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# RADIOCHEMICAL EVALUATION OF FIRE ASSAY METHOD FOR DETERMINATION OF SILVER

by

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# Radiochemical Evaluation of Fire Assay Method for Determination of Silver

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▷ Radiochemical experiments using silver-110 have been performed to evaluate the fire assay method for the determination of silver. The losses of silver during crucible fusion, scorification, and cupellation have been measured under a variety of experimental conditions. The major loss occurs in the cupellation step and it is difficult to reduce this loss to less than 2% of the silver present. In experiments with pure fluxes the loss of silver to the slag during crucible fusion was approximately 0.2%; that to the crucible wall was approximately 0.5%. The loss may exceed 2% in the fusion of charges rich in copper and/or nickel sulfide materials or with charges giving viscous slags. Loss of silver to the slag during scorification of impure lead buttons was approximately 0.2%.

ALTHOUGH methods for determining the precious metals by fire assay have been in use for many years, numerous problems still exist in the accurate determination of the platinum metals (*1, 2, 4, 10*). These methods are successful for gold and silver, although few systematic studies have been performed to provide a sound basis for their acceptance.

This laboratory is engaged in a systematic investigation of the behavior of the precious metals during determination by fire assay. The accuracy of the silver determination was evaluated first, because it is the most common and abundant of the precious metals. It was decided to use radiochemical techniques to trace the silver, as these would provide a direct and sensitive means for determining the losses encountered in crucible fusion, scorification, and cupellation and these studies would be valuable for developing techniques which could be applied to the rarer platinum metals.

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Despite the great advantages of using radioisotopes in this field, few such attempts have been made. Theirs, Graydon, and Beamish (*10*) used ruthenium-106 for studying the behavior of ruthenium during its determination by fire assay. Nakamura and Fukami (*8*) used silver-110 to measure the loss of silver to bone ash-cement cupels during the cupellation, at a fixed temperature, of lead buttons containing very small quantities of silver. The loss varied from 5 to 30% with beads weighing from 2 to 0.1 mg., respectively. By analyzing various portions of the used cupels they showed how the absorbed silver was distributed throughout the cupels.

The present work describes experiments using silver-110, half life 270 days, as a tracer to determine the magnitude of the losses of this metal that may be encountered in the various steps of its determination by fire assay. Under certain conditions (often used in practice) the total error due to losses may be as high as 5 to 10%. However, with careful control of conditions the error can be minimized and should seldom exceed 3%.

Chopin (*3*) and King (*7*) found that volatilization losses of silver during fire assay were negligible. Therefore this factor was not investigated in the present work.

## APPARATUS AND REAGENTS

Stock Silver-110 Tracer Solution was prepared by dissolving radioactive silver wire, containing approximately 10 mc. of silver-110 in dilute nitric acid and finally diluting with water to give a solution containing 1 mg. of silver per ml.

Flux for Fuson of Silver-Bearing Test Samples. Except where otherwise stated, the composition of the flux used (standard flux) was as follows:

| | Grams | Weight % |
|---|---|---|
| PbO | 90 | 53.9 |
| $Na_2CO_3$ | 36 | 21.5 |
| $Na_2B_4O_7$ | 18 | 10.8 |
| $SiO_2$ | 20 | 12.0 |
| Flour | 3 | 1.8 |
| | | 100.0 |

Preset Time Scaler, Model SC15CST, Electronic Associates, Ltd., Toronto, Canada.

Glass Envelope Immersion-Type Geiger-Müller Tube. Type B6, manufactured by 20th Century Electronics, Croydon, Surrey, England. The tube was fitted with a glass collar and a cell capable of holding a 5-ml. sample when fitted around the tube.

Temco portable pyrometer Model 50.

Chromel-Alumel thermocouple.

Assay furnace, 15-kw. Globar-type.

## EXPERIMENTAL PROCEDURE AND RESULTS

**General Experimental Procedure.** The test samples were prepared by mixing appropriate quantities of flux, silver-110 stock solution, powdered silver nitrate carrier, and in certain cases gold or mineral sulfides. The samples were placed in 30-gram glazed fire-assay crucibles and fused at 1030° C. for 45 minutes. The melts were then poured into conical steel molds and when cool the lead buttons were separated from the slag and then either scorified or cupelled.

When scorifications were performed, sufficient lead foil was wrapped around the buttons to bring their total weight to about 70 grams. The buttons were then placed in 3-inch scorifying dishes and scorified at 980° C. for 30 to 40 minutes (until the molten slag covered the "eye" of the button). The melts were then poured into conical steel molds and when cool the buttons were separated from the slag. When necessary, the scorification step was repeated until the color of the slag indicated essentially complete removal of impurities (iron, copper, nickel).

In tests involving cupellation, the lead buttons were placed in preheated cupels of various sizes and shapes and cupelled in the furnace at known temperatures ranging from 895° to 1010° C. to give silver or silver-gold beads.

Because of the difficulty in determining the temperature of the molten buttons, the temperature of cupellation was arbitrarily taken to be that of the average temperature of the air in the furnace during the cupellation period. The air temperature was measured with a Chromel-Alumel thermocouple placed about 0.5 inch from the side and approximately at the same level as the height of the cupel.

After some of the cupellation tests, the cupels were ground to −100 mesh and cautiously added with stirring to about 150 ml. of concentrated nitric acid in a 600-ml. beaker. After the vigorous reaction had ceased, water was added and the beaker and contents were heated until the insoluble matter has dissolved (a slight suspension of silicon dioxide remained). The solutions were finally diluted to 500 ml. with water and the silver-110 content of a 5-ml. aliquot of each solution was measured.

To determine the silver-110 content of the slags from crucible fusion or scorification, all the slag was pulverized in a steel percussion mortar and then thoroughly mixed with a weighed quantity of powdered silver nitrate and a suitable quantity of flux plus flour.

This mixture was fused in the original crucible, and the resulting buttons were cupelled at 895° to 900° C. to give reassay silver beads. The beads were dissolved in warm 1 to 3 nitric acid and the resulting solutions were finally diluted to 25 ml. The silver-110 activity of a 5-ml. aliquot of each solution was measured according to the procedure given below. In certain tests, the slags were assayed a second time by the same procedure as above.

**Salting of Fluxes with Silver-110 and Radiochemical Measurement.** In all tests, a weighed quantity of powdered silver nitrate was thoroughly mixed with each charge to act as a carrier for silver-110. Approximately half of the mixture was then transferred to a 400-ml. beaker. A measured aliquot of the stock silver-110 solution was added to the center of the bed in the beaker. The remaining portion of the charge was added and the beaker and contents were heated at 110° C. in a drying oven for at least 2 hours.

A standard reference solution of silver-110 was prepared at the same time as the test samples by taking the same quantity of powdered silver nitrate and stock silver-110 solution as was used to salt the experimental charges, and then diluting to 100 ml. with 10% nitric acid.

After the test samples which had been heated in the drying oven had cooled, the lumps were broken up in a mortar and the material was thoroughly mixed. They were then carried through the various steps of fire assaying that were under study. The final product to be radiometrically assayed was either pulverized cupel material or silver reassay beads. These materials were dissolved in acid and diluted to a known volume. The radioactivity in a measured aliquot of the test solutions was determined with the immersion Geiger-Müller tube and scaler. The amount of silver present in the solutions was calculated by comparing the radioactivity of the test solutions with that of the standard reference solution.

Experiments with synthetic solutions showed that self-absorption corrections were not needed for the activity figures of any test solutions.

The castle assembly used in the work provided sufficient shielding to maintain the background at approximately 20 counts per minute.

Enough silver-110 was used in all tests, so that the sample aliquots had an activity at least three times as high as the background. The counting times were such that the probable error inherent in the counting data did not exceed 5% in any test.

The silver wire used to prepare the stock tracer solution had a specific activity such that the ratio of inactive to active silver atoms was approximately 10,000 to 1. It is valid to assume that the chemical behavior of the active silver added to the test samples in the form of the tracer solution was representative of the inactive silver in this solution as well as of that added as carrier in most of the tests.

In preliminary experiments, the recovery of silver in the beads varied from 96 to 98%. Because of the inherent error in the radiometric assay of the solutions prepared from these beads, it was impossible to estimate the losses to slags, cupels, etc., by measuring the difference between the recovery figures and 100%. Consequently, techniques were used which would allow the losses to be measured directly.

## LOSS OF SILVER DURING CRUCIBLE FUSION

**To Slag.** Effect of Button Weight. A series of tests was per-

formed to determine the extent of the silver loss to the slag and the degree to which this loss varied with the size of the lead button formed during crucible fusion. In each test a 165-gram portion of the standard neutral flux was salted with 5 mg. of silver (carrier plus tracer), and the amount of flour was varied. The lead buttons obtained ranged in weight from 9 to 35 grams. The slag from each test was assayed for silver as shown in the following experimental results.

| Test No. | Button Weight, Grams | Silver Lost to Slag, % |
|---|---|---|
| 1 | 9 | 1.0 |
| 2 | 13 | 0.5 |
| 3 | 19 | 0.4 |
| 4 | 26 | 0.2 |
| 5 | 35 | 0.2 |

The results indicate that silver is not completely collected, when buttons weighing less than 25 grams are formed during the fusion of charges weighing 165 grams (tests 1 to 3). With buttons weighing 25 grams or over, the silver loss appears to be constant at approximately 0.2%. However, it is not clear from the results of tests 4 and 5 whether the silver found in the reassay beads had dissolved in the slag or had been taken up by the crucible wall during the initial fusion. The latter phenomenon has been observed in work with platinum (*6*). Therefore three further tests (6, 7, and 8) were performed to determine the solubility of silver in the slag and the magnitude of any loss due to retention by the crucible wall during fusion.

The composition of the charges in these tests was identical to those used in tests 4 and 5. The slags obtained after fusion were assayed in fresh crucibles rather than in the original crucibles as in tests 4 and 5. The silver content of the resultant reassay beads corresponded to 0.2, 0.2, and 0.3%, respectively, of the silver originally present in the charge. Therefore it can be concluded that the 0.2% loss in tests 4 and 5 was due to the solubility of silver in the slag rather than to uncollected values or to pickup from the walls of the crucibles.

**Retention of Silver by Crucible Wall during Fusion.** The crucibles used for the initial fusions in tests 6 to 8 were monitored with the immersion type Geiger probe by inserting it in such a fashion that the geometry was constant for each crucible. Considerable activity was detected on the walls of the crucibles (Table I). However, it was impossible to relate the activity on the solid crucibles directly to that of the standard reference solution. To overcome this problem the crucibles were "washed" during the fusion of fresh flux with silver nitrate carrier. The buttons resulting from the fusions were cupelled to give reassay beads which were then assayed radiometrically for silver. After the washing fusion, the crucibles were again monitored with the Geiger probe. By relating the activity of the crucible walls before and after washing to that of the reassay beads, it was possible to determine the approximate percentage of silver lost to the crucibles during the initial fusion (Table I).

The results in Table I show that approximately 0.5% of the silver in the initial charge is retained by the crucible during fusion.

**Loss To Metal Sulfides with Oxidizing Fluxes.** To determine how loss of silver to slag is influenced by variation in the composition of the charge (flux plus sample), a series of tests was performed in which various metal sulfides were fused with appropriate fluxes to produce buttons weighing 30 grams or more. Copper- and nickel-bearing materials were chosen for much of this work, because they are among the most troublesome elements encountered in fire assaying, being difficult to slag off and tending to contaminate the lead button.

The slags from these tests were assayed twice for silver by the procedure described above (Table IV).

The first assay was performed by fusing the ground slag with 170 grams of the standard flux. In the second assay the slag was fused with 50 grams of litharge and 3 grams of flour.

The content of the major constituents in the sulfide materials is given in Table II and the composition of the charge used in each test is given in Table III.

The fluxes used in tests 9, 10, and 12 were similar to those recommended by Shepard and Dietrich (*9*) for charges rich in copper and nickel. The flux used in test 11 was similar to that used by Theirs, Graydon, and Beamish (*10*) for a pyrite charge.

Because the initial fusion in tests 9, 10, and 12 resulted in free-flowing slags together with the reduction of sufficient lead for good collection, the comparatively high loss of silver in these tests was probably due to the increased solubility of silver in the slag, caused by the action of copper and nickel. It is also probable that silver would have been detected in the slags after a third assay. However, it is unlikely that the amount would exceed 0.1 to 0.2% of the amount present in the initial charge and it can be concluded that the total loss of silver in these tests was 2 to 3%.

In test 11 the slag obtained in the initial fusion was very viscous and it is probable that the 3.8% loss of silver to the slag was due primarily to the incomplete collection by lead rather than to the increased solubility caused by the slagging of iron. This is substantiated by the fact that silver was not detected in the slag after the first assay.

Table I. Retention of Silver on Crucible Wall

| Test No. | Activity of Crucible Wall: After initial fusion, counts/min. | Activity of Crucible Wall: After fusion with fresh flux, counts/min. | Silver, %: Found in reassay bead | Silver, %: Retained by crucible after initial fusion (calcd.) |
|---|---|---|---|---|
| 6 | 364 | 296 | 0.07 | 0.4 |
| 7 | 415 | 343 | 0.09 | 0.5 |
| 8 | 542 | 408 | 0.12 | 0.5 |

Table II. Composition of Sulfide Materials

| Sample | Description | Cu, % | Ni, % | Fe, % | S, % |
|---|---|---|---|---|---|
| A | Chalcopyrite | 30 | | 32 | 35 |
| B | Pentlandite plus violarite plus chalcopyrite | 9 | 17 | 23 | |
| C | Pyrite | | | 41 | 47 |
| D | Cu-Ni matte | 13 | 57 | 4 | 22 |

Table III. Composition of Charges

| Test No. | Sample | Sample Weight, Grams | Weight of Flux Components, Grams: PbO | $Na_2CO_3$ | $SiO_2$ | $KNO_3$ | Flour | Weight Base Metal in Charge, Grams: Cu | Ni | Fe |
|---|---|---|---|---|---|---|---|---|---|---|
| 9 | A | 15 | 165 | 20 | 15 | 19 | .. | 4.5 | ... | 4.8 |
| 10 | B | 7.5 | 165 | 20 | 15 | 5 | .. | 0.7 | 1.3 | 1.7 |
| 11 | C | 15 | 54 | 25 | 19 | 27 | .. | ... | ... | 6.2 |
| 12 | D | 3.5 | 245 | 16 | 4 | .. | 1 | 0.5 | 2.0 | 0.1 |

Table IV. Variation of Slag Loss with Composition of Charge

| Test No. | Original Silver in Slag, %: 1st assay | Original Silver in Slag, %: 2nd assay | Remarks |
|---|---|---|---|
| 9 | 1.5 | 0.5 | |
| 10 | 1.8 | 0.1 | |
| 11 | 3.8 | None detected | Viscous slag in initial fusion |
| 12 | 2.3 | 0.3 | |

## LOSS OF SILVER DURING SCORIFICATION

Three impure lead buttons (approximately 30 grams each) containing copper

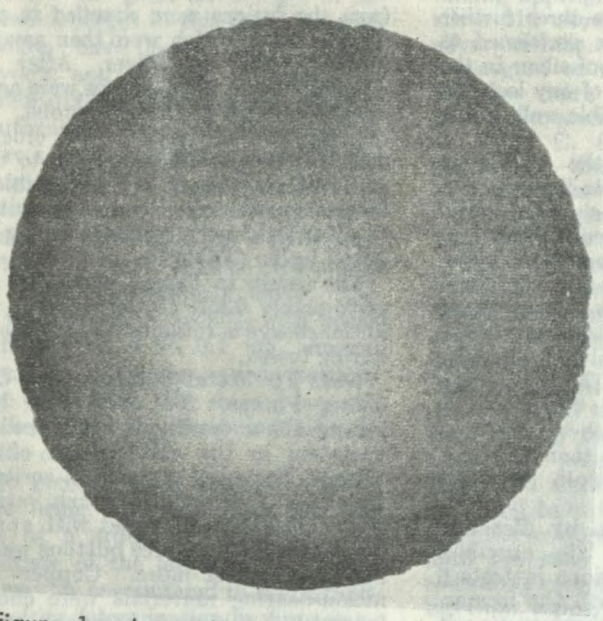

Figure 1. Autoradiograph obtained by inverting the cupel over the film

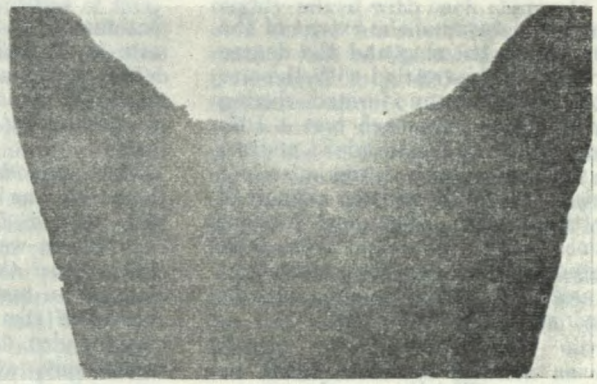

Figure 2. Autoradiograph obtained by using half of a cupel flat on its side on the film

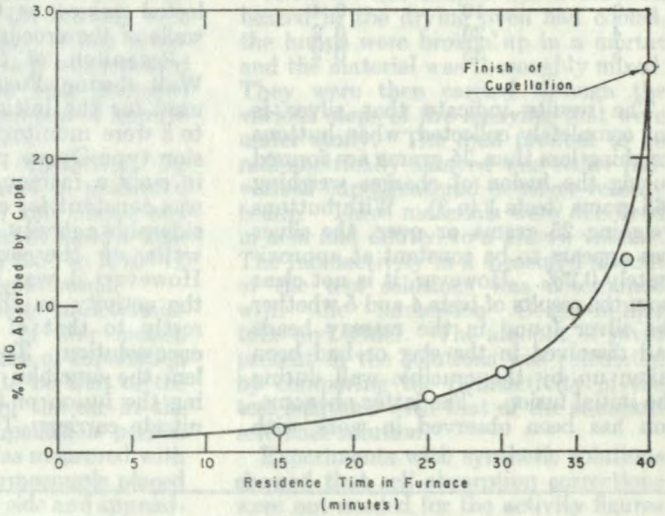


Figure 3. Amount of silver absorbed by each cupel

Buttons contained 5 mg. of silver and weighed 31 to 34 grams
Cupellation temperature 895° C., magnesia cupels

and nickel were prepared by fusion of charges similar to that used in test 12. Each button was scorified three times. The slag from each scorification was assayed radiochemically; in all cases it contained approximately 0.2% of the silver originally present in the initial charge.

## LOSS OF SILVER DURING CUPELLATION

It has long been known that the principal loss of silver in fire assaying occurs during cupellation. Among the important researches on this process are those of King, who studied the effect of air on the cupellation losses to bone ash cupels (*6*), and also the influence of the method of manufacture and of the composition of bone ash and bone ash–cement cupels on these losses (*7*).

The following series of radiochemical experiments were conducted to investigate factors that influence the loss of silver to cupels.

**Effect of Cupellation Temperature.** A number of buttons weighing 30 to 35 grams containing silver-110 and 5 mg. of silver carrier were prepared and cupelled at a series of known temperatures in 1.5-inch magnesia cupels under a suitable draft.

At the end of the cupellation period the silver bead was removed and the amount of silver-110 retained by the cupel was determined. The results of these tests are given in Table V.

Temperatures lower than 895° C. were not investigated, because this temperature approaches that of the freezing point of litharge.

The results in Table V show the importance of maintaining the lowest possible temperature during cupellation. Even at 890° to 900° C. approximately 2.5% of the silver is lost by absorption into the cupel; at temperatures in the vicinity of 1000° C. this loss is doubled.

The cupel from test 15 was used to prepare two autoradiographs (Figures 1 and 2). Figure 1 was prepared by inverting the cupel over the film. The cupel was then cut vertically through the middle and one piece was laid flat on its side on the film to prepare Figure 2. These figures show clearly how extensively the silver is distributed laterally from the bead site of the cupel, and that silver penetrates to a considerable depth into the cupel body.

**Variation of Silver Loss with Time.** To determine how the quantity of silver absorbed by the cupel varies with cupellation time, a number of buttons were prepared which ranged in weight from 31 to 34 grams and contained 5 mg. of silver. Each button was cupelled at 895° C. for a different predetermined length of time, at the end of which the cupel was removed from the furnace and the molten lead was allowed to solidify. The button was then removed from the cupel and the amount of silver absorbed by each cupel was determined.

As expected, Figure 3 shows that the silver loss is greatest near the end of the cupellation period, because the ratio of lead to silver is rapidly decreasing at this time.

**Effect of Bead Size.** To determine the variation in silver loss with bead size, a number of buttons weighing 30 to 33 grams were prepared, with each button containing a different quantity of silver. These buttons were cupelled in 1.5-inch magnesia cupels at 895° C. The amount of silver absorbed by the cupels was determined. The results clearly show that the quantity of silver absorbed by the cupel varies with the quantity of silver present in the button. However, the percentage loss of silver decreases as the weight of the bead increases.

In the range of bead weights investigated by Nakamura and Fukami (*8*) (2 to 0.1 mg.), the loss of silver to the cupel ranged from 5 to 30% of the silver originally taken.

**Effect of Button Weight.** A number of buttons of different weights, each containing 5 mg. of silver carrier, were cupelled at 895° C. The amount of silver absorbed by the cupels was determined as before.

Table V shows that, in the range investigated, the amount of lead in the button to be cupelled has no effect on the extent of the loss of silver to the cupel by absorption.

**Effect of Silver-Gold Ratio.** Because silver and gold often occur together, experiments were performed to determine the effect of the presence

Table V. Effect of Experimental Conditions on Loss of Silver to Magnesia Cupels

| Test No. | Cupellation Temp., ° C. | Silver Lost to Cupel, % |
|---|---|---|
| | Effect of Cupellation Temperature | |
| 13 | 1010 | 5.2 |
| 14 | 980 | 4.5 |
| 15 | 940 | 3.5 |
| 16 | 935 | 3.2 |
| 17 | 923 | 3.1 |
| 18 | 911 | 3.2 |
| 19 | 895 | 2.6 |
| | Effect of Bead Size | |
| | Silver in Button, Mg. | Silver Absorbed by Cupel, Mg. |
| 20 | 2 | 3.0 0.06 |
| 21 | 5 | 2.6 0.13 |
| 22 | 10 | 2.6 0.26 |
| 23 | 25 | 2.3 0.58 |
| 24 | 50 | 1.8 0.90 |
| 25 | 100 | 1.6 1.6 |
| | Effect of Button Weight | |
| | Button Weight, Grams | |
| 26 | 11 | 2.5 |
| 27 | 18 | 2.6 |
| 28 | 24 | 2.5 |
| 29 | 26 | 2.5 |
| 30 | 32 | 2.6 |
| | Effect of Ag/Au Ratio | |
| | Ag/Au | Ag + Au in Button, Mg. |
| 31 | 1:1 | 10 2.0 |
| 32 | 2:1 | 9 2.2 |
| 33 | 3:1 | 12 2.2 |
| 34 | 5:1 | 12 2.5 |
| 35 | 8:1 | 9 2.6 |
| 36 | 10:1 | 11 2.6 |
| 37 | 25:1 | 26 2.2 |
| 38 | 50:1 | 51 1.8 |
| 39 | 100:1 | 101 1.5 |

Table VII. Effect of Cupel Type on Silver Lost to Cupel

| Cupel Type | Silver Lost to Cupel, % | Average |
|---|---|---|
| A | 2.2 | 2.3 |
| | 2.1 | |
| | 2.6 | |
| B | 2.3 | |
| | 2.4 | 2.4 |
| | 2.4 | |
| C | 2.8 | |
| | 2.9 | 3.0 |
| | 3.2 | |
| D | 2.4 | |
| | 2.4 | 2.4 |
| | 2.4 | |

of gold on the cupel absorption of silver during cupellation.

A number of buttons (30 to 35 grams) with a different silver to gold ratio in each, were cupelled at 895° C. to give silver-gold beads. The per cent of silver lost to the cupels was determined in the usual manner (Table V).

It is apparent from the results in Table V that gold affords a protective action on silver during cupellation, when the ratio of silver to gold in the button is 3 to 1 or less (tests 31 to 33). At higher ratios (tests 34 to 39), the loss of silver becomes identical with that obtained with silver alone (tests 22 to 25). It is of interest to note the bead size effect in tests 37 to 39, where the percentage loss decreases with an increase in bead weight.

**Effect of Cupel Shape and Composition.** Most cupels are made from magnesia or bone ash and are available from the factory or the laboratory press in various sizes and shapes. To determine the effect of cupel shape and composition on the silver loss during cupellation, four types of cupels were selected (Table VI).

Table VI. Description of Cupel

| Type | Shape | Diameter, Inches | Weight, Grams | Composition | Manufactured |
|---|---|---|---|---|---|
| A | Cylindrical | 1 | 25 | Magnesia | Factory |
| B | Cylindrical | 1 | 25 | Magnesia | Laboratory |
| C | Cylindrical | 1 | 25 | Bone ash | Laboratory |
| D | Tapered | 1.5 | 45 | Magnesia | Factory |

A series of experiments was performed in which buttons of appropriate weight, containing silver-110 tracer and 5 mg. of silver carrier, were cupelled at 895° C. in the cupels described above. The per cent of the original silver lost to the cupels was determined as before and the results of these tests are given in Table VII.

The results in Table VII show that the silver loss in bone-ash cupels is approximately 25% higher than that in magnesia cupels. In the two types of magnesia cupels investigated, the shape of the cupel did not influence the silver loss.

## CONCLUSIONS

Radiochemical techniques using silver-110 have provided a direct and sensitive means for measuring the loss of silver encountered in its determination by fire assay. Cupellation losses vary with temperature, the size of the bead, the gold content of the bead, and the composition of the cupel. Even under optimum conditions the loss of silver to the cupel is approximately 2%.


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## LITERATURE CITED


(1) Allen, W. F., Beamish, F. E., ANAL. CHEM. 22, 451 (1950).
(2) Barefoot, R. B., Beamish, F. E., *Ibid.*, 24, 840 (1952).
(3) Chopin, L., *Chim. anal.* 33, 94 (1951).
(4) Coburn, H. G., Beamish, F. E., ANAL. CHEM. 28, 1297 (1956).
(5) Hoffman, I., Beamish, F. E., *Ibid.*, 28, 1188 (1956).
(6) King, J. T., *Univ. Toronto Bull.* 6, 255 (1926).
(7) *Ibid.*, No. 147, 50 (1934).
(8) Nakamura, Y., Fukami, K., *Japan Analyst* 6, 687 (1957).
(9) Shepard, O. C., Dietrich, W. F., "Fire Assaying," McGraw-Hill, New York, 1940.
(10) Theirs, R., Graydon, W., Beamish, F. E., ANAL. CHEM. 20, 831 (1948).



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