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DISTRIBUTION OF URANIUM AT LOW ACID - LOW URANIUM
CONCENTRATIONS INTO 4-1/2 PERCENT TBP-AMSCO

R. L. Andelin, E. L. Anderson, W. H. McVey

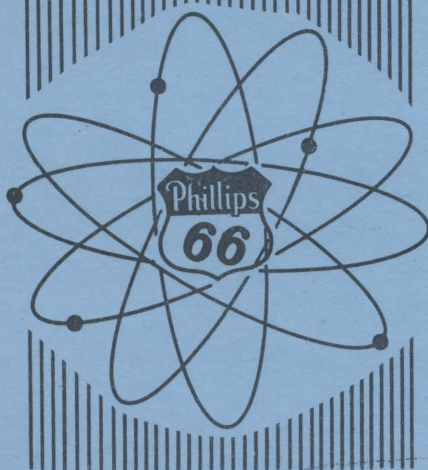
August 23, 1956

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A B S T R A C T

Acid free uranyl nitrate hexahydrate was prepared and its distribution into 4-1/2 percent TBP-Amsco 125-90W in the aqueous concentration range 1 gram per liter to 30 grams per liter was measured. Acid concentrations were varied from 0.00M added acid to 0.06M added acid. Uranium distribution was found to be strongly dependent on nitric acid and uranium concentrations as expected. Hydrolysis effects under these conditions were studied. No attempt was made to measure nitric acid distribution. A value of 171.8 was found for the thermodynamic equilibrium constant in the reaction for complexing of uranyl nitrate by TBP.

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by

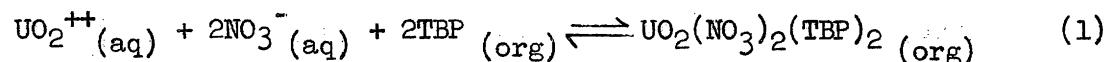
R. L. Andelin
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I. INTRODUCTION

Calculations⁽¹⁾ on the distribution of uranium at low acid, low uranium concentrations into 4 1/2 percent tributyl phosphate-kerosene indicated considerable dependency of distribution ratios on the acid and uranium concentrations. The calculations were based upon distribution data obtained from uranium tracer experiments.⁽²⁾ The work presented in this report was performed to extend the data of reference (2) into the macro uranium range.

II. THEORETICAL

The fundamental equation for uranium extraction as developed by Moore⁽³⁾ is:



The equilibrium constant for this reaction is,

$$\frac{(\text{UO}_2(\text{NO}_3)_2(\text{TBP})_2)}{(\text{UO}_2^{++})(\text{NO}_3^-)^2(\text{TBP})^2} = K \quad (2)$$

Rewriting (2) as a distribution coefficient,

$$E_A^\circ = \frac{U_O}{U_A} = K (\text{NO}_3^-)^2 (\text{TBP})^2 \quad (3)$$

Since most of the uranium distributes in the aqueous phase under stripping column conditions, the molar concentration of uncomplexed TBP in the organic phase may be considered essentially constant. Equation (3) may now be written:

$$\frac{U_O}{U_A} = K' (NO_3^-)^2 \quad (4)$$

In a system with no added acid where,

$$(NO_3^-) = 2U_A, \quad (5)$$

$$U_O = K'' (U_A)^3. \quad (6)$$

Taking logarithms of both sides,

$$\log U_O = 3 \log U_A + \log K''. \quad (7)$$

Therefore, a log-log plot for this system should yield a straight line with a slope of 3.

In a system with added acid where:

$$(NO_3^-) = 2U_A + (H_A^+) \approx (H_A^+), \quad (8)$$

and (H_A^+) is small, equation (4) may be written,

$$\frac{U_O}{U_A} = K' (H^+)^2. \quad (9)$$

Then, since the acid concentration is essentially constant,

$$\frac{U_O}{U_A} = K'', \quad (10)$$

and, taking logarithms,

$$\log U_O = \log U_A + \log K''. \quad (11)$$

Therefore, in the region where the aqueous acid concentration is much greater than the aqueous uranium concentration a log-log plot should give straight line with a slope of 1. For the equilibria studied in this report, a straight line with slope of 1 would occur only at extremely low uranium concentrations, since the highest acid concentration studied was only 0.06M.

III. EXPERIMENTAL

A. Preparation of Acid Free Uranyl Nitrate Hexahydrate.

1. Theoretical. The work of Wendolkowski and Kirsli⁽⁴⁾ was used to demonstrate that essentially acid free uranyl nitrate hexahydrate could be prepared by a vacuum technique. They studied the thermal decomposition of uranyl nitrate hexahydrate at various temperatures by observing the equilibrium pressure over the salt as the water of hydration was gradually removed. They found that in the transition from the hexahydrate to the trihydrate state at 41.8° C, only 4×10^{-4} moles of nitric acid were lost per mole of uranium. Their starting material was Mallinckrodt CP Grade uranyl nitrate hexahydrate. The first sample of water vapor withdrawn from their equilibrium system contained approximately 2×10^{-4} moles of nitric acid per mole of uranium, or about half of the total amount of nitric acid removed. From these data it is obvious that little free nitric acid is present in the original salt, and that what is there can be essentially removed by one or two withdrawals of the vapor phase from the equilibrium system.

2. Apparatus and Experimental Method. The apparatus used for preparing acid free uranyl nitrate hexahydrate was simply a dessicator jar which contained a thermometer, and a Wallace and Tiernan absolute pressure gauge connected to an oil vacuum pump. The jar was first evacuated to a pressure of 0.05 mm of mercury at 23.0° C and the system sealed off. After sixteen hours the pressure was 0.3 mm. A quantity of Bakers A. R. uranyl nitrate hexahydrate crystals was ground to a powder with a porcelain mortar and pestle. Approximately 44 grams of the powder were spread evenly over a watch glass and placed inside the dessicator. The system was then pumped down for 10 minutes to a pressure of 1.40 mm mercury at 23° C and sealed off. The equilibrium pressure after three days was 6.68 mm of mercury. (From Wendolkowski's⁽⁴⁾ equation for this system, $\log P = \frac{-2396}{T} + 1.010$, the equilibrium pressure at 23° C should be 8.32 mm.) The vapor phase was removed by pumping down again for 15 minutes to 1.5 mm of mercury. After another three-day equilibration the vapor phase was again removed, and the salt was considered essentially acid free. A little over 0.3 of a mole of water was lost per mole of uranium.

B. Distribution Measurements

Uranium distribution data were obtained by contacting an aqueous phase containing uranium with an organic phase containing tributyl phosphate and then analyzing each phase for uranium content.

The organic phase consisted of 4-1/2 percent double distilled TBP in silica gel-treated Amsco 125-90W. The Amsco 125-90W yielded a zero chromyl chloride number⁽⁵⁾ after the silica gel treatment. The solvent was washed once with equal volumes of 0.5M sodium carbonate solution, twice with equal volumes of water, and then centrifuged prior to use.

Acid free uranyl nitrate hexahydrate in distilled water (pH 7.5) with known amounts of nitric acid added were used to make up the aqueous phase.

Twenty-five milliliters of each phase were contacted vigorously for 3 to 4 minutes in separatory funnels at 20°C. The organic phase was centrifuged prior to analysis.

Two entirely separate experiments were conducted, each beginning with the preparation of acid free uranyl nitrate hexahydrate. In the first experiment, three acid concentrations were used. These were 0.00M HNO₃, 0.04M HNO₃ and 0.06M HNO₃. Hydrolysis calculations were based on these additions of acid. In the second experiment, 0.02M nitric acid was included in the above series, and pH measurements were made on each aqueous sample. Experimental hydrogen ion concentrations were then used in the hydrolysis calculations. These two separate sets of experiments are referred to as the 3-acid and 4-acid experiments in the discussion to follow.

C. Analytical Methods.

Aqueous uranium standard solutions were analyzed gravimetrically as U₃O₈. The detailed method of analysis is described in IDO-14316.⁽⁶⁾

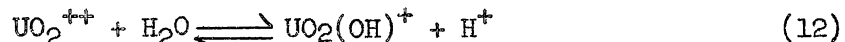
Organic uranium samples were analyzed directly by an ultra violet spectrophotometric technique. The details of this method are described in IDO-14349.⁽⁷⁾ The direct analysis of organic samples was possible because of the specially purified Amsco-125-90W used. A few of the organic analyses were checked by an isotopic dilution method which is described in IDO-14301⁽⁸⁾ and IDO-14366.⁽⁹⁾ Organic uranium samples containing less than 1 microgram per milliliter uranium were analyzed by a direct fluorophotometric method. Nine determinations were made on each sample. This method is also described in IDO-14316.⁽⁶⁾

No acid analyses other than pH measurements were made on the uranium solutions.

IV. DISCUSSION AND RESULTS

A. Effect of Hydrolysis.

The equation for hydrolysis of uranyl nitrate is written by Kolthoff,⁽¹⁰⁾



By polarographic techniques he determined the equilibrium constant to be 8.1×10^{-5} . Sutton⁽¹¹⁾ determined a similar equilibrium constant, but showed oxygen bridging rather than hydroxyl addition as the mechanism. In this study the equation given by Kolthoff was used.

The extent of uranyl nitrate hydrolysis was calculated for aqueous samples in each of the two experiments. In the first experiment, the 3-acid experiment, the added acid and the hydrolysis constant were used to calculate the hydrogen ion concentrations. These data are presented in Table 1. Table 2 presents the data from the second experiment, the 4-acid experiment, in which pH measurements were used for calculating hydrolytic species. For these relatively dilute solutions, concentrations rather than activities were used in the calculations. A plot of the ratio of hydrolytic species to uranyl ion as a function of total uranium with nitric acid concentrations as the parameter is shown in Figure 1. Data from Tables 1 and 2 are used in the plot. The solid points represent data from Table 2. From the graph it may be seen that hydrolysis is negligible except in the system with no added acid. In this system maximum hydrolysis is of the order of 20 percent for a $1 \times 10^{-3}M$ uranium solution.

B. Distribution Data.

The uranium distribution data for each experiment are presented in Table 3. Both the original values of U_A and the values of U_A corrected for hydrolysis are listed. The corrected values are simply the concentrations of the extractable uranyl ion in each sample. Figure 2 is a plot of the data. The open and solid points represent data from the 3-acid and 4-acid experiments, respectively. The line for the zero acid experiment is drawn with a slope of three.

C. Statistical Treatment of Data.

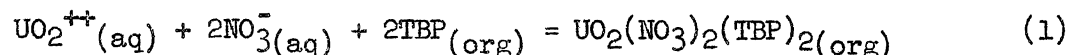
The experimental data were statistically analyzed to determine an empirical relationship between U_O and U_A at each acid concentration. The two experimental runs were analyzed separately and then compared for consistency with regard to the parameters estimated. In determining the relationship, standard regression analyses were made in which U_O was assumed to be the dependent variable. The usual assumptions of normality with regard to the deviations about the regression curve were made. The tests of significance were conducted at the 0.05 significance level and failed to indicate any real difference between corresponding relations for the two series of experiments. Therefore, the data for a given acid concentration were pooled to yield a single curve in each case. The limits are expressed as 95 percent confidence limits. In the case of zero acid level, the two points at the extreme lower concentrations were believed to be suspect due to the lack of a reliable analytical method for uranium at concentrations in this range. Hence the curve at this acid level was fit with and without these two points. A list of the equations are presented in Table 4.

It is interesting to note that the exponent (2.90) of U_A in the equation excluding the two suspect points agrees excellently with the theoretical value of 3.00 derived above and shown in Equation 7.

The distribution data of Table 3 are replotted with their limits in Figures 3, 4, 5, and 6 for the 0.00M, 0.02M, 0.04M, and 0.06M acid levels, respectively.

D. Determination of the Thermodynamic Equilibrium Constant.

In dilute solutions of low ionic strength, it is possible to evaluate the thermodynamic equilibrium constant of a reaction from a limiting slope plot of the log of the observed equilibrium constant as a function of the square root of the ionic strength. The derivation of this for Equation (1) is as follows:



The observed equilibrium constant, Equation 2 is,

$$K_{(obs)} = \frac{(UO_2(NO_3)_2(TBP)_2)}{(UO_2^{++})(NO_3^-)^2(TBP)^2} \quad (2)$$

The thermodynamic equilibrium constant is,

$$K = K_{(obs)} \cdot \frac{\gamma_{UO_2(NO_3)_2(TBP)_2}}{\gamma_{UO_2^{++}} \gamma_{NO_3^-}^2 \gamma_{TBP}^2} \quad (13)$$

where the γ 's are the activity coefficients of the respective ions. Then, since the activities of the uncharged molecules are relatively small compared to charged ions, let,

$$\frac{\gamma_{UO_2(NO_3)_2(TBP)_2}}{\gamma_{TBP}^2} = 1$$

and, therefore,

$$K_{(obs)} = K \gamma_{\pm}^3 UO_2(NO_3)_2 \quad (14)$$

In logarithmic form,

$$\log K_{(obs)} = \log K + 3 \log \gamma_{\pm} UO_2(NO_3)_2 \quad (15)$$

From the Debye-Hückel theory, (12)

$$\log \gamma = \frac{-A |Z_+ Z_-| \sqrt{\mu}}{1 + B a_i \sqrt{\mu}} \quad (16)$$

where A and B are constants, Z is the ion charge, μ is the ionic strength and a_i is the effective diameter of the ions in solution. Substituting the value of $\log \gamma$ from Equation 16 into Equation 15, and using 0.5042 and 0.3273×10^8 as the constants for A and B respectively, the final form of the equation is,

$$\log K(\text{obs}) = \log K - \frac{+ 3.025 \sqrt{\mu}}{1 + 0.327 \times 10^8 a_i \sqrt{\mu}} \quad (17)$$

Although the Debye-Hückel expression for activity coefficients is valid only at ionic strengths less than 0.01, it is felt that the ionic strengths listed here are low enough to warrant an evaluation of the thermodynamic K.

Table IV lists the observed values of K and the calculated values of $\sqrt{\mu}$ for the 3-acid and 4-acid experiments. A constant distribution coefficient for nitric acid of 0.01 was assumed in determining $K(\text{obs})$. Ionic strengths were calculated from the expression,

$$\mu = 1/2 \sum_i m_i Z_i^2$$

where m_i is the molality of each ion. For these calculations, however, molar concentrations were used.

The data of Table IV were used in determining values for $\log K$ and a_i in Equation 17. The method of least squares was used with the problem being programmed and run on the 650 IBM Digital Computer. The values obtained were 171.8 for K, and 4.59×10^{-8} cm for a_i . The value for a_i is well within the range of effective diameters listed by Kielland⁽¹³⁾ for singly and doubly charged inorganic ions.

Figure 7 is a plot of the data of Table IV. The solid line was calculated from Equation 17 using the values for K and a_i listed above. The first three points at the zero acid level in both the 3-acid and 4-acid experiments were omitted for statistical reasons in the calculation of K and a_i . The reason for the large discrepancies observed with the zero-acid points is not known. However, it is believed that the original x-y data are accurate and should be used in column calculations as given in Table III, i.e. $U_{A(\text{uncorrected})}$ vs. U_0 .

V. ACKNOWLEDGMENTS

The authors acknowledge their indebtedness to the analysts who performed the special analytical work required in this study: Bernice E. Paige, for her adaptation of the ultraviolet spectrophotometric method to organic samples; Paul Goris for the isotopic dilution analyses; and R. B. Regier for the gravimetric uranium analyses. The authors also acknowledge their indebtedness to F. H. Tingey for his statistical treatment of the data and his "least squares" evaluation of K and a_i on the 650 IBM Digital Computer.

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TABLE 1

SUMMARY OF HYDROLYSIS CALCULATIONS
FROM 3-ACID EXPERIMENT

<u>Uranium</u> <u>moles/liter</u>	<u>Added HNO₃</u> <u>moles/liter</u>	<u>(H⁺)*</u> <u>moles/liter</u>	<u>(UO₂(OH)⁺)</u> <u>moles/liter</u>	<u>(UO₂⁺⁺)</u> <u>moles/liter</u>	<u>(UO₂(OH)⁺)</u> <u>(UO₂⁺⁺)</u>
1.99 x 10 ⁻³	0	3.63 x 10 ⁻⁴	3.63 x 10 ⁻⁴	1.627 x 10 ⁻³	2.23 x 10 ⁻¹
5.97 x 10 ⁻³	0	6.56 x 10 ⁻⁴	6.56 x 10 ⁻⁴	5.314 x 10 ⁻³	1.24 x 10 ⁻¹
1.39 x 10 ⁻²	0	1.02 x 10 ⁻³	1.02 x 10 ⁻³	1.288 x 10 ⁻²	7.91 x 10 ⁻²
5.97 x 10 ⁻²	0	2.16 x 10 ⁻³	2.16 x 10 ⁻³	5.754 x 10 ⁻²	3.76 x 10 ⁻²
1.99 x 10 ⁻³	4.0 x 10 ⁻²	4.0 x 10 ⁻²	4.02 x 10 ⁻⁶	1.986 x 10 ⁻³	2.02 x 10 ⁻³
5.97 x 10 ⁻³	4.0 x 10 ⁻²	4.0 x 10 ⁻²	1.21 x 10 ⁻⁵	5.958 x 10 ⁻³	2.03 x 10 ⁻³
1.39 x 10 ⁻²	4.0 x 10 ⁻²	4.0 x 10 ⁻²	2.81 x 10 ⁻⁵	1.387 x 10 ⁻²	2.02 x 10 ⁻³
5.97 x 10 ⁻²	4.0 x 10 ⁻²	4.0 x 10 ⁻²	1.21 x 10 ⁻⁴	5.958 x 10 ⁻²	2.03 x 10 ⁻³
1.99 x 10 ⁻³	6.0 x 10 ⁻²	6.0 x 10 ⁻²	2.7 x 10 ⁻⁶	1.987 x 10 ⁻³	1.36 x 10 ⁻³
5.97 x 10 ⁻³	6.0 x 10 ⁻²	6.0 x 10 ⁻²	8.06 x 10 ⁻⁶	5.962 x 10 ⁻³	1.35 x 10 ⁻³
1.39 x 10 ⁻²	6.0 x 10 ⁻²	6.0 x 10 ⁻²	1.88 x 10 ⁻⁵	1.388 x 10 ⁻²	1.35 x 10 ⁻³
5.97 x 10 ⁻²	6.0 x 10 ⁻²	6.0 x 10 ⁻²	8.06 x 10 ⁻⁵	5.962 x 10 ⁻²	1.35 x 10 ⁻³

*H⁺ concentration calculated from hydrolysis constant.

TABLE 2

SUMMARY OF HYDROLYSIS CALCULATIONS
FROM 4-ACID EXPERIMENT

<u>Uranium</u> <u>moles/liter</u>	<u>Added HNO₃</u> <u>moles/liter</u>	<u>pH</u>	<u>(H⁺)*</u> <u>moles/liter</u>	<u>(UO₂⁺⁺)</u> <u>moles/liter</u>	<u>(UO₂(OH)⁺)</u> <u>moles/liter</u>	<u>(UO₂(OH)⁺)</u> <u>UO₂⁺⁺</u>
5.34 x 10 ⁻³	0	3.45	3.56 x 10 ⁻⁴	4.984 x 10 ⁻³	3.56 x 10 ⁻⁴	7.1 x 10 ⁻²
1.07 x 10 ⁻²	0	3.20	6.31 x 10 ⁻⁴	1.007 x 10 ⁻²	6.31 x 10 ⁻⁴	6.2 x 10 ⁻²
2.65 x 10 ⁻²	0	3.00	1.00 x 10 ⁻³	2.550 x 10 ⁻²	1.00 x 10 ⁻³	3.9 x 10 ⁻²
1.27 x 10 ⁻¹	0	2.50	3.16 x 10 ⁻³	1.238 x 10 ⁻¹	3.16 x 10 ⁻³	2.5 x 10 ⁻²
5.34 x 10 ⁻³	2.0 x 10 ⁻²	1.75	1.78 x 10 ⁻²	5.316 x 10 ⁻³	2.42 x 10 ⁻⁵	4.55 x 10 ⁻³
1.07 x 10 ⁻²	2.0 x 10 ⁻²	1.70	2.00 x 10 ⁻²	1.066 x 10 ⁻²	4.31 x 10 ⁻⁵	4.04 x 10 ⁻³
2.65 x 10 ⁻²	2.0 x 10 ⁻²	1.68	2.09 x 10 ⁻²	2.640 x 10 ⁻²	1.02 x 10 ⁻⁴	3.86 x 10 ⁻³
1.27 x 10 ⁻¹	2.0 x 10 ⁻²	1.64	2.30 x 10 ⁻²	1.266 x 10 ⁻¹	4.45 x 10 ⁻⁴	3.52 x 10 ⁻³
5.34 x 10 ⁻³	4.0 x 10 ⁻²	1.40	3.99 x 10 ⁻²	5.329 x 10 ⁻³	1.08 x 10 ⁻⁵	2.03 x 10 ⁻³
1.07 x 10 ⁻²	4.0 x 10 ⁻²	1.40	3.99 x 10 ⁻²	1.068 x 10 ⁻²	2.17 x 10 ⁻⁵	2.03 x 10 ⁻³
2.65 x 10 ⁻²	4.0 x 10 ⁻²	1.38	4.17 x 10 ⁻²	2.645 x 10 ⁻²	5.14 x 10 ⁻⁵	1.95 x 10 ⁻³
1.27 x 10 ⁻¹	4.0 x 10 ⁻²	1.35	4.48 x 10 ⁻²	1.268 x 10 ⁻¹	2.29 x 10 ⁻⁴	1.81 x 10 ⁻³
5.34 x 10 ⁻³	5.95 x 10 ⁻²	1.23	5.89 x 10 ⁻²	5.333 x 10 ⁻³	7.33 x 10 ⁻⁶	1.38 x 10 ⁻³
1.07 x 10 ⁻²	5.95 x 10 ⁻²	1.23	5.89 x 10 ⁻²	1.069 x 10 ⁻²	1.47 x 10 ⁻⁵	1.38 x 10 ⁻³
2.65 x 10 ⁻²	5.95 x 10 ⁻²	1.22	6.04 x 10 ⁻²	2.647 x 10 ⁻²	3.55 x 10 ⁻⁵	1.34 x 10 ⁻³
1.27 x 10 ⁻¹	5.95 x 10 ⁻²	1.20	6.31 x 10 ⁻²	1.268 x 10 ⁻¹	1.79 x 10 ⁻⁴	1.41 x 10 ⁻³

*H⁺ concentration calculated from pH measurement.

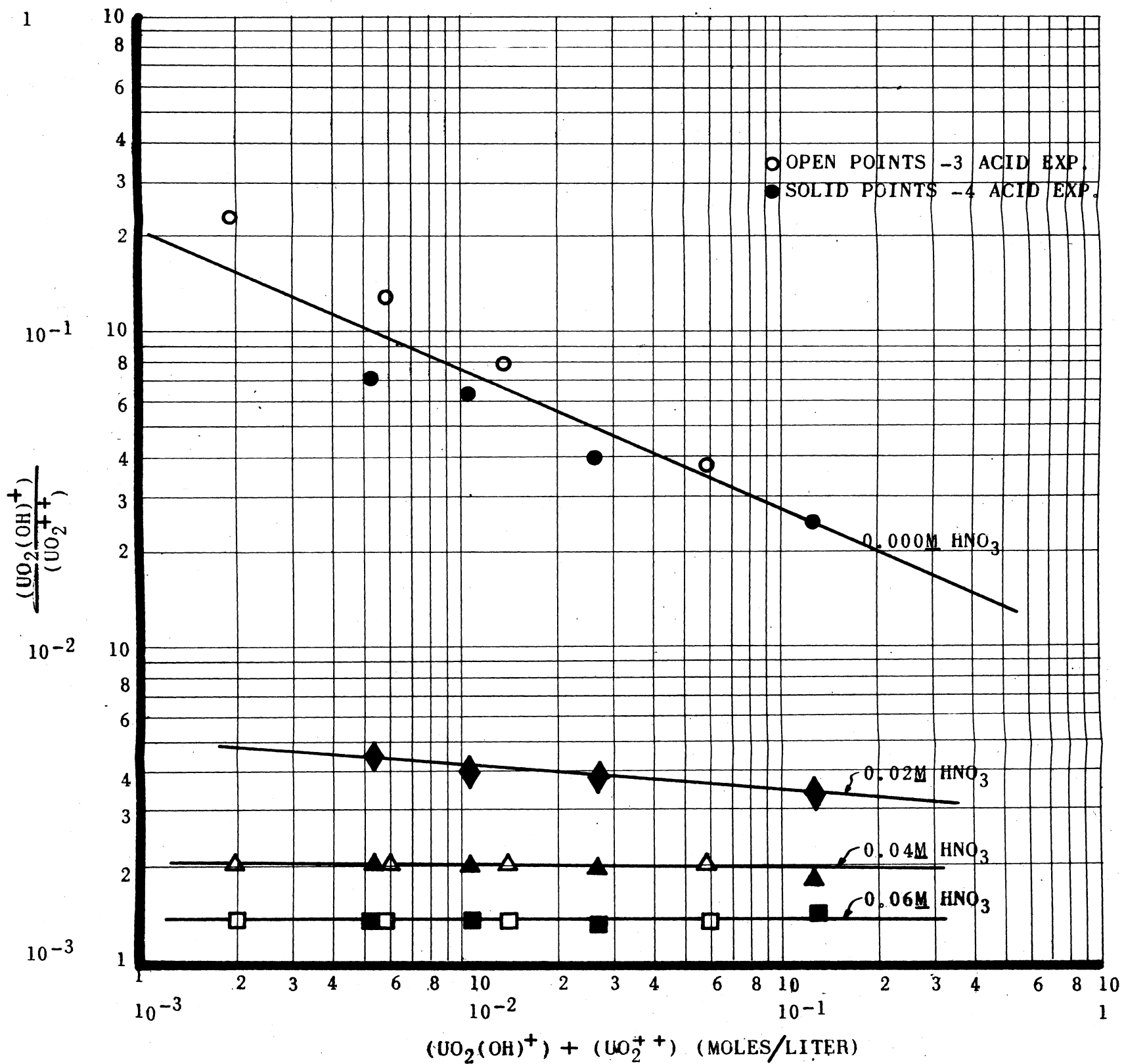


FIGURE 1

HYDROLYSIS OF URANYL NITRATE AT LOW ACID CONCENTRATIONS

TABLE 3

SUMMARY OF DISTRIBUTION DATA

<u>3-Acid Experiment</u>				<u>4-Acid Experiment</u>		
<u>HNO₃</u> <u>moles/liter</u>	<u>U_A</u> <u>moles/liter</u>	<u>Corrected</u> <u>U_A</u> <u>moles/liter</u>	<u>U_O</u> <u>moles/liter</u>	<u>U_A</u> <u>moles/liter</u>	<u>Corrected</u> <u>U_A</u> <u>moles/liter</u>	<u>U_O</u> <u>moles/liter</u>
0	1.99 x 10 ⁻³	1.627 x 10 ⁻³	4.0 x 10 ⁻⁸	5.34 x 10 ⁻³	4.984 x 10 ⁻³	8.82 x 10 ⁻⁸
	5.97 x 10 ⁻³	5.314 x 10 ⁻³	6.72 x 10 ⁻⁸	1.07 x 10 ⁻²	1.007 x 10 ⁻²	-----
	1.39 x 10 ⁻²	1.288 x 10 ⁻²	6.30 x 10 ⁻⁶	2.65 x 10 ⁻²	2.550 x 10 ⁻²	4.12 x 10 ⁻⁵
	5.97 x 10 ⁻²	5.754 x 10 ⁻²	6.60 x 10 ⁻⁴	1.27 x 10 ⁻¹	1.238 x 10 ⁻¹	3.84 x 10 ⁻³
0.02				5.34 x 10 ⁻³	5.316 x 10 ⁻³	5.04 x 10 ⁻⁶
				1.07 x 10 ⁻²	1.066 x 10 ⁻²	2.14 x 10 ⁻⁵
				2.65 x 10 ⁻²	2.640 x 10 ⁻²	1.43 x 10 ⁻⁴
				1.27 x 10 ⁻¹	1.266 x 10 ⁻¹	4.54 x 10 ⁻³
0.04	1.99 x 10 ⁻³	1.986 x 10 ⁻³	-----	5.34 x 10 ⁻³	5.329 x 10 ⁻³	1.81 x 10 ⁻⁵
	5.97 x 10 ⁻³	5.958 x 10 ⁻³	2.69 x 10 ⁻⁵	1.07 x 10 ⁻²	1.068 x 10 ⁻²	4.75 x 10 ⁻⁵
	1.39 x 10 ⁻²	1.387 x 10 ⁻²	6.30 x 10 ⁻⁵	2.65 x 10 ⁻²	2.645 x 10 ⁻²	2.13 x 10 ⁻⁴
	5.97 x 10 ⁻²	5.958 x 10 ⁻²	1.08 x 10 ⁻³	1.27 x 10 ⁻¹	1.268 x 10 ⁻¹	5.13 x 10 ⁻³
0.06	1.99 x 10 ⁻³	1.987 x 10 ⁻³	1.05 x 10 ⁻⁵	5.34 x 10 ⁻³	5.333 x 10 ⁻³	3.15 x 10 ⁻⁵
	5.97 x 10 ⁻³	5.962 x 10 ⁻³	4.12 x 10 ⁻⁵	1.07 x 10 ⁻²	1.069 x 10 ⁻²	7.73 x 10 ⁻⁵
	1.39 x 10 ⁻²	1.388 x 10 ⁻²	9.92 x 10 ⁻⁵	2.65 x 10 ⁻²	2.647 x 10 ⁻²	3.20 x 10 ⁻⁴
	5.97 x 10 ⁻²	5.962 x 10 ⁻²	1.29 x 10 ⁻³	1.27 x 10 ⁻¹	1.268 x 10 ⁻¹	5.76 x 10 ⁻³

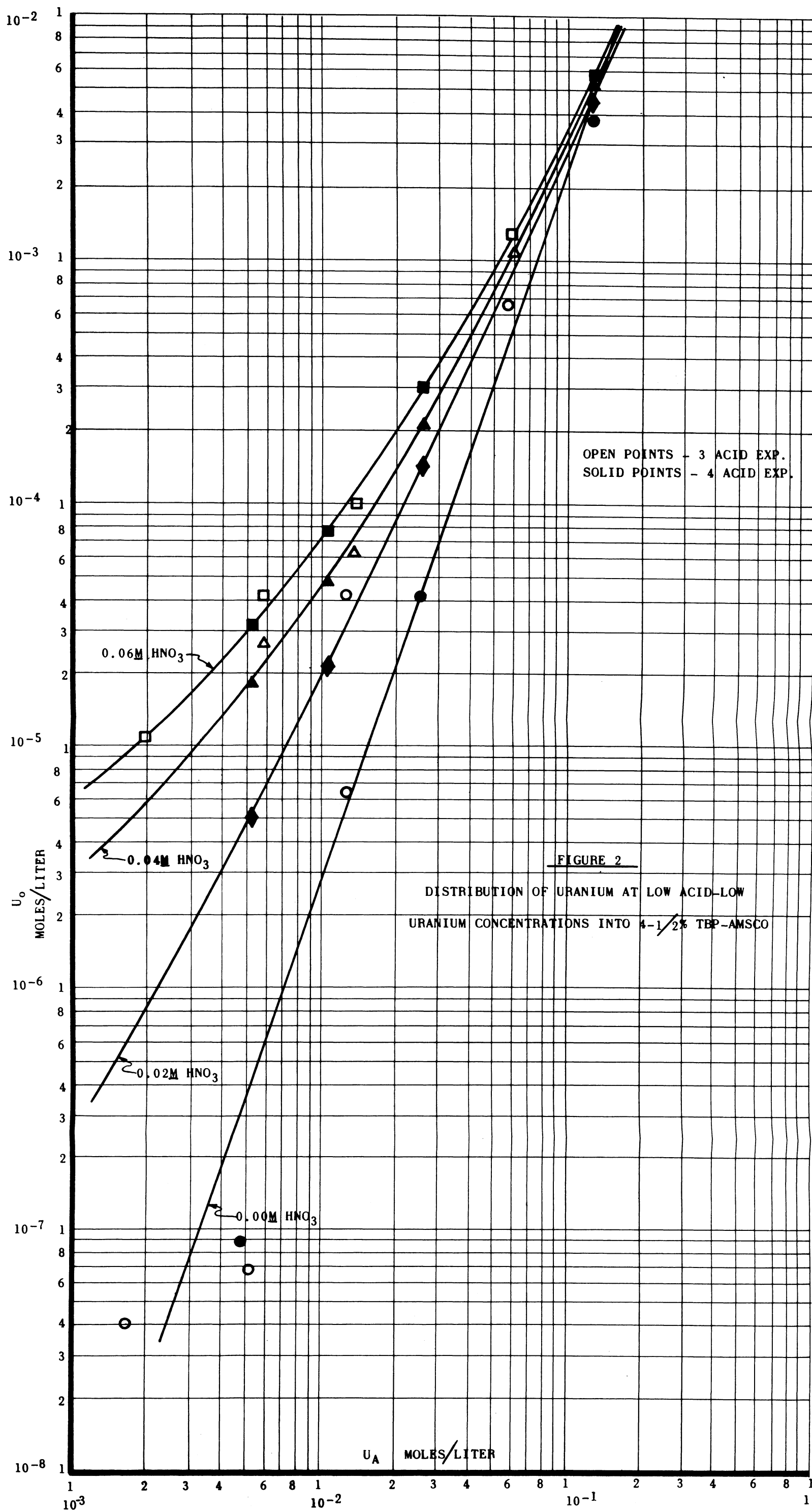


TABLE 4
SUMMARY OF STATISTICAL TREATMENT
OF DATA FROM TABLE 3

<u>Acid Concentration</u> <u>moles/liter</u>	<u>Equation</u>	<u>Limits</u>
0 (suspect points included)	$U_0 = U_A^{3.46} \cdot 10^{0.90}$	$U_0 \cdot 10^{\pm 0.43}$
0 (suspect points excluded)	$U_0 = U_A^{2.90} \cdot 10^{0.28}$	$U_0 \cdot 10^{\pm 0.27}$
2×10^{-2}	$U_0 = U_A^{2.15} \cdot 10^{-0.43}$	$U_0 \cdot 10^{\pm 0.04}$
4×10^{-2}	$U_0 = U_A^{3.20 + 0.45 \log U_A} \cdot 10^{0.24}$	$U_0 \cdot 10^{\pm 0.11}$
6×10^{-2}	$U_0 = U_A^{2.67 + 0.32 \log U_A} \cdot 10^{-0.11}$	$U_0 \cdot 10^{\pm 0.07}$

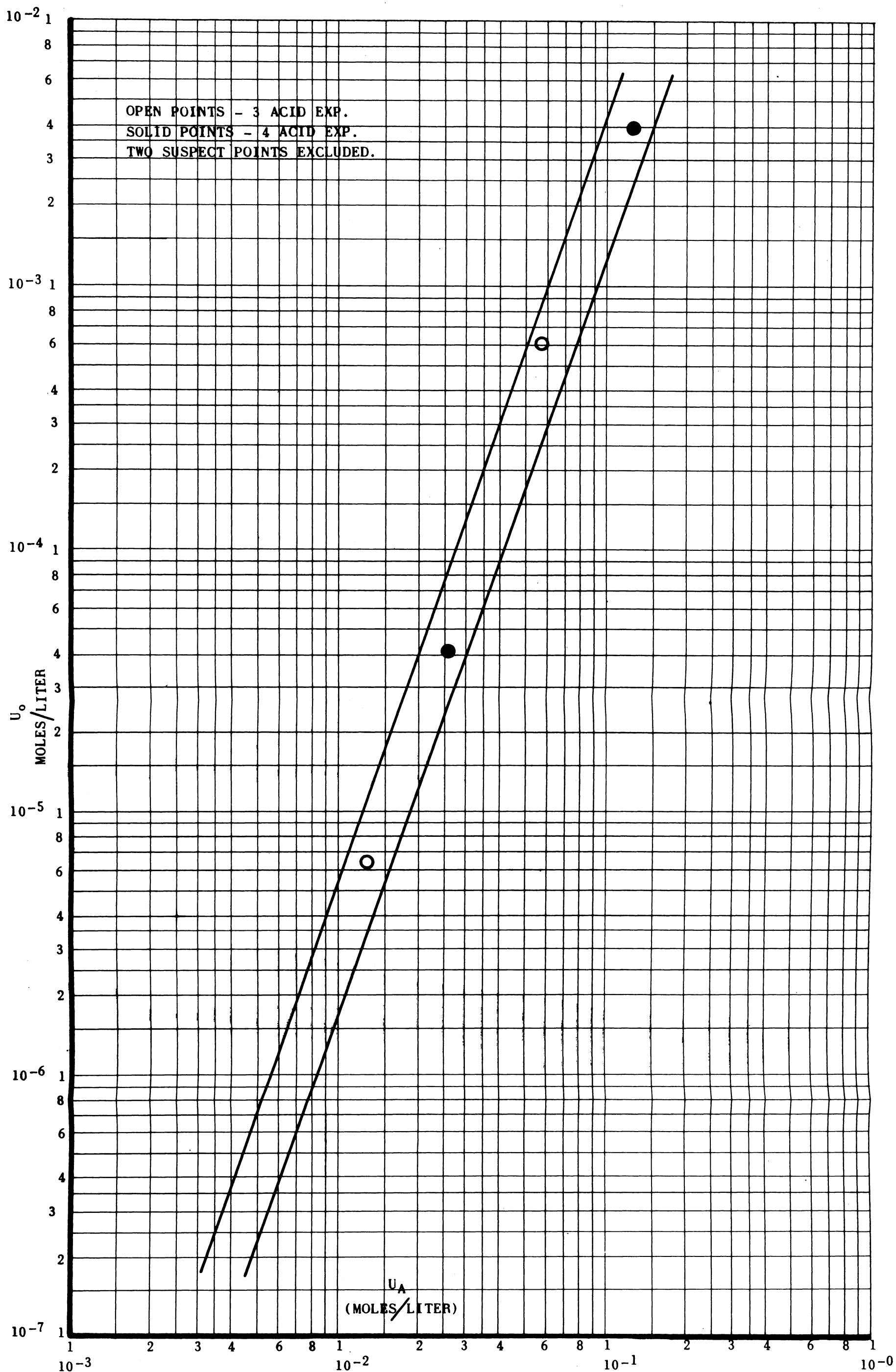


FIGURE 3

DISTRIBUTION OF URANIUM AT LOW CONCENTRATIONS BETWEEN 0.00M
 NITRIC ACID SOLUTION AND 4-1/2% TBP-AMSCO. STATISTICAL LIMITS SHOWN.

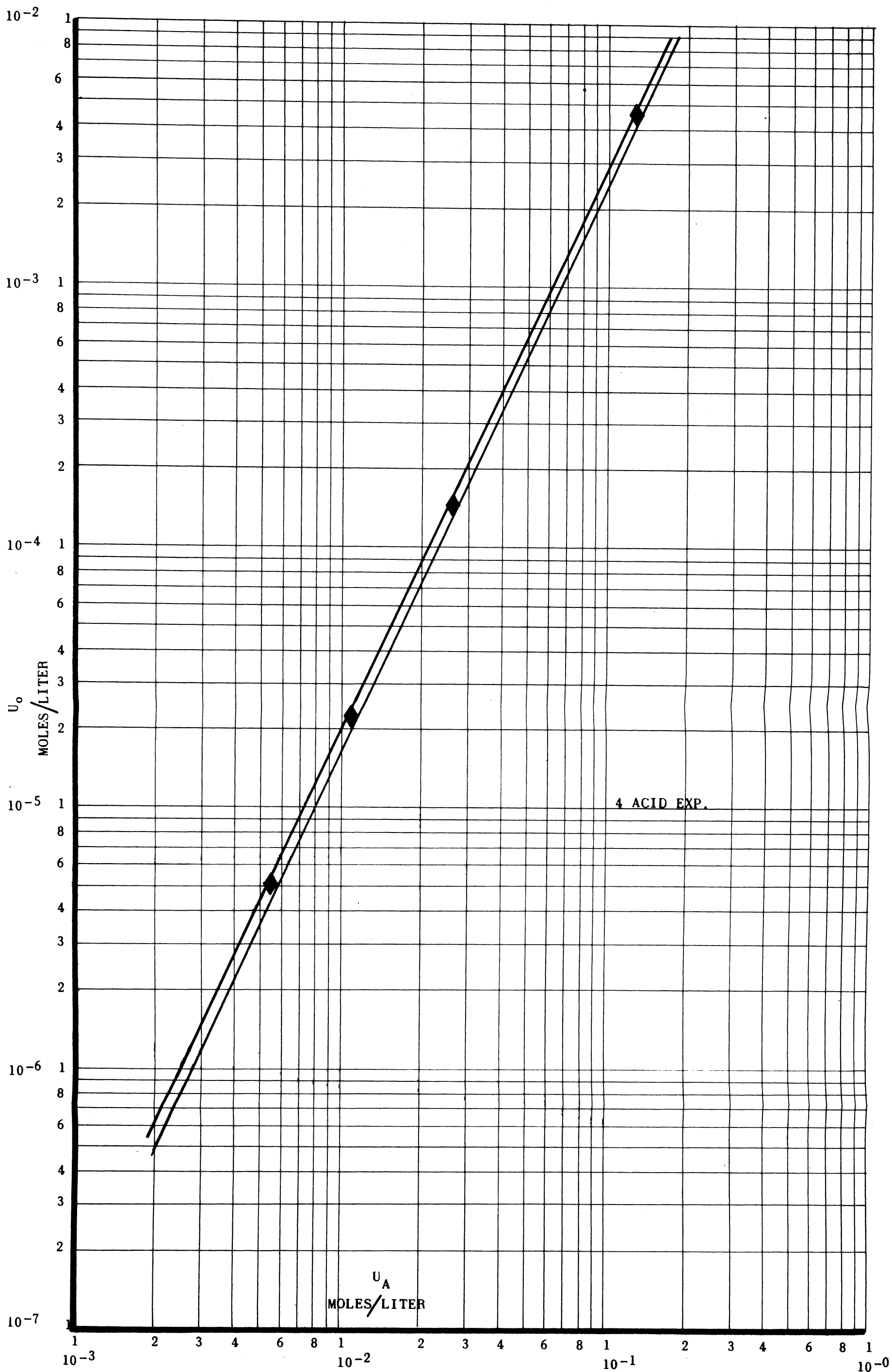


FIGURE 4

DISTRIBUTION OF URANIUM AT LOW CONCENTRATIONS BETWEEN 0.02M
 AQUEOUS NITRIC ACID SOLUTION AND 4-1/2% TBP-AMSCO. STATISTICAL LIMITS SHOWN.

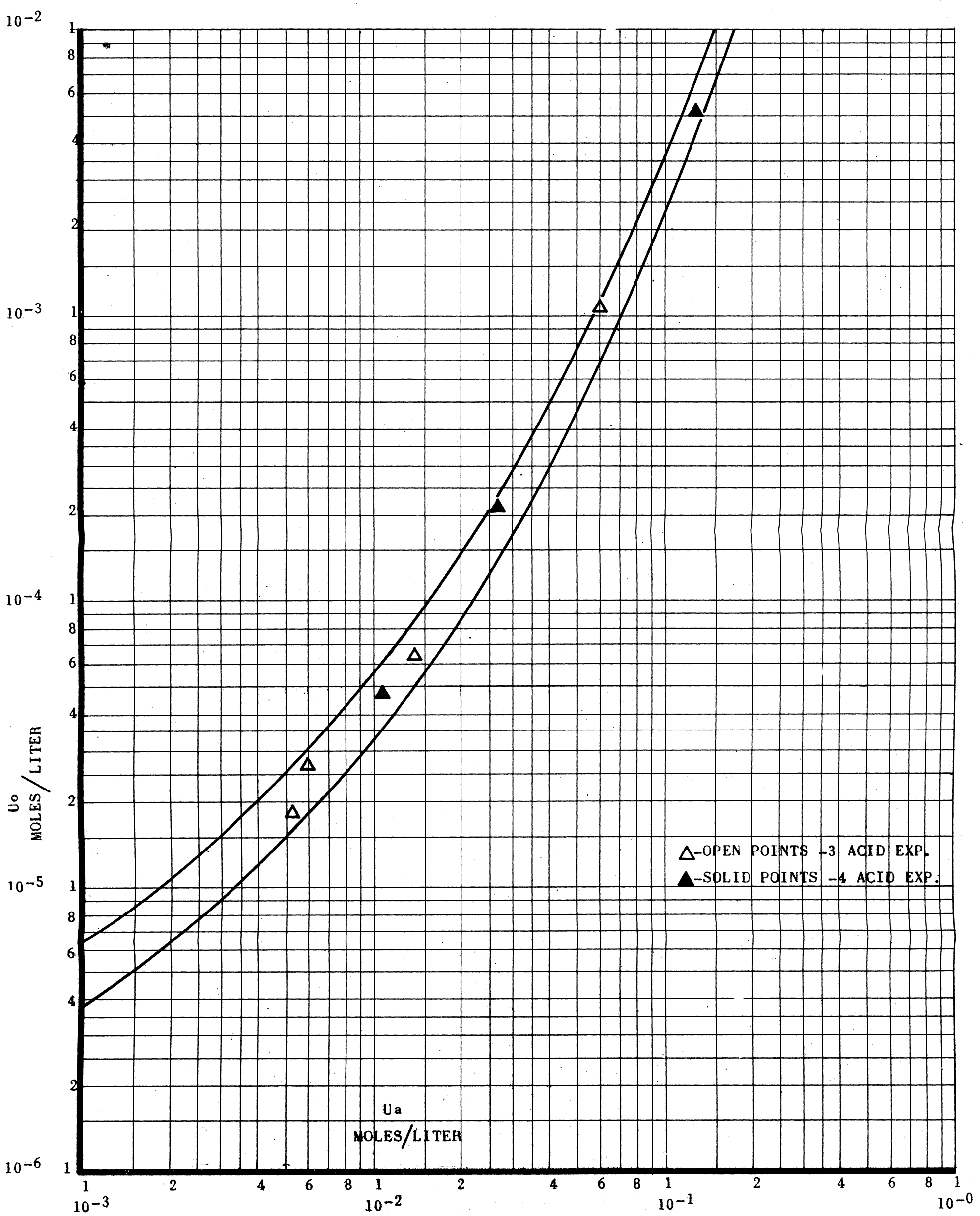


FIGURE 5
 DISTRIBUTION OF URANIUM AT LOW CONCENTRATIONS BETWEEN 0.04M AQUEOUS NITRIC
 ACID SOLUTION AND 4-1/2% TBP-AMSCO. STATISTICAL LIMITS SHOWN.

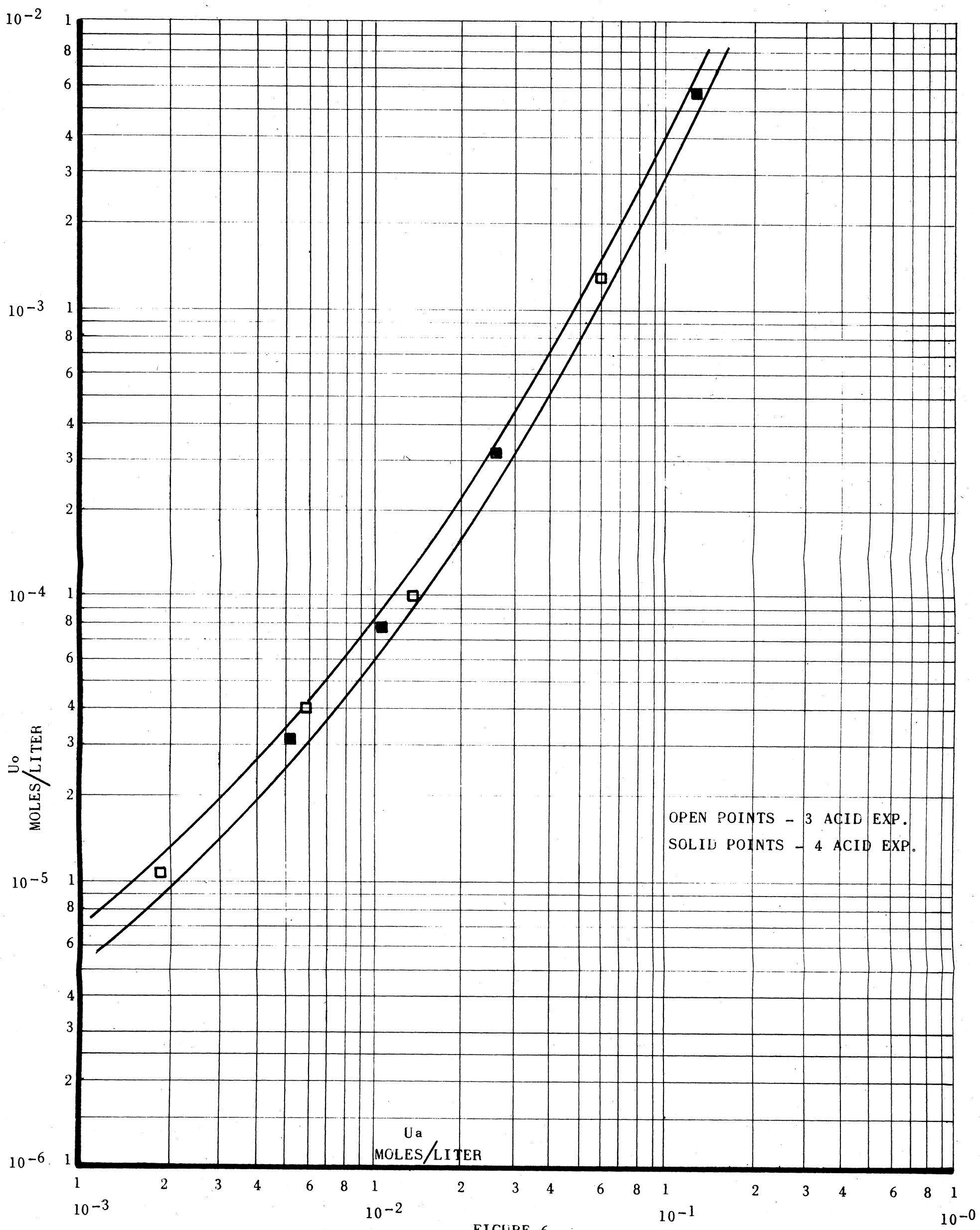


FIGURE 6

DISTRIBUTION OF URANIUM AT LOW CONCENTRATIONS BETWEEN 0.06M
 AQUEOUS NITRIC ACID SOLUTION AND 4-1/2 % TBP - AMSCO. STATISTICAL LIMITS SHOWN.

TABLE 5

SUMMARY OF THE OBSERVED VALUES OF
K AND CALCULATED VALUES OF $\sqrt{\mu}$

Acid Level (Molarity)	<u>3-Acid Experiment</u>		<u>4-Acid Experiment</u>	
	<u>K(obs)</u>	<u>$\sqrt{\mu}$</u>	<u>K(obs)</u>	<u>$\sqrt{\mu}$</u>
0.00	56.9	0.075	56.9	0.125
0.00	32.4	0.132	-----	-----
0.00	23.6	0.202	21.5	0.280
0.00	30.0	0.420	18.0	0.615
0.02			37.5	0.236
0.02			43.6	0.228
0.02			38.3	0.316
0.02			18.0	0.636
0.04	-----	-----	50.8	0.236
0.04	62.6	0.240	44.0	0.268
0.04	36.7	0.286	34.7	0.346
0.04	26.9	0.467	17.5	0.649
0.06	48.8	0.256	44.3	0.276
0.06	49.9	0.278	40.6	0.302
0.06	34.7	0.320	35.1	0.360
0.06	25.4	0.488	17.2	0.664

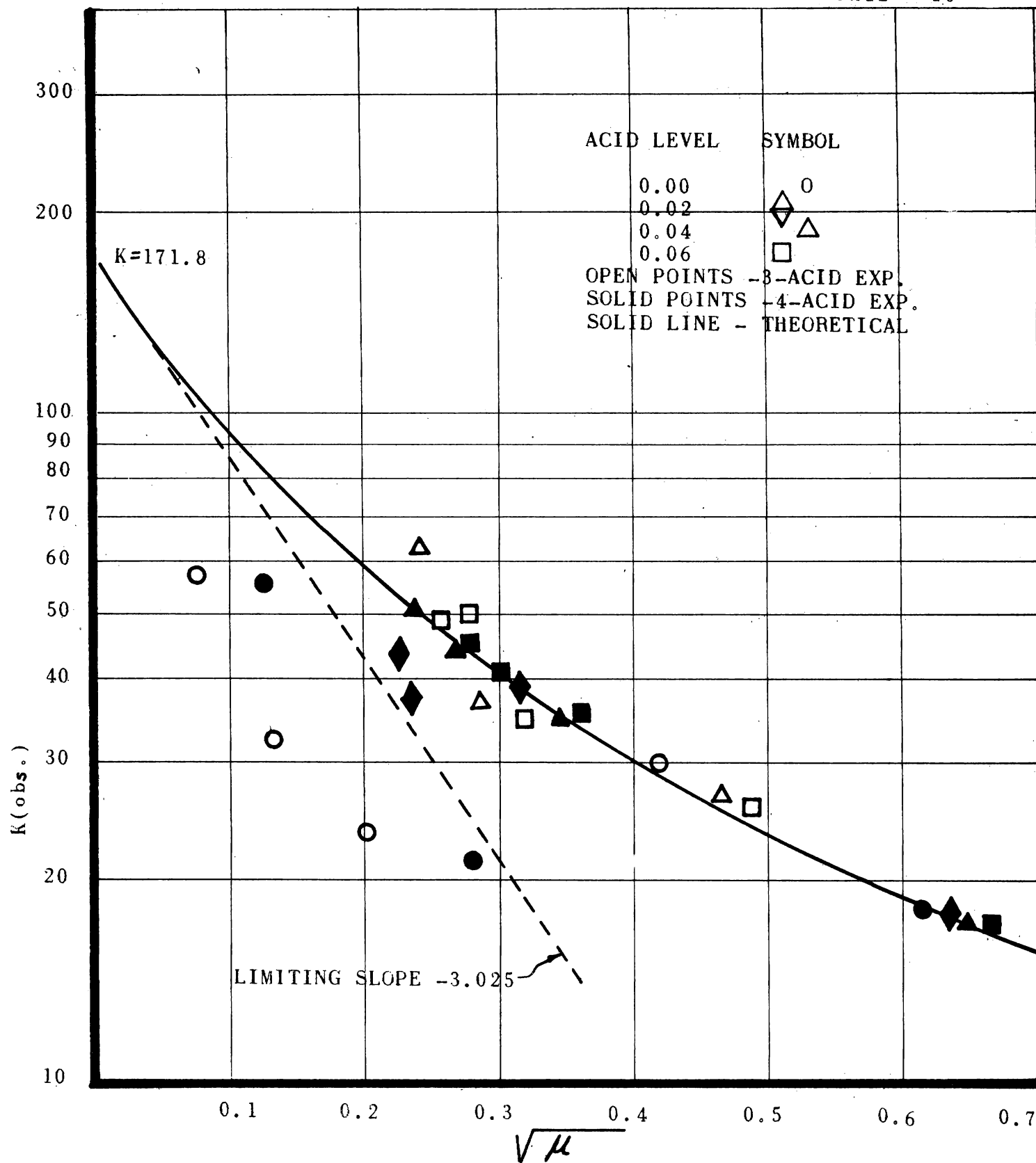


FIGURE 7

OBSERVED EQUILIBRIUM CONSTANT AS A FUNCTION
OF THE SQUARE ROOT OF THE IONIC STRENGTH



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