto aluminum discs, one inch in diameter. These discs were then placed on a 2-1/4 x 3-1/4 inch card under a geiger tube. The tube was mounted vertically, face downward in a lead shield below a plastic rack designed to hold the cards from 1/2 to 2-1/2 inches from the tube face. An automatic counter was connected to the geiger tube. With the disc under the tube, each phase was counted separately. The distribution ratio was then determined from the results.

The remainder of the 10 milliliters was returned to the reaction vessel and a certain volume of 0.20 NNaOH was added to raise the pH. The procedure was then repeated and the distribution ratio determined for that pH until the range of pH from four to eleven was covered. The curve of D_o/a versus pH was then drawn for each run.

The results shown in Figure 1 indicate that most of the increase in distribution ratio, that is, most of the chelation, took place between a pH of four and eight.

B. Run Data

Tables I and II show a tabulation of all the significant runs together with the objectives of each run and compositions of the original solutions.

Data on pertinent runs have been extracted and plotted in Figures 1 through 8 to indicate effects of various variables.

C. Results

Figure 1 shows the results of runs made with mixed strontium, barium and cesium in the aqueous phase. These runs were made to determine if one salt affects another in the chelation extraction process. In these figures, the distribution ratio, organic over aqueous, phases are plotted against pH of the aqueous phase. By comparing the data in Figure 1 with that of Figure 7, it is evident that an increase in the metal ion concentration in the aqueous phase increases the ultimate distribution ratio. The ratio of Sr^{++} , Ba^{++} , Cs^+ in this run is the same as would occur in gross fission products. The magnitude of the distribution coefficients for Sr^{++} and Ba^{++} is somewhat surprising for such alkaline elements.

Data in Figure 2 was plotted to determine if a split could be made between strontium and barium. Although the figure indicates that a difference in distribution ratio does exist, the exact magnitude of the partition coefficient would have to be determined more accurately before it can be definitely stated that Sr and Ba splits can be made. Maximum distribution coefficients shown in Figure 2 are not as high as in Figure 1. This is due to the difference in TTA excesses used in the two runs.

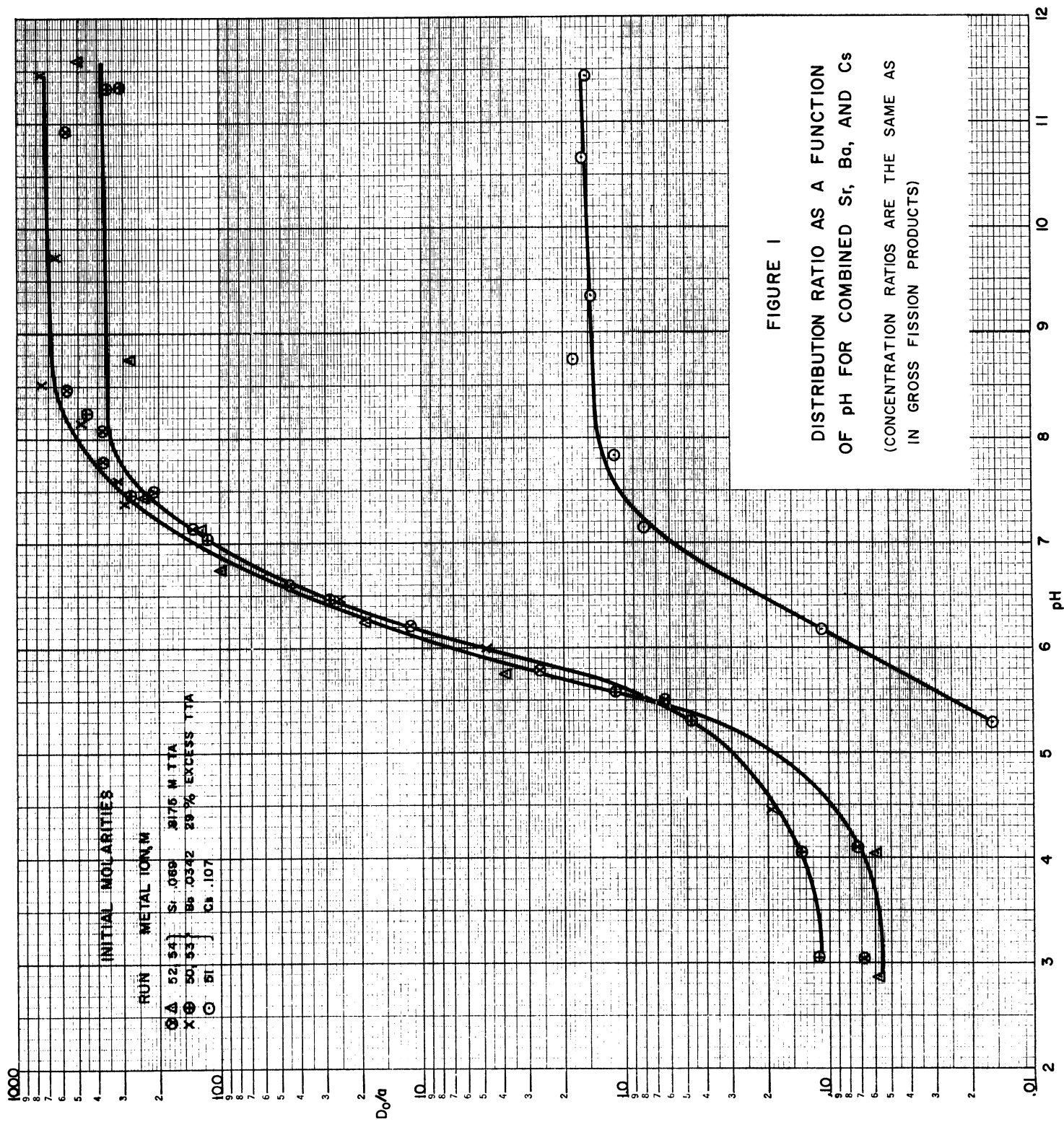
Figures 3, 4, and 5 show the effects of varying excesses and varying concentration of TTA upon the chelation of Cs^+ . The chelation of Sr^{++} and Ba^{++} is well underway at a pH of 6, while appreciable Cs^+ does not chelate until a pH of 7 is reached; therefore separates of Sr^{++} and Ba^{++} from Cs^+ should be easily accomplished. The same conclusions can be reached from the data in Figure 1. Chelation of Cs^+ is quite sharp at a pH of 7, and there is no reason why the distribution ratio should not increase to high values if the TTA would not hydrolyze. This hydrolysis begins to occur at a pH of 7.0 and thus accounts for the

abrupt curvature shown in all the curves above this pH. The maximum distribution for Cs^+ of about 1.5 is shown in Figure 1. This value would be adequate for Cs^+ extraction in countercurrent equipment if chelating agent hydrolysis could be avoided. Figure 5 quite markedly illustrates the effects of TTA concentration upon Cs^+ distribution coefficients.

Figure 6 shows the effects of TTA concentrations upon Ba^{++} distribution. In this case the changes in D_o/a are not as great as in Figure 5, since the TTA concentrations are not varied as widely. Over the TTA concentrations changes given the qualitative effects are the same on Ba^{++} as on Cs^+ in that an increase in TTA concentration increases the distribution ratio. This agrees with theory as will be discussed later.

Figure 7 illustrates an important point, in that changes in metal ion concentrations are shown to have much more effect on distribution coefficient than changes of far greater magnitude in TTA excess.

Figure 8 illustrates the effects of excess TTA upon the distribution ratio of Sr^{++} . This effect can be predicted reasonably well by theory.



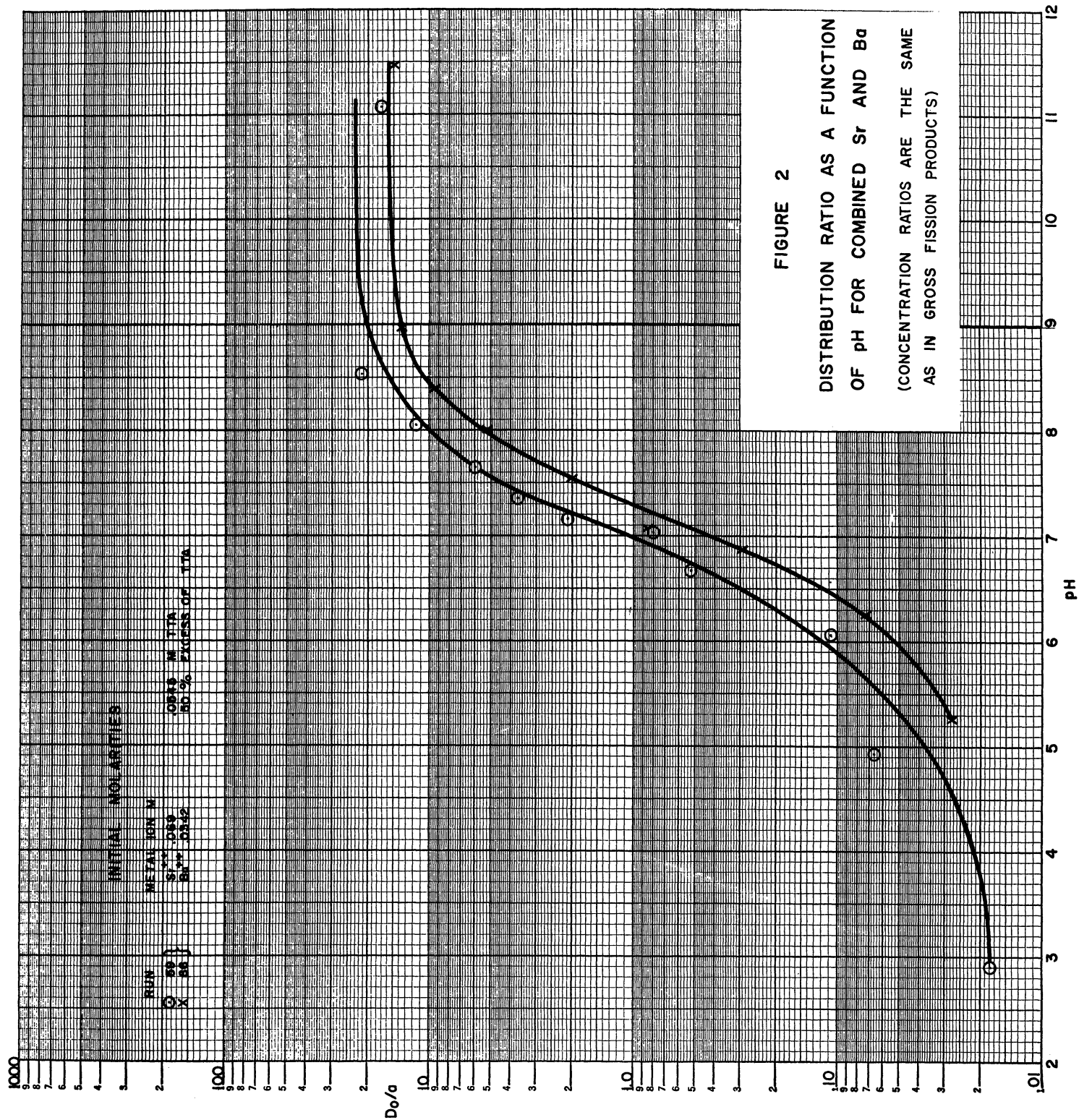
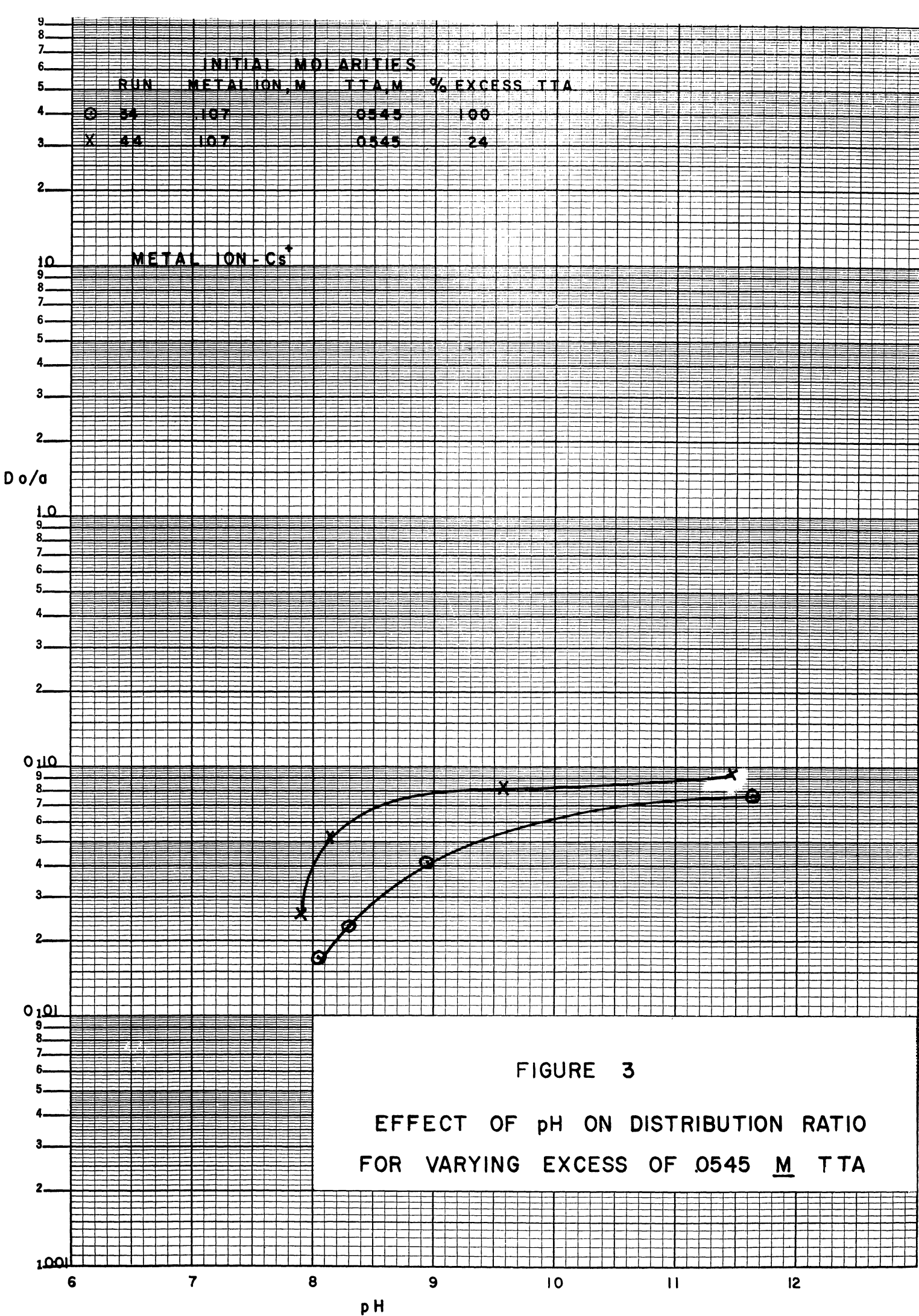
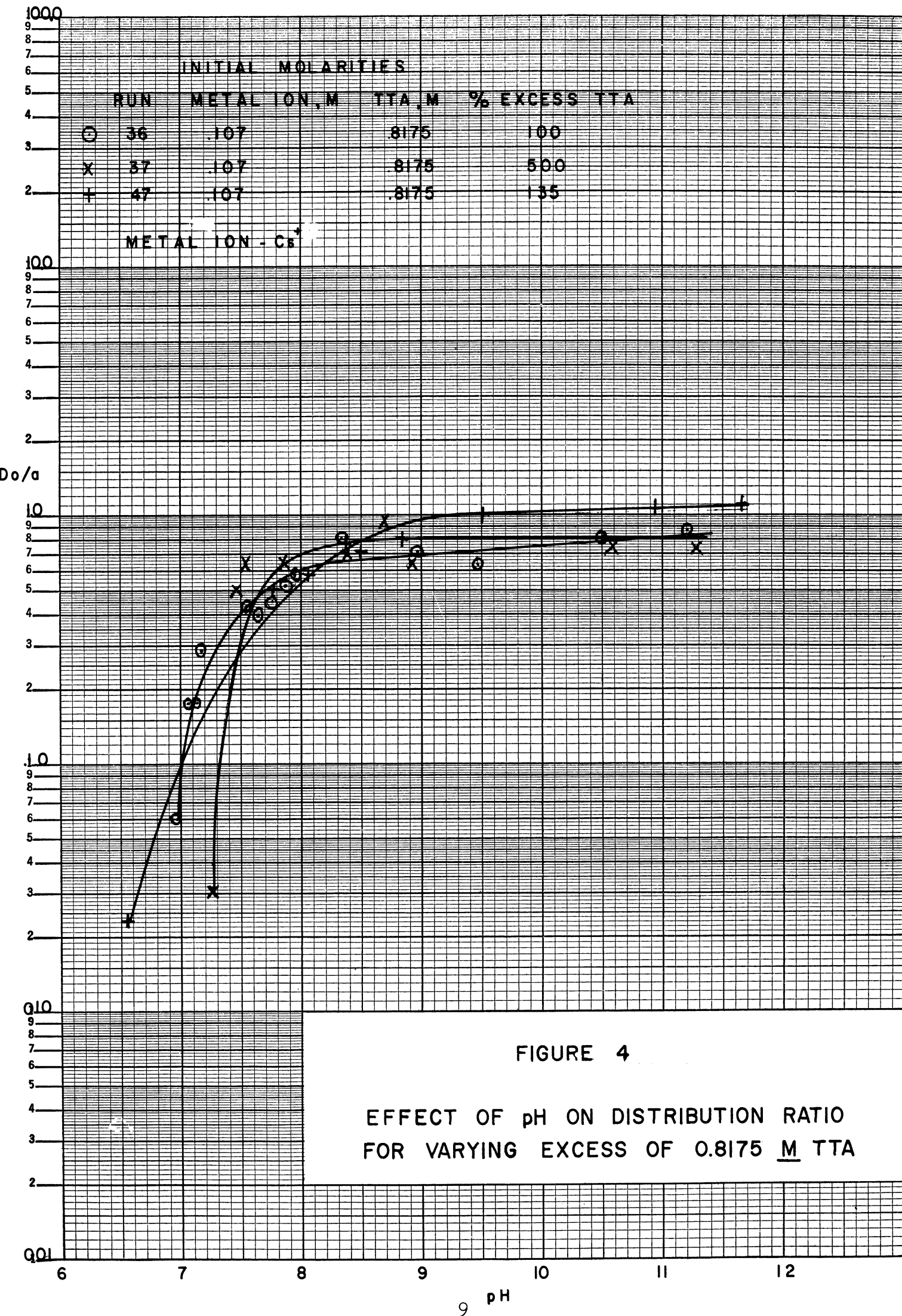
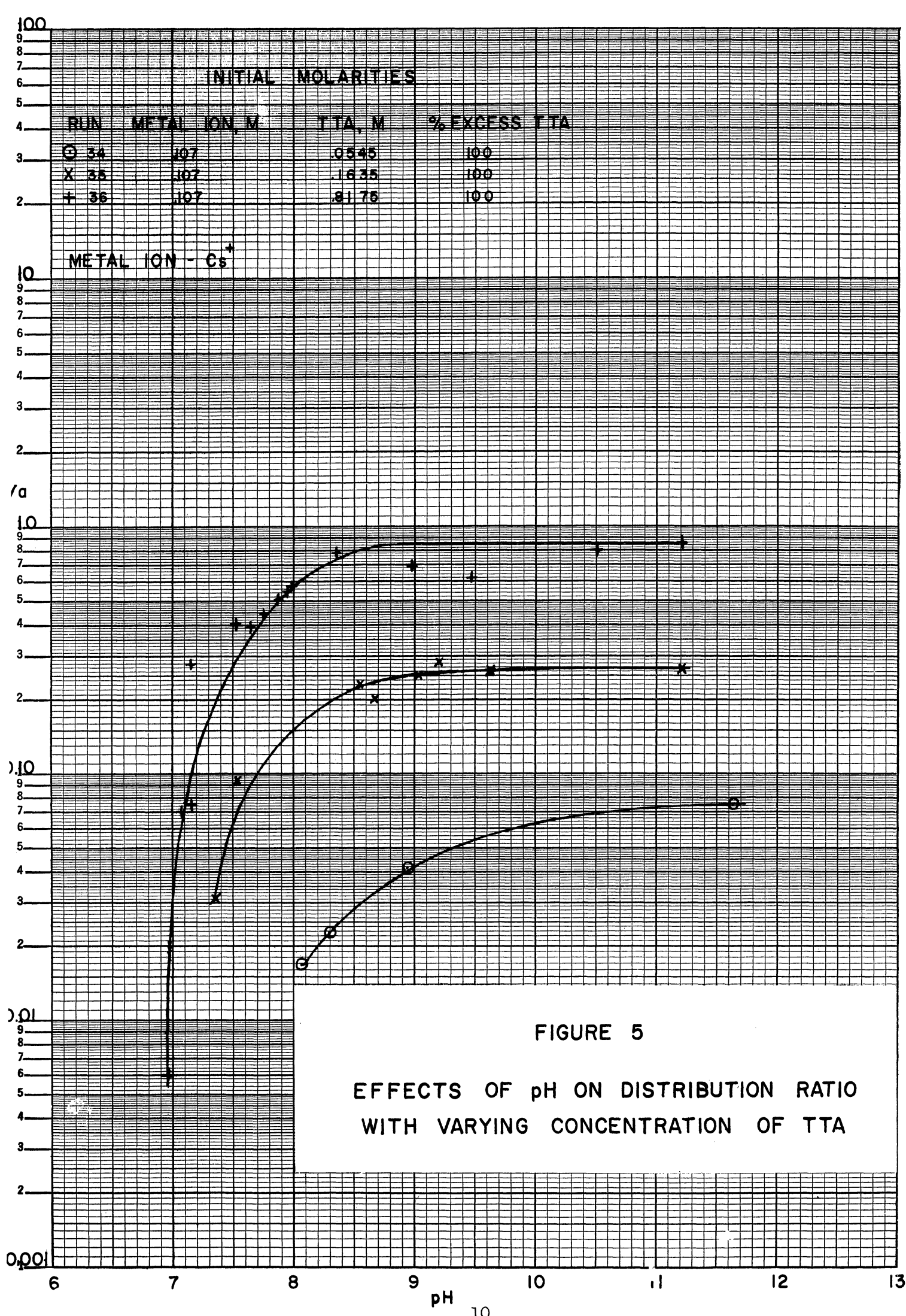


FIGURE 2

DISTRIBUTION RATIO AS A FUNCTION
OF pH FOR COMBINED Sr AND Ba
(CONCENTRATION RATIOS ARE THE SAME
AS IN GROSS FISSION PRODUCTS)







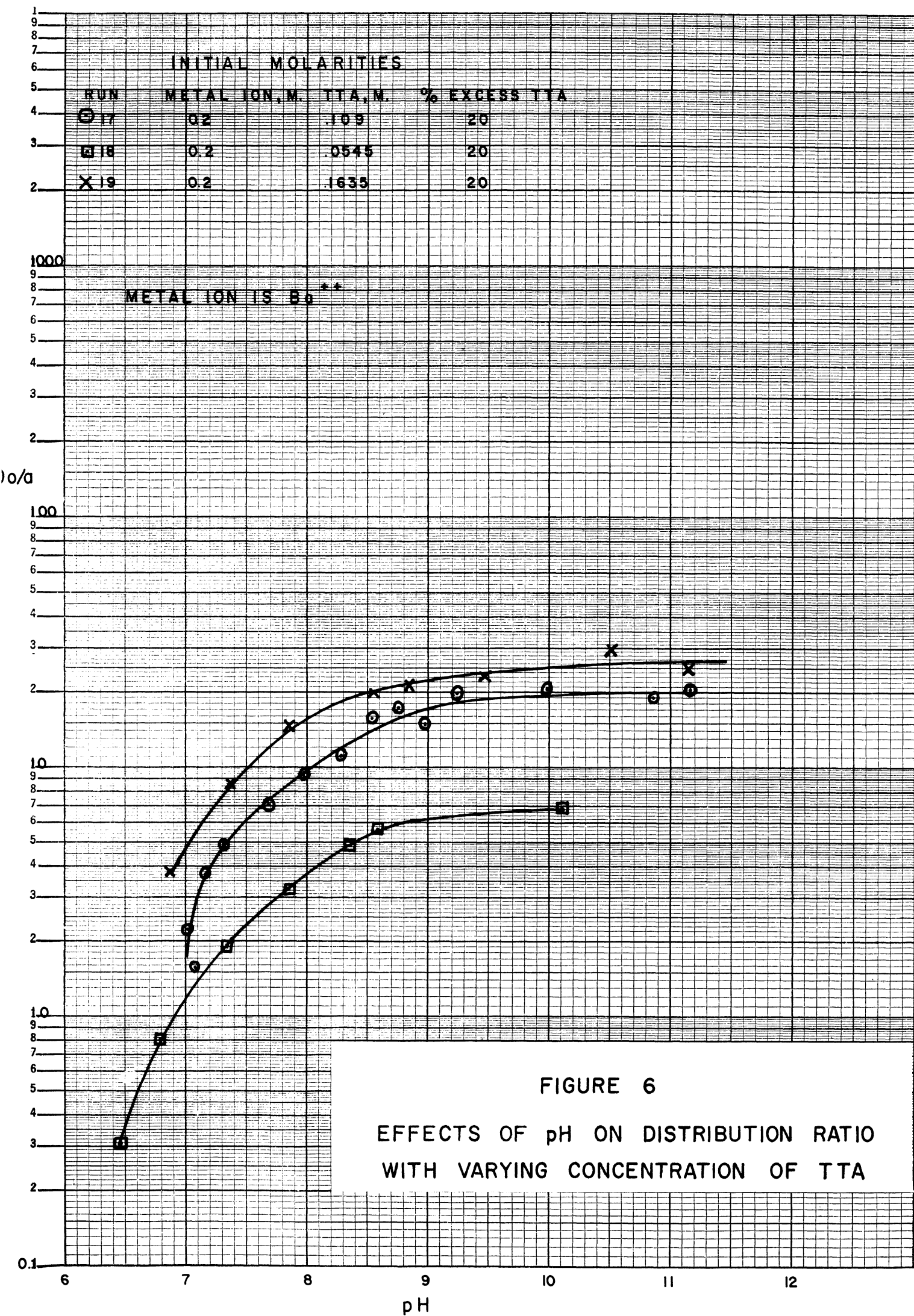
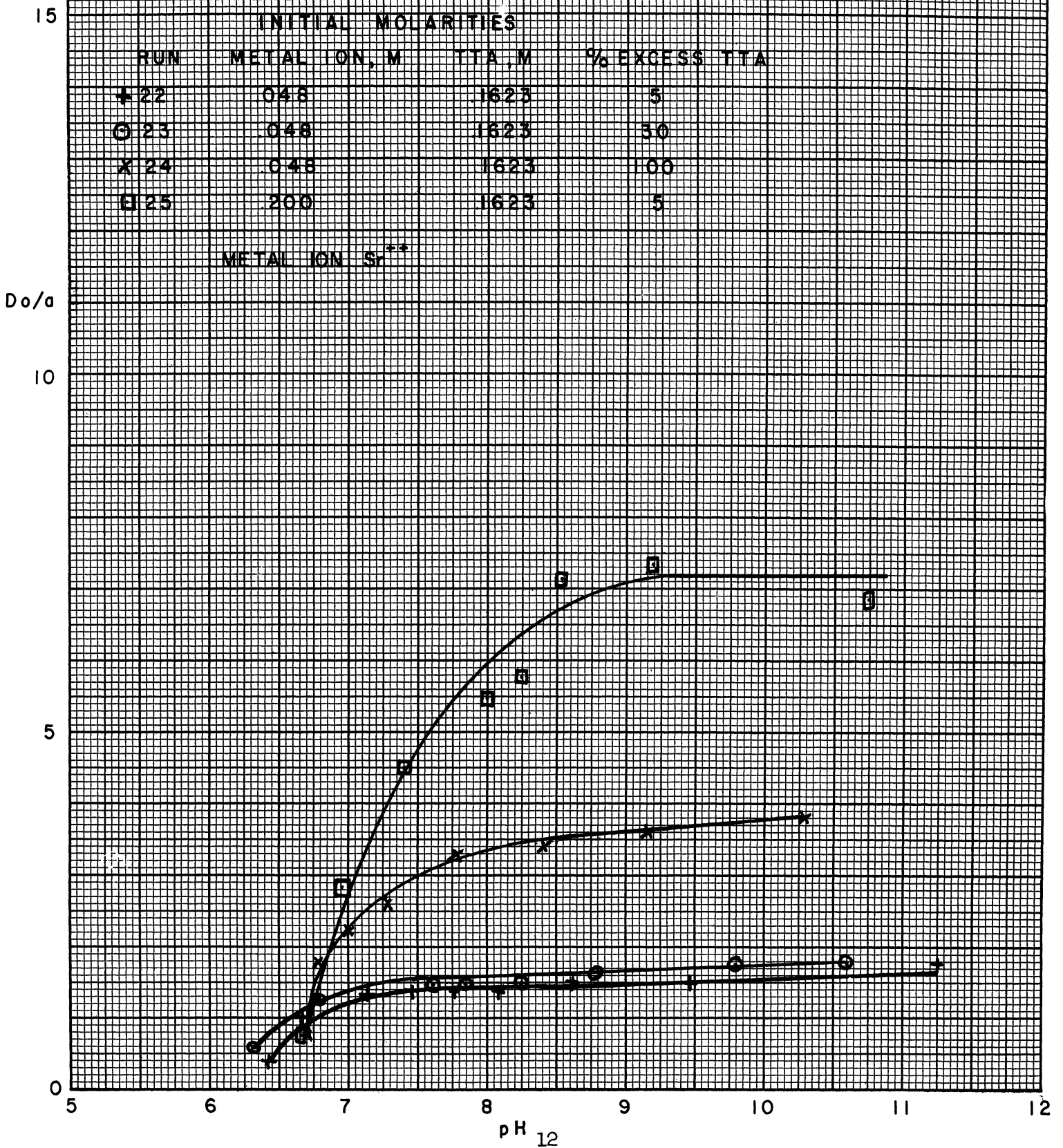
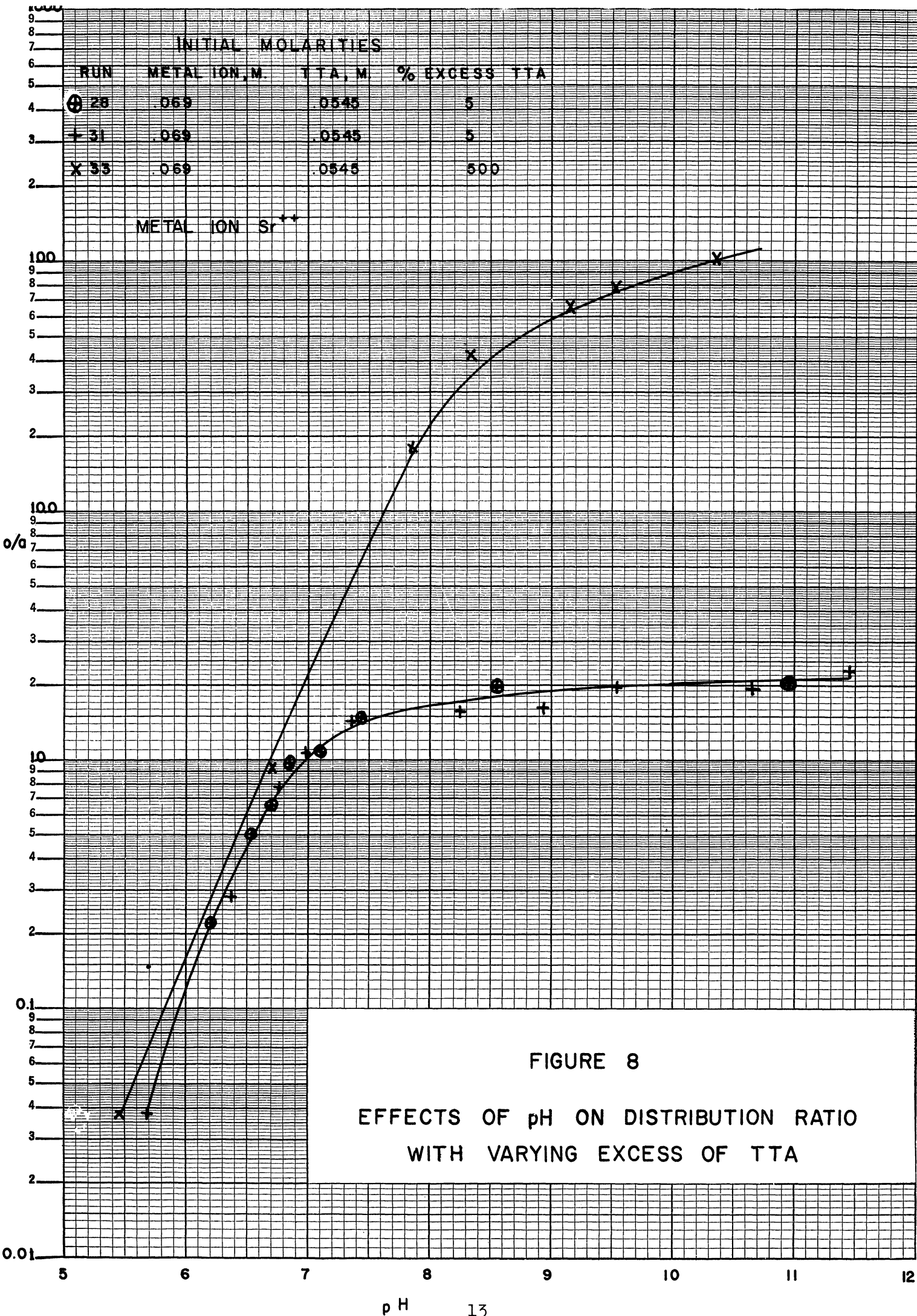


FIGURE 7

EFFECTS OF pH ON DISTRIBUTION RATIO
WITH VARYING EXCESS OF TTA



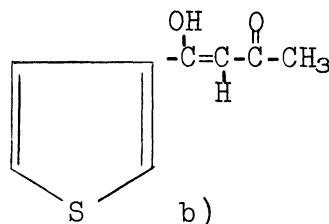
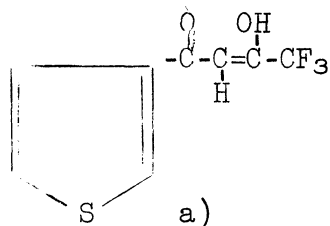


IV THEORY AND DATA CORRELATION OF CHELATION - SOLVENT EXTRACTIONA. Chelating Agent

Salicylaldehyde and thenoyltrifluoroacetone (TTA) were the chelating agents tried. Only the TTA was found to be capable of chelating the desired ions and forming organic soluble compounds.

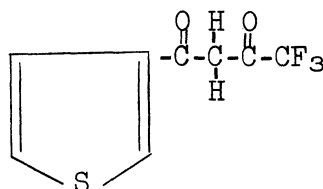
TTA has been found to exist in four forms by a number of workers (6), (7), (8), (9), (10), and (11). These forms are given below.

1.



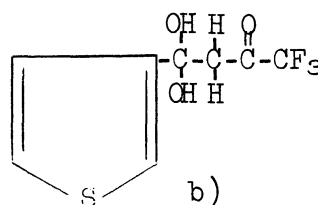
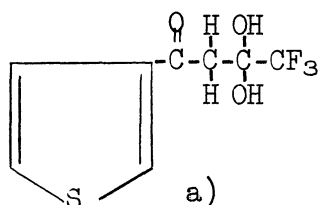
Enol Form

2.



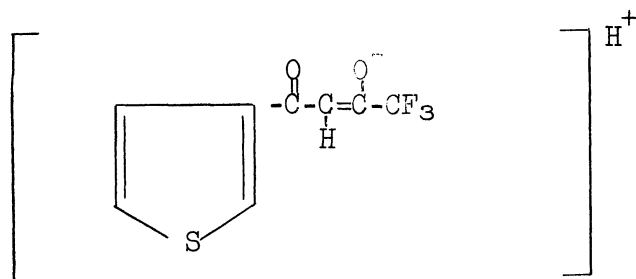
Keto Form

3.



Keto Hydrate Form

4.



Enolate

In the organic phase, the TTA exists mainly in the enol form (1) with its two resonant forms. In the aqueous phase, the keto and keto hydrate (3) A and B forms both exist with the A form predominating due to steric hindrance. The quantities of TTA in an aqueous phase in

equilibrium with an organic phase will be very low because of the low solubility of TTA in aqueous media. The amount of keto-hydrate (III) present increases from a pH of 4 to a pH of 9. Above a pH of 8 the enolate form (IV) predominates. It is the enolate form that is most effective in forming the covalent enolate compounds.

Above pH of 7 to 7.5 the TTA molecule hydrolyzes, this forming two, acetyl-thiophene and trifluoroacetic acid. Hydrolysis essentially destroys TTA, so that no more chelation compounds can be formed.

B. Theory and Correlation of Data

Ionic bonds of a metal ion must be tightly complexed if the ion becomes soluble in organic solvents. The proper chelating agent accomplishes this complexing by forming covalent bonds with the metal ion.

A reaction giving such bonding could be written as:

$M_{(w)}^{+2} + 2 HK_{(o)} \rightleftharpoons MK_2(o) + 2 H_{(w)}^+$ with $M_{(w)}^{+2}$ being a divalent metal ion preferentially soluble in the aqueous phase, $HK_{(o)}$ the enol form of the chelating agent preferentially soluble in organic solvents, $MK_2(o)$ the chelate compound formed that is soluble in the organic phase, and H^+ the hydrogen ion liberated by the reaction.

An equilibrium constant for such a reaction is given as

$$(2) \quad K = \frac{a_{MK_2} a_{H^+}^2}{a_{M^{+2}} a_{HK}^2}$$

This equation can be expressed in activity coefficient form as shown in

$$(3) \quad K = \frac{[M_{MK_2}]}{[M^{+2}][HK]^2} \cdot \frac{\gamma_{MK_2} a_{H^+}^2}{\gamma_{M^{+2}} \gamma_{HK}^2}$$

The distribution ratios are usually expressed in terms of concentrations rather than activity so if

$$(4) \quad \frac{D_O}{A} = \frac{[MK_2]_{(o)}}{[M^{+2}]_{(w)}}$$

then

$$(5) \quad K = \frac{\frac{D_O}{A}}{[HK]^2} \cdot \frac{\gamma_{MK_2} a_{H^+}^2}{\gamma_{M^{+2}} \gamma_{HK}^2}$$

the logarithmic form of equation (5) is convenient for plotting purposes so that with $\text{pH} = -\log a_{\text{H}^+}$, this can be rewritten as:

$$(6) \quad \log K = \log \frac{D_0}{A} - 2 \log [\text{HK}] - 2 \text{pH} + \log \frac{\gamma_{\text{MK}_2}}{\gamma_{\text{M}^{2+}} \gamma_{\text{HK}^2}}$$

Activity coefficients for ions in aqueous solution are given by Keilland (12) and for TTA in benzene by King and Reas (8). No activities have been determined for TTA in MIBK solution so that equation (6) cannot be used to give precise values for K. However, assuming the activity coefficients to be unity, concentrations can be used to give qualitative K's. In Table III some K values are presented.

TABLE III

K Values for Cs^+ , Sr^{++} , and Ba^{++} in TTA - Methylisobutylketone
Phase Volumes, ml.

Metal Ion	Organic	Aqueous	TTA, M.	pH	D_0/a	K
Cesium*	9.0	13.0	0.812	6.96	.060	1.2×10^{-8}
	8.9	18.4	0.806	7.06	.173	1.9×10^{-8}
	8.8	23.3	0.808	7.14	.176	1.6×10^{-8}
	Average					1.5×10^{-8}
Strontium**	19.9	11.8	.0436	6.20	0.22	4.2×10^{-11}
	19.8	12.7	.0302	6.52	0.51	6.5×10^{-11}
	19.7	13.1	.0272	6.68	0.66	3.9×10^{-11}
	19.6	13.5	.0225	6.84	0.96	4.0×10^{-11}
	Average					4.7×10^{-11}
Barium***	50	20.0	.0373	6.48	0.30	2.4×10^{-11}
	49.9	22.9	.0291	6.78	0.80	2.6×10^{-11}
	49.8	24.8	.0253	7.00	1.34	2.1×10^{-11}
	Average					2.3×10^{-11}

* Data from run 36. Initial conditions were Aq. Vol. 5 ml., Org. Vol. 9.0 ml., Cs^+ 0.107 M, TTA 0.8175 M.

** Data from run 28. Initial conditions were Aq. Vol. 7.9 ml., Org. Vol. 20.0 ml., Sr^{++} 0.069 M, TTA 0.0545 M.

*** Data from run 18. Initial conditions were Aq. Vol. 5.0 ml., Org. Vol. 50 ml., Ba^{++} 0.200 M, TTA 0.0545 M.

These K values were not determined at the same ionic strengths, and they can be used, therefore, as order of magnitude values only when different runs are compared. Since hydrolysis of the TTA would make K values unreliable none were computed for pH values exceeding 7.1. A qualitative comparison of K values for Sr^{++} and Ba^{++} would indicate that separation of the two by chelation-solvent extraction might be difficult, and this is verified by Figures 1 and 2.

A plot of $\log K$ versus pH would give a straight line which should agree with the data very closely between pH's of 5 to 7. The influence of high concentrations of metal ion as shown in Figure 7 cannot be correlated by the theory presented. This phenomena will have to be investigated further.

C. Hydrolysis of TTA

Zebroski (13) measured rates of hydrolysis for TTA at various pH's. His equation takes the form:

$$\frac{1}{t} \ln \left[\frac{E_{\bar{n}0}}{E_{\bar{n}t}} \right] = k'f(\text{OH}^-)$$

where:

$E_{\bar{n}0}$ = Evol form of TTA present initially.

$E_{\bar{n}t}$ = Evol form of TTA present after time t.

t = Time in hours.

$k'f(\text{OH}^-)$ = A factor determined experimentally.

Table IV below summarizes the experimental data and gives the ratio of $\frac{E_{\bar{n}0}}{E_{\bar{n}t}}$ for various pH values.

TABLE IV

Hydrolysis of TTA as a function of Ph

$(\text{OH}^-) \text{ M}$	$k'f (\text{OH}^-)$	$\frac{E_{\bar{n}0}}{E_{\bar{n}t}}^*$
1.04	.271 x 2 hrs.	1.72
0.105	.0482 x 2	1.11
0.0105	.0330 x 2	1.068
0.001	.0324 x 2	1.067
pH 9.44	.0767 x 2	1.17
pH 8.42	.0245 x 2	1.050
pH 6.99	.0090 x 2	1.018

* Time basis 2 hours which approximates the time required for a run.

It is evident from the above table that hydrolysis will be high for any pH above 7.0, and that TTA would be too expensive as a chelating agent for basic reactions. It is also evident that hydrolysis will influence the distribution ratios of a metal ion if reaction times are long or pH values above 7.0. Figures 1 through 8 indicate that where the excess of TTA is large curvature in the $\log D_0/a$ versus pH relationship occurs at a higher pH than with small TTA excesses. This is exactly the results one would expect according to the hydrolysis rates of TTA.

Two conclusions were reached in the light of these rates:

- (1) TTA cannot be used in a continuous process where pH ranges above 7.0, and
- (2) hydrolysis accounts for the curve in the D_0/a versus pH plot, whereas theory would predict a straight line.

PROCESS APPLICATIONA. Chelation of Other Fission Products

The objective of this work was to develop a process for the selective removal of Sr-90 and Cs-137 from gross fission products. It is necessary to know at what pH the remaining fission products extract to make the desired separations.

The fission product spectra can be divided roughly into two groups: the rare earths, and the transition elements, such as Zr, Nb, Ru, and Rb. Cesium and barium are the rare earth groups while Sr is in the transition group. In addition to the fission products there will be corrosion products from the lines and vessel walls of the process equipment. Corrosion products will consist mostly of Fe, Cr, and Ni.

Bronaugh and Suttle (14) have reported data on the rare earths which indicate the group will extract between pH's of 2.4 to 4.5. One exception is Scandium which apparently chelates at a pH of 1.2.

Zebroski (13) reported the chelation of Fe, Cr, and Al at pH values of 1, 3, 7, and 2.2. Chelation of gross fission products at a pH of 2.4 would remove Sc, Fe, Al, Zr, and probably all of the transition group except Sr. A chelation at a pH of 4.5 would remove the remainder of the rare earths and Al, but it would leave Ba and Cs in the aqueous phase.

B. Discussion of Proposed Flowsheet

Figure 9 is a tentative flowsheet illustrating how the chelation solvent extraction could be used for an easy separation of Sr-Ba and Cs from gross fission products.

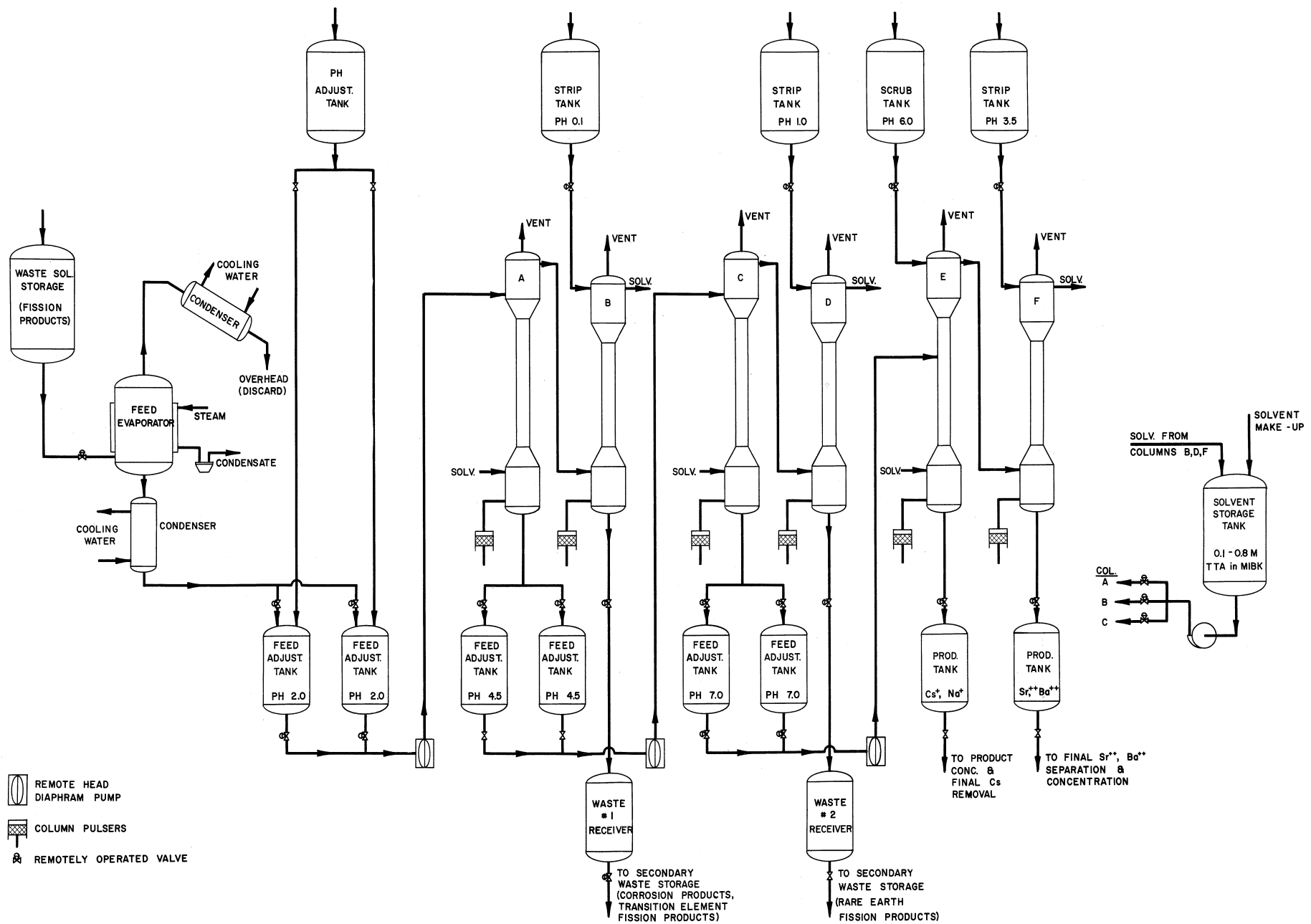
The starting material is assumed to be aqueous waste solution from a fuels processing plant such as Purex. The material in the waste is assumed to have been stored for a minimum period of 120 days to one year before being processed.

The solution is concentrated to adjust the fission product concentration to the desired range and to boil off nitric acid. The solution is then cooled and routed to feed adjustment tanks. Here the pH is adjusted to 2.0 to 2.5, and the solution is then pumped to the first solvent extraction contactor A. The aqueous solution is contacted countercurrently with TTA dissolved in Methylisobutylketone, and the transition elements and corrosion products are extracted into the organic phase. Organic phase is stripped of metal ions in a stripping column (B) by a highly acidic scrub stream while the MIBK-TTA solution is routed to solvent storage, and the solution of metal ions goes into Waste Receiver #1 for disposal back into the Purex waste facilities.

Aqueous phase from (A) contactor is adjusted to a pH of 4.5

FIGURE 9

FISSION PRODUCT SEPARATION BY CHELATION SOLVENT EXTRACTION



and routed to other contactors where the operations described above are repeated. Contactors C and D remove the rare earths from the aqueous stream while contactors E and F make the final split between Sr-Ba and Cs. The Ba coming off with the Sr is not radioactive because of the long decay time prior to processing. The Sr-Ba product will thus be essentially a pure beta emitter. The Cs product will contain cold Rb plus any Na^+ that was used in the process.

Further efforts should be directed toward final splits between Sr and Ba and Cs from Rb and Na. In order to make the process consistent and turn out a high purity product, these final splits should be done preferably by solvent extraction.

The waste solutions from the process that are collected in Waste Receivers #1 and #2 now contain no long-lived activity and this can be discarded in approximately six years by conventional means.

The solvent used in the process can be recycled repeatedly if pH values are maintained at 7.0 or below. This reuse should hold solvent costs to a minimum.

Production costs of Cs^{137} and Sr^{90} from such a process can be estimated from costs of similar solvent extraction processes extracting fissionable materials from fission products. Estimated costs by such means will vary considerably with the assumed processing rates. One such study (15), gives costs for processing Pu-239 at \$6.30, \$3.60, \$2.70, and \$2.20 per gram for 25, 50, 75 and 100% production capacity. Pu-239 production basis was 800 kg/year at 100% capacity. Approximately equal quantities of 800 kg/yr of gross fission products would be formed. This corresponds to about 20 kg of Sr^{90} and 29 kg of Cs^{137} or 4×10^6 and 2.3×10^6 curies respectively. Processing costs on this basis would be extremely small on a dollars per curie basis. Production rates of these magnitudes cannot be reached for some years in the future. Present output of fission products would be on the order of several hundred thousand curies of combined Sr^{90} and Cs^{137} . Processing costs at these rates would be approximately 7-10¢ per curie.

VI FUTURE PROBLEMS

The problems remaining in this process are:

1. Final separation methods for Sr and Ba and Cs and Rb have not been developed.
2. A chelating agent having the reaction characteristics of TTA, but possessing more stability in basic solutions would be very valuable. Such an agent would probably make possible a solvent extraction separation between Sr and Ba, Cs and Rb, and it would reduce costs of the process.
3. Refinements should be made of the methods used to determine distribution ratios and to reduce or eliminate the errors present here.
4. The exact pH versus D_0/a is not known for some of the transition elements. More data is needed to substantiate the meager data currently available.
5. The action of gross fission products as a whole are not accurately known. Complete process runs should be made on simulated gross fission products.
6. Costs are unknown for such a process. A cost per curie figure for such a process must be determined.
7. Pilot plant data are not available. If such a process were incorporated into a plant, data using larger scale equipment would need to be obtained.

It is believed, however, that the work done thus far has demonstrated the overall feasibility of the process, and that these problems can be answered satisfactorily.

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TABLE I

SUMMARY OF RUN CONDITIONS AND PURPOSES

Run No.	TTA		Metal	Tracer		Conc. NaOH	X.S. TTA	Purpose		
	ml.	Conc.		ml.	M					
17	49.0	0.1090M	Ba ⁺⁺	10.0	0.20M	Ba ¹⁴⁰	2.0	0.2N	33%	Effect of varying concentration of TTA on barium chelation.
18	50.0	0.0545M	Ba ⁺⁺	5.0	0.20M	Ba ¹⁴⁰	2.0	0.2N	33%	Effect of varying concentration of TTA on barium chelation.
19	20.0	0.1635M	Ba ⁺⁺	7.0	0.20M	Ba ¹⁴⁰	1.0	0.2N	33%	Effect of varying concentration of TTA on barium chelation.
22	20.0	0.1635M	Sr ⁺⁺	31.0	0.048	Sr ⁸⁹	0.5	0.2N	5%	Effect of varying excesses of TTA on strontium chelation.
23	20.0	0.1635M	Sr ⁺⁺	25.0	0.048	Sr ⁸⁹	2.0	0.2N	30%	Effect of varying excesses of TTA on strontium chelation.
24	20.0	0.1635M	Sr ⁺⁺	16.0	0.048	Sr ⁸⁹	2.0	0.2N	100%	Effect of varying excesses of TTA on strontium chelation.
25	20.0	0.1635M	Sr ⁺⁺	7.7	0.20	Sr ⁸⁹	2.0	0.2N	5%	Comparison with #22 for more concentrated strontium.
28	20.0	0.0545M	Sr ⁺⁺	7.9	0.069	Sr ⁸⁹	2.0	0.2N	5%	Effect of low concentration of TTA
31	40.0	0.0545M	Sr ⁺⁺	15.0	0.069	Sr ⁸⁹	3.0	0.2N	5%	Rerun of #28
33	40.0	0.0545M	Sr ⁺⁺	2.6	0.069	Sr ⁸⁹	4.0	0.2N	500%	Test of 500% excess on Sr chelation.
34	80.0	0.0545M	Cs ⁺	6.8	0.107	Cs ¹³⁷	1.0	0.2N	100%	Test of various TTA concentrations and excesses; for cesium chelation.
35	30.0	0.1635	Cs ⁺	7.6	0.107	Cs ¹³⁷	1.0	0.2N	100%	Test of various TTA concentrations and excesses; for cesium chelation.
36	9.0	0.8175	Cs ⁺	11.5	0.107	Cs ¹³⁷	1.0	0.2N	100%	Test of various TTA concentrations and excesses; for cesium chelation.
37	9.0	0.8175	Cs ⁺	3.8	0.107	Cs ¹³⁷	1.5	0.2N	500%	Test of various TTA concentrations and excesses; for cesium chelation.
44	40.0	0.0545	Cs ⁺	6.0	0.107	Cs ¹³⁷	2.0	0.2N	24%	Test of various TTA concentrations and excesses; for cesium chelation.
46	5.0	0.8175	Ba ⁺⁺	6.0	0.0342	Ba ¹⁴⁰	4.0	0.2N	1000%	See Note 1.
47	6.0	0.8175	Cs ⁺	7.2	0.107	Cs ¹³⁷	2.0	0.2N	135%	See Note 1.
48	7.0	0.8175	Sr ⁺⁺	8.4	0.069	Sr ⁸⁹	1.0	0.2N	392%	See Note 1.
50	15.0	0.8175	F ⁻	18.0	Note 2	Ba ¹⁴⁰	8.0	0.2N	40%	Attempt at separations with all 3 present as in the reactor waste stream.
51	15.0	0.8175	F ⁻	18.0	Note 2	Cs ¹³⁷	2.0	0.2N	40%	Attempt at separations with all 3 present as in the reactor waste stream.

TABLE I

SUMMARY OF RUN CONDITIONS AND PURPOSES

Run No.	TTA		Metal		Tracer	Conc.	M	Conc.	X.S.	Purpose
	ml.	Conc.	ml.	ml.						
52	15.0	0.8175	F ¹	18.0	Note 2	Sr ⁸⁹	2.0	0.2N	40%	Attempt at separations with all 3 present as in the reactor waste stream
53	15.0	0.8175	F ¹	18.0	Note 2	Ba ¹⁴⁰	8.0	0.2N	40%	Repeat of #50.
54	15.0	0.8175	F ¹	18.0	Note 2	Sr ⁸⁹	2.0	0.2N	40%	Repeat of #52.
55	20.0	0.1635	F ¹¹	40.0	Note 3	Sr ⁸⁹	1.0	0.2N	-	Attempt at separation of Sr and Ba by using excess feed instead of excess TTA.
56	15.0	0.1635	F ¹¹	30.0	Note 3	Ba ¹⁴⁰	3.0	0.2N	-	Attempt at separation of Sr and Ba by using excess feed instead of excess TTA.
59	60.0	0.0545	F ¹¹	10.6	Note 3	Sr ⁸⁹	2.0	0.2N	50%	Effect of 50% excess and weak TTA on the Sr-Ba separation.
66	60.0	0.0545	F ¹¹	10.6	Note 3	Ba ¹⁴⁰	3.0	0.2N	50%	Effect of 50% excess and weak TTA on the Sr-Ba separation.

Note 1: From data on the products of slow neutron fission it was calculated that the waste stream from an atomic reactor would have a composition ratio of:

0.0342 moles Ba (NO₃)₂
 0.0690 moles Sr (NO₃)₂
 0.1070 moles Cs (Cl)
 per liter of solution.

These concentrations are used in all subsequent runs.

Note 2: F¹ is a feed composition of that given in Note 1.

Note 3: F¹¹ is a feed consisting only of Ba(NO₃)₂, and Sr(NO₃)₂ in the concentrations given in Note 1. This feed is used since it can be seen from the data that Sr and Ba would be separated easily from the total waste stream and an attempt was being made in these runs to separate Ba and Sr from each other.

TABLE II
SUMMARY OF RUN DATA

DISTRIBUTION RATIO AS A FUNCTION OF pH AT SELECTED INITIAL AQUEOUS PHASE CONCENTRATIONS OF
BARIUM, CECROTIN, AND CURTUM AND INITIAL ORGANIC PHASE CONCENTRATIONS OF TTA

[illegible]

