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DETERMINATION OF MERCURY BY CONTROLLED POTENTIAL COULOMETRY: AN OUTLAY FOR AUTOMATIC PERFORMANCE

by

H. MUNTAU and R. CENCI

1974

Joint Nuclear Research Centre
Ispra Establishment - Italy
Chemistry Division
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The mercury amounts needed for each determination are ranging from ca. 40 mg to 10 μg. The precision obtainable is better than 0.25 % for the higher concentration range, e.g., between 1 and 40 mg.
The reduction does not need to be completed when plotting \( \log i \) vs \( t \).

Full automation of the entire analytical procedure, including sampling, sample treating, analysis and data processing, seems possible.
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ABSTRACT

The determination of mercury by controlled-potential coulometry in presence of CaBr₂-concentrations between 0.05 and 1.5 M and HBr-concentrations between 0.1 and 1.0 M is possible using perchloric acid as a basal electrolyte.

The mercury amounts needed for each determination are ranging from ca. 40 mg to 10 μg. The precision obtainable is better than 0.25 % for the higher concentration range, e.g., between 1 and 40 mg.

The reduction does not need to be completed when plotting log i vs t.

Full automation of the entire analytical procedure, including sampling, sample treating, analysis and data processing, seems possible.
TABLE OF CONTENTS

1. INTRODUCTION 5
2. EXPERIMENTAL 6
  2.1. Apparatus and reagents 6
  2.2. Procedure 8
  2.3. Preparation of standards 8
  2.4. Measurements on Standard I 9
    2.4.1. Concentration range 10
    2.4.2. Concentration of basal electrolyte 11
  2.5. Measurements on Standard II 12
    2.5.1. Influence of CaBr₂-concentration 12
3. OUTLINE OF AN AUTOMATIC SYSTEM 14
4. DISCUSSION 16
5. CONCLUSION 19
1. INTRODUCTION

Production of hydrogen by nuclear heat [1, 1a] involves certain cycles of intermediate reactions, the most extensively studied one being the following:

1) \[ \text{CaBr}_2 + 2 \text{H}_2\text{O} \xrightarrow{700^\circ} \text{Ca(OH)}_2 + 2 \text{HBr} \]

2) \[ 2 \text{HBr} + \text{Hg} \xrightarrow{250^\circ} \text{HgBr}_2 + \text{H}_2 \]

3) \[ \text{HgBr}_2 + \text{Ca(OH)}_2 \xrightarrow{200^\circ} \text{CaBr}_2 + \text{HgO} + \text{H}_2\text{O} \]

4) \[ \text{HgO} \xrightarrow{600^\circ} \text{Hg} + \frac{1}{2} \text{O}_2 \]

The efficiency of any production plant using such a cycle will depend strongly on the yield of the single reactions. Analytical control of the reaction products is therefore unavoidable.

Whereas the analytical problems involved with the hydrogen production step were studied in detail [2], the present study is dedicated to the HgO-precipitation-and CaBr\textsubscript{2}-regeneration step (3).

From an analytical point of view, this step seems the most incomplete one, mainly due to complexation phenomena occurring in the presence of the virtually high Br\textsuperscript{-} concentration.

Marcus [3] pointed out, that in cases of Br\textsuperscript{-} concentrations of \( n > 2 \) in the presence of \( n \) Hg\textsuperscript{2+} exclusively the complexes \[ \text{HgBr}_3 \] and \[ \text{HgBr}_4 \] will be formed whereas the formation of Hg\textsuperscript{2+}, HgBr\textsuperscript{+} and HgBr\textsubscript{2} will be negligible. Regarding the CaBr\textsubscript{2} concentration, certainly part of the HgBr\textsubscript{2} will escape the precipitation and be transferred to the hydrolysis step 1).
Therefore, the residual mercury bromide should be monitored, if possible, completely automatically, regarding also the relatively high and unstable temperature of the CaBr$_2$- solution.

As an analytical method, which appears to meet best the above mentioned requirements, we have chosen controlled potential coulometry (CPC) using a mercury cathode. We did so for the following reasons:

- CPC as an absolute method does not need chemical calibration and standards;
- it will not suffer from electrode poisoning, passivation effects and other surface alterations, as solid electrodes necessarily do;
- varying temperatures of the samples do not show unfavourable influence;
- varying composition of the sample, e.g. increasing or decreasing concentrations of CaBr$_2$, Ca(OH)$_2$, HBr are not expected to influence the results;
- wide concentration ranges between 40 mg/ml and 20/ug/ml;
- the precision to be obtained (RSD = 0,1 - 0,2 %)
- regarding the actual design of now-a-days coulometers and the constancy of its cell elements, CPC appears to be the most suitable technique for automatization.

2. EXPERIMENTAL

2.1. Apparatus and reagents

The ORNL electronic controlled-potential coulometer 2005-x50 was used throughout this study. This instrument is an improved version of the one designed by BOOMAN and has been
described in more details by LE DUIGOU and LAUER [5].

It is designed to supply electrolysis current to the electrode system while maintaining the potential of the working electrode - in most cases a mercury electrode - at some chosen value with respect to a reference electrode. Polarity of the working electrode and direction of current flow can be chosen to run either reduction or oxidation titrations.

A portion of the current that flows is integrated electronically by an integrating operational amplifier such that the charge accumulated is a measure of the coulombs that have been used. The integral is read out as a voltage with aid of a Keithley Multimeter. (FIG. 1).

The quantity of the substance reduced or oxidized is then calculated from the voltage by means of Faraday's law.

The reaction vessel used in this study was designed and is used for routine measurements of uranium (FIG. 2).

It consists of a 50-ml glass vessel with a sealed-in platinum contact, a plexiglas cap, which supports a Radiometer Calomel Reference electrode, a separated electrode compartment and an argon inlet tube.

Stirring is provided by a glass disk, about 1 cm in diameter and positioned horizontally at the mercury-electrolyte interface, with a 6 mm glass rod sealed at its center and protruding through the cap, so that it can be driven by an 1500 r.p.m. motor.

The cell is equipped with a two-way inlet for argon, which in a first step deaerates the electrolyte prior to analysis and in second step keeps the space over the electrolyte free
from penetrating air.

The mercury used as pool electrode was a Mercure-Industries product and is 99.99999 % pure. The argon used for sweeping out air from the cell and the electrolyte was U.P.F. quality. Other reagents used, as mercury bromide, calcium bromide and several acids, were reagent grade and used without further purification.

2.2. Procedure

All measurements were performed in the same way:
After pouring 7 ml of mercury and usually 5 ml of the basal electrolyte, mostly 5 M HClO₄, a potential of - 0.3 V was applied to the mercury electrode and while stirring the solution and sweeping argon through it, the current through the cell has allowed to reach its background value, say 30/μA.

Then the integrator was zeroed and a small sample, defined either by volume or weight, introduced into the cell. The argon then was allowed to pass over the surface of the liquid and the electrolysis continued until the background current is reached again. The coulombs used are then a direct measure of the quantity of mercury reduced.

2.3. Preparation of standards

The questions to be answered during this study were the following:
- is it in principle possible to apply CPC to the determination of mercury?
- if yes, how would complexing agents, as for example CaBr₂ and HBr, influence the reduction?
- would it be possible to automate the several steps involved with sampling and measurement, regarding the special condi-
tions of the system, from which the samples are to be taken?

To answer the first question, an extremely pure mercury-sulphate solution was prepared by dissolving a weighed amount of 99.99999 % pure mercury in supra-pure nitric acid by gentle warming after covering the mercury by double-distilled water. The mercury solution obtained was fumed twice with suprapure sulphuric acid and the solution diluted to defined weight and volume (Standard I = 12.76 mg Hg$^{2+}$/g or 15.85 mg Hg$^{2+}$/ml.

A second standard solution has been prepared from HgBr$_2$, suspended in excess in twice-distilled water and allowed to stand for 3 days at room temperature. The remaining excess of HgBr$_2$ was then filtered off. Cyanometric analysis of the solution yielded a value of 2.90 mg Hg/ml, corresponding to 5.21 mg HgBr$_2$/ml or a molarity of 0.0145, value considerably lower than the maximum solubility at 25° of 0.0170, given by GMELIN [7]. It has been reported, on the other hand, that HgBr$_2$-solutions, prepared by heating, after cooling will contain more HgBr$_2$ due to hydrolysis occurring while heating and owing to the much higher solubility of HgBr$_2$ in even very diluted HBr-solutions [8] (Standard II = 2.9 mg/ml).

2.4. Measurements on Standard I

The cell was prepared as described under 2.2, using 1 M HClO$_4$ as basal electrolyte. Weighed amounts of Standard I were introduced and reduced.

A second series of measurements were performed introducing each 1 ml with aid of a calibrated pipette. Results are shown in Table 1.
2.4.1. Concentration range

The ORNL Automatic Coulometer has been designed for measurements in the Milligram range. Nevertheless, stepwise amplification is possible and uranium amounts as low as 9/ug have been determined successfully [9].

Maximum capacity of our integrator is equal to about 40 mg of mercury. Diluting standard I a series of measurements has been performed down to a total amount of about 12/ug of mercury. 1 M HClO₄ was used as a basal electrolyte. Results are shown in Table II.

* RSD (Relative Standard Deviation) = \( \frac{6 \cdot 100}{\bar{x}} \)
**TABLE II**

<table>
<thead>
<tr>
<th>Amplification</th>
<th>Mercury given* (mg)</th>
<th>$\bar{x}$</th>
<th>n</th>
<th>RSD (%)</th>
<th>Basal electrolyte</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 x 1</td>
<td>38,28</td>
<td>38,39</td>
<td>6</td>
<td>0,17</td>
<td>1 M HClO₄</td>
</tr>
<tr>
<td>1 x 1</td>
<td>12,76</td>
<td>12,83</td>
<td>5</td>
<td>0,14</td>
<td></td>
</tr>
<tr>
<td>1 x 10</td>
<td>1,276</td>
<td>1,289</td>
<td>5</td>
<td>0,18</td>
<td></td>
</tr>
<tr>
<td>10 x 10</td>
<td>0,128</td>
<td>0,129</td>
<td>5</td>
<td>0,20</td>
<td></td>
</tr>
<tr>
<td>10 x 100</td>
<td>0,013</td>
<td>0,014</td>
<td>5</td>
<td>2,2</td>
<td></td>
</tr>
</tbody>
</table>

* Values approximatively defined by diluting Standard I.

2.4.2. **Concentration of basal electrolyte**

Sulphuric acid and perchloric acid due to their indifferent chemical nature are favorite basal electrolytes in controlled-potential coulometry. Regarding the high Ca$^{2+}$ concentration of our actual samples, perchloric acid has been chosen as basal electrolyte. The concentration of the basal electrolyte very often influences both reduction potential and completeness of reaction. Hence, 1-ml-amounts of standard I were reduced varying the concentration of perchloric acid from 0.5 M to 5 M (Table III).
TABLE III

<table>
<thead>
<tr>
<th>Mercury given</th>
<th>HClO₄</th>
<th>n</th>
<th>$\bar{x}$ (mg)</th>
<th>RSD (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>12.76</td>
<td>0.5</td>
<td>6</td>
<td>12.81</td>
<td>0.17</td>
</tr>
<tr>
<td>12.76</td>
<td>1.0</td>
<td>5</td>
<td>12.83</td>
<td>0.14</td>
</tr>
<tr>
<td>12.76</td>
<td>2.0</td>
<td>6</td>
<td>12.78</td>
<td>0.18</td>
</tr>
<tr>
<td>12.76</td>
<td>3.0</td>
<td>6</td>
<td>12.80</td>
<td>0.15</td>
</tr>
<tr>
<td>12.76</td>
<td>4.0</td>
<td>6</td>
<td>12.79</td>
<td>0.17</td>
</tr>
<tr>
<td>12.76</td>
<td>5.0</td>
<td>6</td>
<td>12.83</td>
<td>0.14</td>
</tr>
</tbody>
</table>

Considering the mercury reduction not influenced by varying HClO₄ concentrations, we decided to perform all further measurements in 5 M HClO₄.

2.5. Measurements on Standard II

A series of measurements has been performed on 1 ml-aliquotes of standard II yielding $\bar{x} = 2.93$ (theoretical values = 2.90 mg Hg) and RSD = 0.22 %.

Obviously a small positive bias occurred during the measurements, owing probably to certain differences in the analytical techniques employed, e.g., the cyanometric technique and CPC.

2.5.1. Influence of CaBr₂-concentration

Several CaBr₂-solutions, varying in concentration between 10 and 30 % (W/V) previously saturated with Ca(OH)₂ were prepared.
1 ml-Aliquots of Standard II were introduced into the cell, (containing 5 M HClO₄ as basal electrolyte), added the appropriate amounts of CaBr₂-solution and mixed by bubbling through argon. Table V shows the results.

### TABLE V

<table>
<thead>
<tr>
<th>CaBr₂ (M)</th>
<th>Mercury given (mg)</th>
<th>Mercury found (mg)</th>
<th>n</th>
<th>RSD</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.05</td>
<td>2.90</td>
<td>2.92</td>
<td>5</td>
<td>0.21</td>
</tr>
<tr>
<td>0.15</td>
<td>2.90</td>
<td>2.93</td>
<td>5</td>
<td>0.19</td>
</tr>
<tr>
<td>0.25</td>
<td>2.90</td>
<td>2.94</td>
<td>5</td>
<td>0.17</td>
</tr>
<tr>
<td>0.75</td>
<td>2.90</td>
<td>2.95</td>
<td>5</td>
<td>0.19</td>
</tr>
<tr>
<td>1.5</td>
<td>2.90</td>
<td>2.93</td>
<td>5</td>
<td>0.20</td>
</tr>
</tbody>
</table>

### 2.5.2. Influence of free HBr

Without attempting to decide whether or not free HBr may occur under operation conditions, its possible presence has to be considered from an analytical point of view.

Using a calcium bromide solution containing no Ca(OH)₂ for maintaining a constant CaBr₂-concentration level (1.5 M) the influence of increasing amounts of HBr on the determination of 1 ml-aliquots Standard II was investigated. Results are shown in Table VI.
TABLE VI

<table>
<thead>
<tr>
<th>HBr (M)</th>
<th>CaBr₂ (M)</th>
<th>Mercury given (mg)</th>
<th>Mercury found (mg)</th>
<th>n</th>
<th>RSD</th>
</tr>
</thead>
<tbody>
<tr>
<td>0,1</td>
<td>1,5</td>
<td>2,90</td>
<td>2,93</td>
<td>5</td>
<td>0,22</td>
</tr>
<tr>
<td>0,5</td>
<td>1,5</td>
<td>2,90</td>
<td>2,95</td>
<td>5</td>
<td>0,16</td>
</tr>
<tr>
<td>1,0</td>
<td>1,5</td>
<td>2,90</td>
<td>2,95</td>
<td>5</td>
<td>0,19</td>
</tr>
</tbody>
</table>

3. OUTLINE OF AN AUTOMATIC SYSTEM

An attempt has been undertaken outlining roughly the possibilities of a system that enables fully automatic sampling, sample treatment, coulometric analysis and evaluation of data obtained.

Of course, this first attempt is far from being complete and perfect and closer studies of the design problems will be necessary.

Roughly, the sequence of operation could be the following: (See also FIG. 3).

- The central programming unit (CPU) activates the coulometer which starts reduction, in order to clean the vessel and the surface of the mercury cathode. At some pre-determined value, say 30/uA of background current, the coulometer stops and the argon stream is switched to blow on the basal electrolyte surface using a magnetically operated two-way valve.

- CPU opens an electromagnetic valve attached to the loop allowing to flow a portion of the sample into the expansion vessel.
After appropriate expanding and cooling and diluting the sample is forced into the sampling system by means of a small pump.

The sampling system takes an aliquot of the sample and injects it into the cell. A similar sampling system was just recently described by ROSSI and PICKFORD [10] for automatic sampling and sample injection, applied to the problem of high purity water analysis by flameless atomic-absorption spectroscopy.

The coulometer is switched on and electrolysis proceeded until the background current (30 μA) is reached.

The voltage accumulated by integrating the electrolysis current is supplied to the CPU, thus zeroing the integrator for the next measurement. The values obtained for single determinations are to be compared automatically to given nominal values. Large deviations from these values may trigger an immediate repetition of the measurement and/or may operate an alarm system.

CPU operates the pump in order to remove the basal electrolyte from the cell and causes the injection of fresh basal electrolyte into the cell.

Finally the argon valve is switched to bubble argon through the basal electrolyte and the system is ready for the next cycle.
4. DISCUSSION

Obviously chemical parameters hitherto considered do not influence the determination of mercury in Br\textsuperscript{-} containing complexing media.

The relative standard deviations obtained are somewhat higher than the optimum values of about 0.05 % which are to be obtained in high precision coulometry, they agree, however, well with average values observed in routine measurements.

More critical is the somewhat embarrassingly long time needed for the accomplishment of the electrolyte process. Here remedial measures are possible paying a closer look to the fundamentals governing the electrode reactions.

Keeping the potential of the working electrode constant, the current at the time \( t \) will be:

\[
 i_t = n F \left( \frac{\text{d}N}{\text{d}t} \right) = n \cdot F \cdot N \cdot D \cdot q/v \cdot \delta \quad (1)
\]

\[
 \text{d}N/\text{d}t = \text{number of moles reacting each second at the electrode surface}
\]

\[
 D = \text{Diffusion coefficient}
\]

\[
 q = \text{electrode surface}
\]

\[
 v = \text{volume}
\]

\[
 \delta = \text{thickness diffusion layer}
\]

\[
 N = \text{number of moles}
\]

and correspondingly \( i_0 \), the current at the beginning of the electrolysis is:

\[
 i_0 = n \cdot F \cdot N_0 \cdot D \cdot q/v \cdot \delta \quad (2)
\]
Hence, the current at any time \( t \) is proportional to the number of moles of substance not yet reacted.

Since the transport of depolarizer to the electrode surface is diffusion controlled, \( \frac{dN}{dt} \) is also approximately:

\[
\frac{dN}{dt} \approx N \cdot D \cdot q / \nu \cdot \delta \quad (3)
\]

which yields:

\[
\frac{dN}{N} = \frac{D \cdot q}{r \cdot \delta} \cdot dt \quad (4)
\]

and:

\[
N = N_0 \cdot e^{-D \cdot q \cdot t / \nu \cdot \delta} \quad (5)
\]

combined with equations (1) and (2) \( i_t \) is:

\[
i_t = i_0 \cdot e^{-k \cdot t} \quad (6)
\]

where \( k \) is:

\[
k = D \cdot q / \nu \cdot \delta \quad (7)
\]

From (6) it becomes evident that \( i_t \) decreases exponentially. Plotting log \( i \) vs \( t \) a straight line is obtained which yields \( k \). This behaviour has been experimentally verified (11). The equations (7) and (8) show how shorter electrolysis intervals may be obtained. As the constant \( k \) is directly proportional to \( q / \nu \), high stirring rates will decrease and higher temperature will increase \( D \), hence decreasing \( t \).
Moreover, it is not necessary to accomplish the reduction process down to its background current. The amount of electricity used is defined as:

\[ Q = \int_{0}^{\infty} i_0 \cdot e^{-kt} \cdot dt = i_0 / 2,303 \cdot k \]  

(8)

and may be determined simply by plotting \( \log i \) vs \( t \). Then \( i_0 \) is the intersection with the ordinate and \( k \) the slope. Therefore, only a short interval must be recorded. Extrapolation and evaluation of data could be performed automatically.

A second point of interest is the concentration range to be covered. The currently used ORNL coulometer will work with agreeable accuracy between 40 mg/ml and 10/ug/ml or 0.2 M to 5 \( \cdot \) \( 10^{-5} \) M respectively. The new GOKA coulometer, designed at the KFA Karlsruhe for work in the low concentration range, will reach \( 5 \cdot 10^{-6} \) M, but is limited to about 0.02 M (4 mg) as maximum capacity.

Only a few potentiostat-coulometers are recently offered on the market and they seldom meet the special requirements wanted by the customer.

It is therefore understandable that people, interested in this branch of electroanalytical chemistry, usually attempts to construct their own coulometer. To our knowledge, high precision coulometers were built and are currently in use at CEA Fontaney-aux-Roses [12], BCMN Geel [13] and the Lawrence Radiation Lab., Livermore, Calif. [6]. With special effort it is possible to extend the concentration range over two more orders of magnitude and to lower the relative standard deviation to 0.01 %.
5. **CONCLUSION**

The determination of mercury by controlled-potential coulometry in presence of CaBr$_2$-concentrations between 0.05 and 1.5 M and HBr-concentrations between 0.1 and 1.0 M is possible using perchloric acid as a basal electrolyte.

The mercury amounts needed for each determination are ranging from ca. 40 mg to 10/ug. The precision obtainable is better than 0.25 % for the higher concentration range, e.g., between 1 and 40 mg.

The reduction does not need to be completed when plotting log $i$ vs $t$.

Full automation of the entire analytical procedure, including sampling, sample treating, analysis and data processing, seems possible.
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    BCMN Geel (Personal Communication).
FIGURE 2

1 PLATINUM CONTACT
2 REFERENCE ELECTRODE
3 COUNTER ELECTRODE
4 MERCURY CATHODE
5 ARGON - INLET
6 ARGON AIR - PROTECTION JET
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Alfred Nobel
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