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# RaD PURIFICATION, Po PREPARATION, AND CALIBRATION OF MEASURING APPARATUS

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## ABSTRACT

RaD is purified from solution by electrolysis at various current densities, and a procedure is described for extracting Po sources on Pt electrodes. In measuring the strength of the sources, difficulty was encountered with non-linearity of the microammeter of an integrating ionization chamber, and the method of calibrating this in several degrees of sensitivity is described in detail.

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# RaD PURIFICATION, Po PREPARATION, AND CALIBRATION OF MEASURING APPARATUS

## I. Purification of RaD and Preparation of Po Sources.

The solution of RaD, RaE and RaF (Po) obtained from Dr. J. G. Hamilton in Berkeley contained considerable impurity, and it was decided to extract the RaD from these impurities and thus have a clean source of Po. The solution as received contained 350 ± 25 millicuries of Po and 500 ± 25 millicuries of RaD. The method used to extract the RaD from the solution was that used by O. Erbacher and K. Phillip, Zeitschrift fur Physik, Vol. 51, page 309.

The procedure they used consisted of electroplating the RaD from 1.0N $HNO_3$ on to platinum electrodes with a current density of $3 \times 10^{-3}$ amps. per $cm^2$ at each electrode, dissolving the RaD off and replating at $3 \times 10^{-4}$ amps/$cm^2$. This gave them a pure solution of RaD when the activity was dissolved from the platinum. The first operations done on the original solution are outlined in the first table at the end of this report.

The results obtained in the first successful plating of RaD differed from those of Erbacher in that we obtained all the RaD on the anode and he found some 67% of the RaD on the cathode. This can be accounted for by the fact that our solution was partially purified by an ammonium hydroxide precipitation and one gets anodic deposition of Pb when the solution is relatively clean. Erbacher got anodic deposition on his second electrolysis.

The recovery of the RaD in the first electrolysis had better than a 99.5% yield. This excellant recovery was due to the fact that there was 190 mg. of Pb carrier in the solution. Dr. Hamilton believed that the sample contained no Pb carrier but the history of the solution is so uncertain that it was not surprising to find the presence of this added Pb.

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The $PbO_2$ samples from the anodes were combined and assayed. They contained 500± 25 mc. of RaD and 130 mc. of Po. The remainder of the Po had plated out on the cathode during the plating operations. The solution of 500 mc. of RaD and 130 mc. of Po was called 1-B. By reducing the current to $5 \times 10^{-6}$ amps. per $cm^2$ it was possible to plate the Po out of 1-B and not the Pb. This was done in 1.0N $HNO_3$ and over 90% of the Po was removed on the two electrodes from 1-B leaving all of the Pb in solution. After this operation the Pb was plated from 1-B with a current density of $2 \times 10^{-3}$ amps. per $cm^2$. The yield was 99%. The $PbO_2$ was dissolved from the anodes with dilute nitric acid and peroxide solution. The $Pb(NO_3)_2$ obtained (containing about 500 mc. of RaD) was then converted to the chloride with concentrated HCl, finally evaporated to dryness and put aside to allow the Po to grow. The Po which was not in 1-B was plated from a separate solution and at present is on platinum foils which are in use.

The Po which grows from the 500 mc. of RaD will be extracted as needed by using a hydrogen cell with a platinum electrode. When platinum is revolved in a 0.1N HCl solution of RaD and a stream of $H_2$ passed over it the Po plated out onto the platinum leaving the RaD in solution. The obvious advantage to this method is no reagents or impurities are introduced into the RaD solution so periodic extraction of Po is possible without contamination of the source.

## II. Calibration of Measuring Apparatus.

The measuring apparatus used for this work consisted of a Lauritsen electroscope and an alpha-integrating machine which was designed and built by Dr. Segre and his group. The alpha-measuring apparatus is a large plane condenser with guard ring in which the total ionization current of the Po

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alphas is collected. A D-C amplifier and a microammeter measure the current. This machine has five sensitivities on its ranging from 0.54 x $10^9$ disintegrations per minute per division on the microammeter to 0.375 x $10^5$ disintegrations per minute per division. Each sensitivity is approximately a factor of ten less than the preceding one. Appropriate samples of Po were counted on Dr. Segre's demultiplier and then put into the alpha machine. In this way sensitivities 4 and 5 were calibrated. It was noted that the microammeter was not linear over its range of from 0 to 100. In order to get a plot of this non-linearity of the meter several samples of Po ranging around 10 divisions on step 5 were put into the machine and the deflection of the meter from each sample carefully read. Then, by combining these samples in various combinations, readings could be obtained over the full scale on step 5. It was found that the value obtained by adding the individual deflections of the samples numerically differed from the one obtained collectively on the machine. The same thing was done on step 4 sensitivity with the same results. The data are shown in tables 4 and 5. A plot of these data are shown in figure 1. The experimental points are not given but the curve is the average of these points. When this curve was used, and the Po samples which had been counted on the demultiplier were again put into the machine, correct factors were obtained for steps 4 and 5 which were consistent throughout the range of the microammeter. When these two steps were calibrated the next thing was the calibration of the Lauritsen electroscope. To accomplish this a radioactive equilibrium source of RaD, RaE and RaF (Po) was needed. Dr. Segre had part of an old Radon pump system which had been sealed about 6 years, so contained within this was an equilibrium of RaD, RaE and RaF. This container was made of soft glass and was in the form of two bulbs, each about 1.5 inches

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in diameter and connected together by a short glass tube. The whole thing had the appearance of a small dumbbell. We broke one of the bulbs with a hammer and assumed that each individual chip of glass would have the required equilibrium mixture on it, but when several of these chips were put into the alpha machine and then under the electroscope it was found that the ratio of the alpha and beta counts were not consistent. (Dr. Segre was informed recently by Paneth that he has observed the same phenomenon although it does not appear in the literature.) It was then decided to dissolve the activity from the other bulb as a whole since it is unreasonable to believe an equilibrium did not exist in half of the container. The activity was removed from the bulb with hot $HNO_3$. The solution obtained was then counted to see if it was in equilibrium. This was done by mounting two samples on mica and counting the alpha particles on a linear amplifier and the beta particles on a Geiger counter. It was found that the solution was in equilibrium. Several samples from this solution were mounted on platinum and counted on both the alpha-integrator and electroscope. The data are given in table 6. The average of fourteen of these samples gives a value of 11.4 ± 5% microcuries of RaE per division per second on the electroscope.

With these two calibrated machines it was very simple to follow the alpha and beta activity of the RaD solution that was purified. The alphas of course are emitted by the Po and the betas by the RaE. The beta particles from RaD are so soft that it is impossible to measure them so they do not enter into the activity measurements at all. Each RaE sample that is measured, however, has to be followed for several days to see whether it grows or decays in order to know where the RaD is.

The calibration of steps 1, 2 and 3 on the alpha machine were simply done by measuring two or three samples which gave a large deflection on step 4

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then changing the machine to step 3 and taking a reading. After step 3 was calibrated you do the same from step 3 to step 2 and then from step 2 to step 1. It is not known whether the machine will function properly over the entire range of the microammeter on step 1 because at present we do not have samples large enough that have been accurately counted. Also there is some doubt as to whether in this high-activity range the current will stay in the same proportion to the activity as in the lower ranges. The machine is good, however, up through step 2 and the first 10 divisions on step 1. Following are the factors for the five steps obtained as described above.

| Sensitivity Step | Disintegrations per minute per division on microammeter |
|---|---|
| 1 | $0.540 \times 10^9$ |
| 2 | $0.500 \times 10^8$ |
| 3 | $0.425 \times 10^7$ |
| 4 | $0.420 \times 10^6$ |
| 5 | $0.375 \times 10^5$ |

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## TABLE I UNSUCCESSFUL PLATING OF ORIGINAL SOLUTION

300 cc dark green solution with white precipitate on bottom

Solution contained RaD, E, F
0.1N HCl
0.5N $HNO_3$
Large amount of $Ni^{++}$
Some $Cu^{++}$
Probably $Hg^{++}$
Plus many unknowns

**Solution + Sup. on Right**

Evap. to thick green past, would not evap. further because $Ni(NO_3)_2 \cdot 6H_2O$ melts at 56°C. Put in vacuum for 6 hrs. and dried. Made up to 200 cc. and 1.0 N in $HNO_3$. Divided into two 100 cc. solutions

**Decanted Precipitate**

+conc. $HNO_3$
Digested 2 days
on steam bath

**Supernatant**

Added to evaporating solution on the left. Contained 20% of activity originally in the green soln.

**Precipitate**

Digested again in conc. $HNO_3$

**Supernatant**

2.9% of activity in orig. green soln. was put aside

**Ppt.**

Digested in Aqua Regia and put aside

**Solution 1-A-B**

Set aside; purified later by the chemical procedure shown in the next table.

**Solution 1-A-A**

Plated for 24 hrs. at 3 milliamps per 20 sq. cm.
Plated for 50 hrs. at 40 milliamps per 20 sq. cm.
Plated for 22 hrs. at 50 milliamps per 20 sq. cm.
Added Xs $AgNO_3$ solution to remove chloride. This ppt. carried no activity.
Plated 22 hrs. at 100 milliamps per 20 sq. cm. [redacted]
Plated 15 hrs at 60 milliamps per 20 sq. cm.
All the above plating did not remove any RaD so solution 1-A-A had the following chemistry done on it. Tests were run first on small samples.
(See Table 2)

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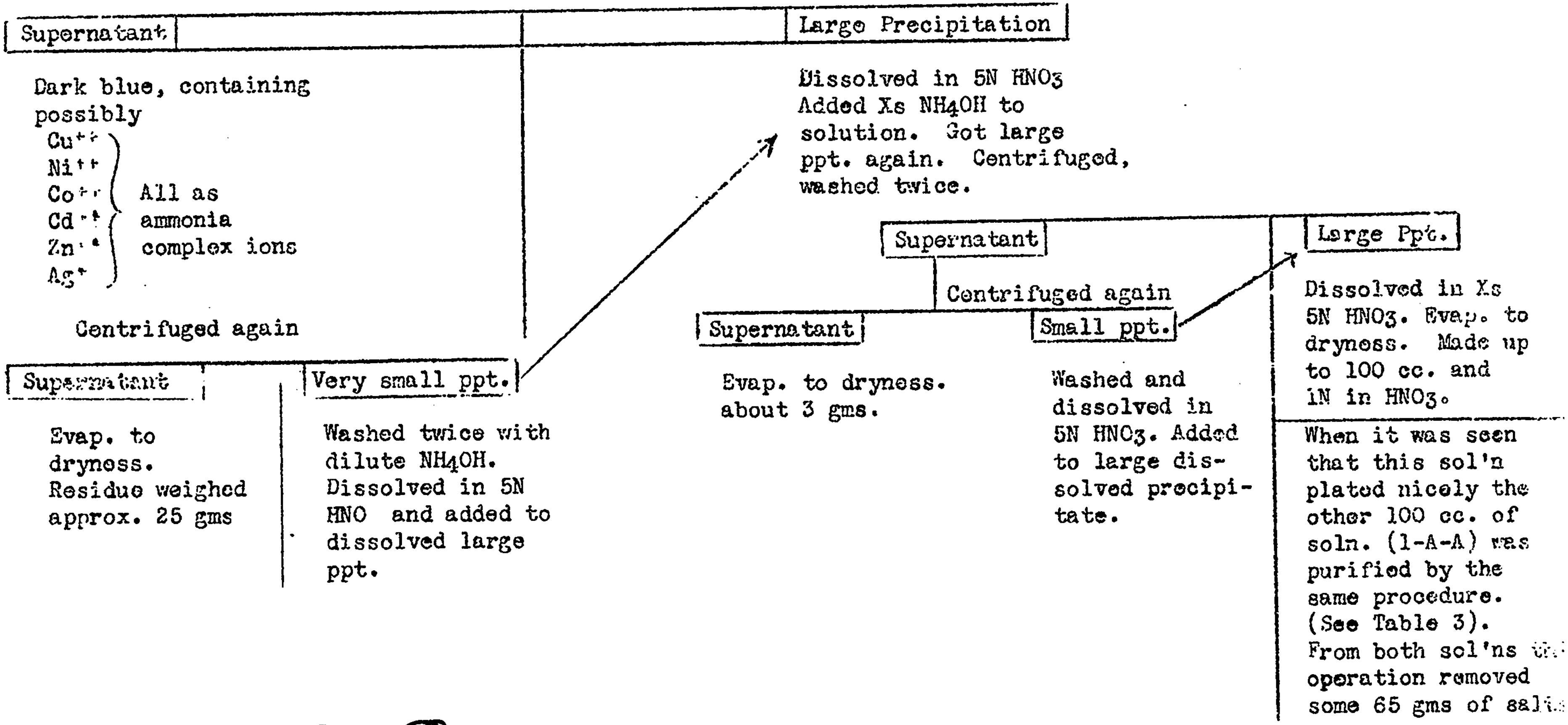
## TABLE 2 AMMONIUM HYDROXIDE PRECIPITATION



100 cc. Solution 1-A-A

+Xs 6N $NH_4OH$
Got large slimy precipitate
Centrifuged, washed twice
with dilute $NH_4OH$

**Supernatant**

Dark blue, containing possibly $Cu^{++}$, $Ni^{++}$, $Co^{++}$, $Cd^{++}$, $Zn^{++}$, $Ag^{+}$ } All as ammonia complex ions

Centrifuged again

**Supernatant**

Evap. to dryness. Residue weighed approx. 25 gms

**Very small ppt.**

Washed twice with dilute $NH_4OH$. Dissolved in 5N HNO and added to dissolved large ppt.

**Large Precipitation**

Dissolved in 5N $HNO_3$
Added Xs $NH_4OH$ to solution. Got large ppt. again. Centrifuged, washed twice.

**Supernatant**

Centrifuged again

**Supernatant**

Evap. to dryness. about 3 gms.

**Small ppt.**

Washed and dissolved in 5N $HNO_3$. Added to large dissolved precipitate.

**Large Ppt.**

Dissolved in Xs 5N $HNO_3$. Evap. to dryness. Made up to 100 cc. and 1N in $HNO_3$.

When it was seen that this sol'n plated nicely the other 100 cc. of soln. (1-A-A) was purified by the same procedure. (See Table 3). From both sol'ns th operation removed some 65 gms of sal

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### TABLE 3 DATA ON PLATING OF RaD

The values of the activities given below are in divisions per second obtained on a Lauritsen electroscope of the RaE beta-particles, and were measured on identical aliquots of the solutions after evaporation to dryness.

Solution 1-A-A

| Hours Plated | Current Density Milliamps per $cm^2$ | Activity div. per sec. |
|---|---|---|
| --- | 0 | 0.995 |
| 96 | 1 | 0.706 |
| 120 | 0.5 | 0.526 |
| 24 | 1.0 | 0.433 |
| 6 | 2.0 | 0.008 |
| 18 | 3.0 | 0.006 |
| 7 | 3.0 | 0.005 |
| Total 271 | | |

Solution 1-A-B

| Hours Plated | Current Density Milliamps per $cm^2$ | Activity div. per sec. |
|---|---|---|
| --- | 0 | 1.000 |
| 48 | 0.5 | 0.903 |
| 224 | 1.0 | 0.698 |
| 5 | 3.0 | 0.009 |
| 13 | 3.0 | 0.392 * |
| 0.25 | 3.0 | 0.251 |
| 6 | 3.0 | 0.057 ⁑ |
| Total 96 | | |

* This aliquot was taken after the anode had been removed from the solution and cleaned of $PbO_2$. Now if the cathode had RaE on it, it could have dissolved off when the current was interrupted because the cathode was not removed from the solution.

⁑ This sample decayed with the five day half-life of RaE, indicating that no RaD was left in the solution.

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## TABLE 4 NON-LINEARITY OF STEP 5

| Readings From Machine | Numerical Addition of Samples | Difference |
|---|---|---|
| 14.0 | 14.0 | 0.0 |
| 20.5 | 20.5 | 0.0 |
| 28.0 | 27.5 | 0.5 |
| 38.5 | 37.5 | 1.0 |
| 48.5 | 47.0 | 1.5 |
| 61.5 | 58.0 | 3.5 |
| 63.5 | 60.0 | 3.5 |
| 69.0 | 65.0 | 4.0 |
| 76.5 | 71.5 | 5.0 |
| 79.5 | 74.0 | 5.5 |
| 86.0 | 80.0 | 6.0 |
| 92.5 | 84.5 | 8.0 |

## TABLE 5 NON-LINEARITY OF STEP 4

| Readings From Machine | Numerical Addition of Samples | Difference |
|---|---|---|
| 20.0 | 20.0 | 0.0 |
| 28.0 | 28.0 | 0.0 |
| 39.5 | 38.0 | 1.5 |
| 48.0 | 46.0 | 2.0 |
| 59.0 | 56.0 | 3.0 |
| 72.0 | 66.5 | 5.5 |
| 84.0 | 76.0 | 8.0 |
| 99.5 | 91.0 | 8.5 |

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TABLE 6 CALIBRATION OF LAURITSEN ELECTROSCOPE FOR RaE

| Sample # | Division on alpha machine | Sensitivity Step of alpha machine | Division per second on electroscope | Corrected Values due to non-linearity of alpha machine | Microcuries of RaE per divisi per second on electroscope |
|---|---|---|---|---|---|
| 9-6-1 | 46 | 5 | 0.0624 | 44.5 | 12.0 |
| 9-6-2 | 60 | " | 0.0871 | 57.0 | 11.1 |
| 9-6-3 | 33.5 | " | 0.0476 | 33.0 | 11.7 |
| 9-6-4 | 16 | " | 0.0238 | 16.0 | 11.4 |
| 9-6-5 | 43 | " | 0.0601 | 41.8 | 11.8 |
| 9-7-1 | 83 | " | 0.1182 | 76.7 | 10.9 |
| 9-7-2 | 90 | " | 0.1272 | 82.4 | 11.0 |
| 9-7-3 | 85 | " | 0.1222 | 78.5 | 10.8 |
| 9-7-4 | 84 | " | 0.1202 | 77.7 | 10.9 |
| 9-7-5 | 58 | " | 0.0852 | 55.2 | 11.0 |
| 9-7-6 | 63 | " | 0.0893 | 59.8 | 11.7 |
| 11-22-1 | 26 | 4 | 0.421 | 26.0 | 11.8 |
| 11-22-2 | 15 | " | 0.245 | 15.0 | 11.8 |
| 11-22-3 | 8.8 | " | 0.141 | 8.8 | 11.6 |
| | | | | Ave. = | 11.4 ± 5% |

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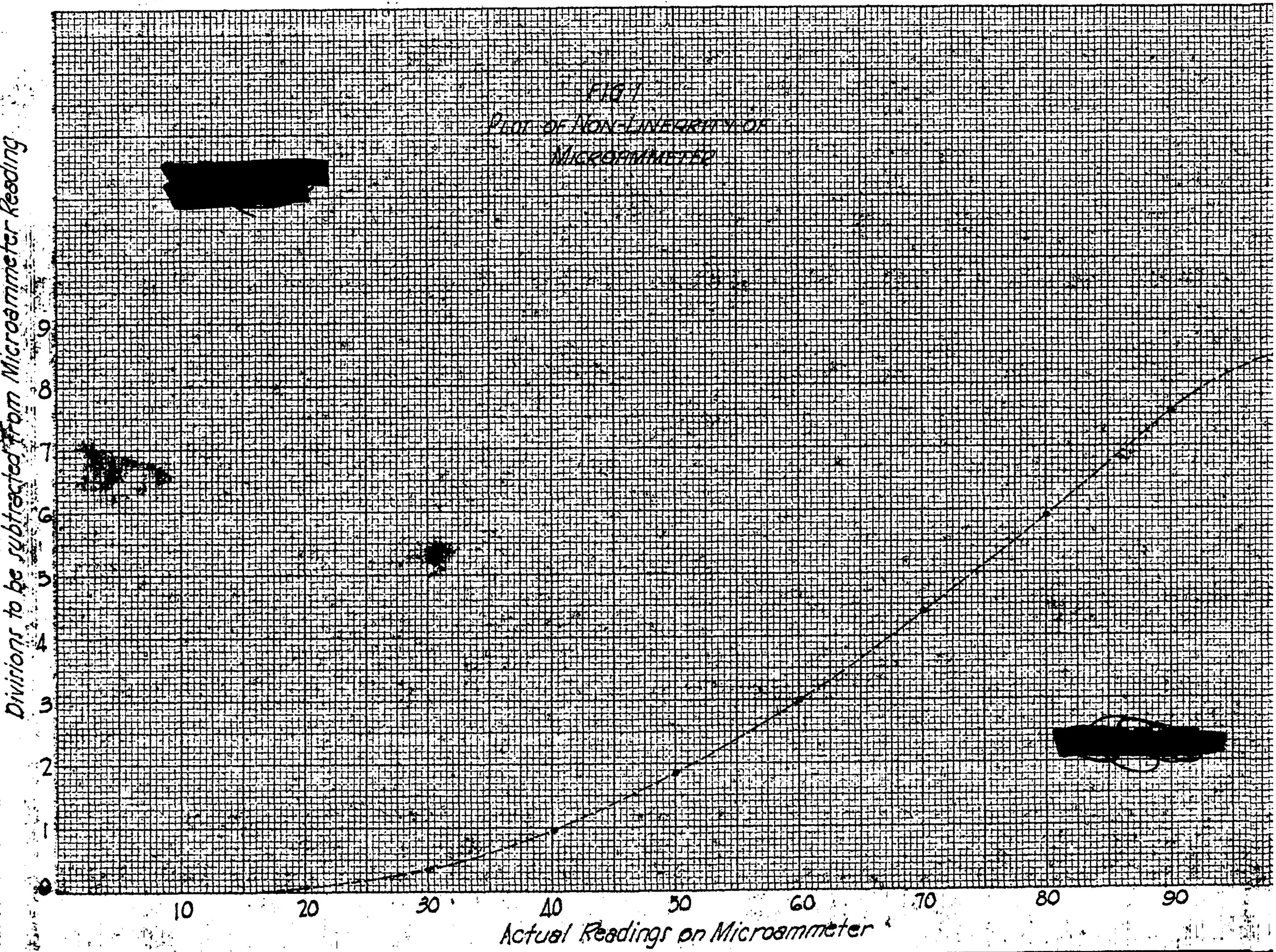

FIG 1
PLOT OF NON-LINEARITY OF
MICROAMMETER

[redacted]

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