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Automated Controlled-Potential Coulometer for Plutonium Determination

University of California

LOS ALAMOS SCIENTIFIC LABORATORY

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Automated Controlled-Potential Coulometer for Plutonium Determination

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D. D. Jackson

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AUTOMATED CONTROLLED-POTENTIAL COULOMETER FOR PLUTONIUM DETERMINATION

by

R. M. Hollen and D. D. Jackson

ABSTRACT

The automated controlled-potential coulometer for the determination of plutonium described in this report is the second in a series of automated instruments designed to determine plutonium and uranium contents in nuclear fuel cycle materials. The measurement precision of the instrument is 0.1% relative standard deviation at the 5-mg plutonium level.

A highly selective method of analysis was developed, involving reduction of plutonium to Pu(III) in a 5.5 *M* hydrochloric acid-0.015 *M* sulfamic acid electrolyte; oxidation of diverse ions, but not Pu(III); addition of phosphate complexant to reduce the Pu(III)-Pu(IV) potential; and oxidation of Pu(III) to Pu(IV) as the measurement step.

A turntable rotates as many as 24 electrolysis cells to a measurement station where all chemical and electrical operations take place. Reagents are added, solutions electrolyzed, data printed and plotted, and the cell and electrodes cleaned. The instrument controller is a desk-top programmable calculator. A paper-tape printout gives date and time, a sample identification number, electrolysis elapsed times, and numbers of coulombs for each electrolysis.

Construction details of the mechanical and electrical systems of the instrument and control-system software are described, along with instrument preoperational adjustments and tests and sample analysis operations.

I. INTRODUCTION

This automated controlled-potential analyzer, the second in a series of automated instruments designed to determine uranium and plutonium contents, was developed under sponsorship of the Office of Safeguards and Security, US Department of Energy. The first instrument was an automated spectrophotometer for the highly selective determination of low milligram amounts of uranium and plutonium with precisions of about 1% relative standard deviation (RSD).¹ This instrument was

later modified² to use an additional spectrophotometric method³ to selectively determine microgram amounts of uranium. The automated controlled-potential analyzer described here determines low milligram amounts of plutonium with high selectivity and a 0.1% RSD.

The chemical method developed specifically for the instrument has been published⁴ and is discussed briefly here. Major mechanical and electrical components of the instrument are discussed, and test data that show its precision capability are given. Appendix A provides preoperational adjustments and tests, and an operational

procedure for determining plutonium is given in Appendix B. Other Appendixes provide details of mechanical components including construction drawings (Appendix C), electrical components including circuit diagrams (Appendix D), and control-system software (Appendix E).

II. CHEMICAL METHOD

The major criteria for the chemical method were high selectivity, precision of 0.1% RSD at the 5-mg plutonium level, determination of total plutonium regardless of initial oxidation state, and long-term electrode stability. Because no available method met all requirements, we developed a highly selective, controlled-potential coulometric method for determining total plutonium.⁴ In a 5.5M HCl-0.015M sulfamic acid electrolyte, plutonium is reduced at 0.25 V to Pu(III). (All potentials are relative to the saturated-calomel electrode.) Many diverse ions, including iron, are oxidized at a potential of 0.57 V. Phosphate (as NaH_2PO_4) is added to make the electrolyte 1M in phosphate, which lowers the Pu(III)-Pu(IV) potential, and Pu(III) is oxidized to Pu(IV) at 0.68 V as the measurement step. All electrolyses are performed to a 50- μA current endpoint. The tolerances of 53 metal ions and 25 nonmetal anions are given in Ref. 4. Metal cations normally present in nuclear fuel cycle materials do not interfere at an equal mole ratio relative to 0.02 mmol (5 mg) of plutonium. Many anions are without effect, and most interfering ones are removed by perchloric acid fuming. Under normal operating conditions, the platinum-gauze working electrode remains stable for months.

Precision for the method is typically <0.1% RSD for a single determination at the 5-mg plutonium level. This represents the total random measurement error, including components from weighing and chemical operations.

III. INSTRUMENT DESCRIPTION

A. General Features

The automated controlled-potential coulometer, shown in Fig. 1, features a turntable that rotates up to 24 electrolysis cells to a measurement station. All operations are automated, including sample transport, reagent

Fig. 1.
Automated controlled-potential coulometer.

deliveries, the reduction and two oxidation electrolyses, data computation with output on paper tape and a display of the electrolyses (current vs time), and cleaning of cells, stirrer, and electrodes. The analyst delivers weight or volume portions of samples, containing nominally 5 mg of plutonium in a volume up to 2 mL, into cells, places the cells in the turntable, and initiates the automated operation. The analysis rate is about one per 30 min. A programmable calculator controls all mechanical and electrical operations and processes data.

The mechanical system is designed to fit in a single open front box 0.91 m wide, 0.79 m deep at the base, and 0.84 m high. A 0.30-m-high front opening gives ready accessibility to the turntable, measurement station, and reagent containers. Instrument layout provides accessibility to the measurement station components and ease of servicing. The mechanical components of the instrument are designed for long-term reliability. Most parts are fabricated of aluminum, Teflon, Kel-F, or nylon to minimize corrosion. Waste solution is collected in a bottle outside the box. Interlocks are incorporated to

minimize the possibility of instrument damage or a faulty analysis. Indication of possible damage to the instrument terminates operation. If only the analysis is affected, automated operation continues, but the analysis is terminated.

Most electronic components are available commercially for ready replacement and simplified maintenance. These components, located outside the box, connect to the mechanical system by four cables.

B. Control System

The control system is shown schematically in Fig. 2. A Hewlett-Packard 9825A Programmable Calculator with 98210A String-Advanced Programming and 98216A Plotter-General I/O - Extended I/O ROMs and 98034A HPIB and 98033A BCD Interface Cards controls all mechanical and electrical operations, processes electrolysis data, prints results on paper tape, and plots graphs of the electrolyses. Programs are written in HPL, a high-level language. The sequence and timing of the operations are controlled accurately by the calculator. Real-time decisions affecting the analysis are made and acted upon. Separate programs (Appendix E) are used for processing blanks (for background correction), analyzing samples, and verifying correct electrical operation. Special function keys on the calculator allow manual control of all operations to facilitate adjustments.

Interfaced and under calculator control are a digital-to-analog converter (DAC), a digital multimeter (DMM), a scanner, a real-time clock, and a digital plotter. The DAC allows selection of electrode potentials by supplying a highly stable voltage to the external signal input of

the potentiostat. The DMM measures electrolysis current and applied potential and serves as an analog-to-digital converter to read in data to the calculator. The DMM also monitors the interlocks. Instrument operation stops if (1) the inert-gas supply ceases, (2) a cell does not lower correctly into position in the turntable, (3) the turntable locking mechanism fails, (4) the turntable does not rotate to its correct angular position, or (5) the cell does not raise correctly into position at the measurement station. The potentiostat output is monitored. If an electrical malfunction is indicated, the potentiostat output is switched off and mechanical operation is stopped. After the last sample is analyzed and the cell, stirrer, and electrodes are cleaned, rinse solution is added to immerse the working electrode, and operation is stopped.

The real-time clock monitors electrolysis times. If elapsed time exceeds preset values considered to affect results adversely, that analysis is terminated and automated operation continues. The produced graph of log current vs time provides diagnostic information. The scanner selects the input signal to the DMM and controls the mechanical operations through contact closures. Photo-isolated relays, shown on the left in Fig. 3, isolate the calculator and potentiostat from large transients generated by solenoids and motors. A Princeton Applied Research Corporation 173 Potentiostat-Galvanostat and 179D Digital Coulometer do the electrolysis. The potentiostat is modified for calculator control of the selector switch, and the coulometer is interfaced for calculator control, as detailed in Appendix D.

Fig. 2.
Control system schematic.

Fig. 3.
Relays and mechanical components.

C. Measurement Station

At this station, a rigid Teflon support holds the electrodes, reagent and inert-gas delivery tubes, stirring unit, and rinsing unit (Fig. 4). The fixed support prevents flexing of electrical leads and tubing, which could cause failure. The positions of the cell components provide reliable electrolyses and servicing access.

The working electrode is a three-layer, folded, 45-mesh platinum gauze with two 14-gauge platinum wires fused to it. The large surface area provides fast electrolyses. The two wires pass through close-fitting holes in the Teflon support to attain alignment rigidity, required for correct cell positioning and effective cleaning. The alignment procedure is detailed in Appendix A.

The counter electrode is 18-gauge platinum wire with one end folded for increased surface area. The reference electrode is a Peckman Model 392-70 saturated-calomel electrode. Commercial bridge tubes with porous Vycor frits, detailed in Appendix C, are used for both the counter and reference electrodes. The Vycor frits, which provide high electrical conductivity at very low flow

rates of electrolyte, are attached with heat-shrinkable Teflon tubing. The tip of the reference bridge tube is located adjacent to the working electrode for best control. The counter electrode is in the geometric center of the working electrode to attain a uniform potential gradient. A drawing of the cell is given in Appendix C.

Effective stirring is provided by a glass paddle driven by an 1800-rpm synchronous motor mounted rigidly above the Teflon support. The paddle, about 1.0 cm wide, 0.6 cm high, and 0.3 cm thick, is made by flattening the heated end of a 0.5-cm-diam glass rod. The Teflon support provides a low-friction bearing surface preventing wobble. The stirring rod is connected to the motor spindle by a flexible coupling of rubber tubing.

A 2.7-atm inert gas supply of argon or nitrogen with <400 ppm O_2 provides the cell purge and drives all pneumatic and hydraulic operations. The purge gas pressure is reduced to 0.2 atm with a miniature pressure regulator inside the mechanical system. A needle valve regulates the purge flow to 750 cm^3/min , and an in-line flow switch monitors the purge flow. The inlet and outlet tubes are 0.5-cm-diam glass tubing bent 90° at the point of emergence from the upper side of the Teflon support.

Fig. 4.
Measurement station.

D. Sample Transport and Electrolysis Cells

The turntable (Fig. 5) is rotated by a Geneva-drive, intermittent-motion assembly that provides slow acceleration and accurate positioning. A cylindrical rod driven by a pneumatic cylinder that engages indentations

Fig. 5.
Sample transport turntable.

milled in the turntable rim at each of the 24 positions provides a table locking mechanism and ensures position accuracy. As part of an interlock system, a switch on the locking mechanism monitors its status. The analyst reads in the position number (1-24 on the turntable) of the first cell in a series, and the calculator prints and plots the position number associated with each analysis result.

The turntable and pneumatic-hydraulic cell-raising mechanism (Fig. 6) provide accurate positioning of a cell

Fig. 6.
Cell-raising mechanism.

relative to the close-fitting working electrode, as well as a tight seal with the Teflon support. The close-fitting holes in the upper portion of the turntable guide cells into position. A Teflon sheet on the lifting disk provides a low-friction surface that prevents wedging of cells during the raising operation. To lessen force on the cells, the hydraulic cylinder of the cell-raising mechanism is driven with a reduced pressure of 1.4 atm by a miniature pressure regulator. The slow speed required for correct operation is set with a needle valve. A low-torque, whisker-actuated switch at the measurement station informs the calculator that a cell is correctly in position. A small scanner assembly, consisting of a light and phototransistor mounted in the same housing, detects the last cell, and instrument operation stops.

The electrolysis cells are simple to fabricate from glass tubing of 4.8-cm diam and 0.2-cm wall thickness. The total cell height is 4.5 cm with ± 0.05 -cm tolerance to prevent damage to the working electrode and to attain effective aspiration of solution. The flat cell bottom avoids interference with the stirrer paddle and attains effective aspiration. The cell top is ground flat and fire-polished lightly to produce a gas-tight seal with the Teflon support.

E. Reagent Dispensing System

This system (Fig. 7) is patterned on those developed for the automated spectrophotometer. These systems have provided years of reliable service and maintained high precision while handling corrosive reagents. In the automated coulometer system, only glass and Teflon contact the corrosive reagents. The system combines pneumatic and hydraulic transducers that provide a smooth, reproducible motion to gas-tight syringes. Delivery volume is set by adjustable stops on the hydraulic cylinder. Precision, determined by weighing dispensed reagents, is better than 0.01% RSD for 23-mL volumes of the 5.5M HCl-0.015M sulfamic acid electrolyte and 5-mL volumes of the 5M phosphate solution. Construction is detailed in Appendix B and preoperational adjustment is given in Appendix A.

F. Cell, Stirrer, and Electrode Cleaning System

All cell components must be cleaned thoroughly between samples. The large surface-area working electrode is difficult to clean without large volumes of rinse. Of

Fig. 7.
Reagent-dispensing system.

many approaches tested, the most effective is to place the tip of an aspiration tube at an exact position at the junction of the working electrode and cell bottom. Exact positioning of the Teflon aspirator tube is attained by moving it pneumatically through a Teflon guide tube (Fig. 8). Aspiration is provided by a 1.8-cfm-diaphragm air pump operating at -0.6 atm. The percentage of solution removed using an initial aspiration and two rinse-aspiration combinations is >99.98 .

A Teflon nutating-disk pump delivers the rinse solution. A concurrently activated two-way valve in the output line prevents solution flowing back to the pump. The pump voltage controls the delivery rate, and the elapsed time of operation controls the volume.

Fig. 8.
Cell and electrode cleaning system.

The ~ 80 -mL waste volume per analyzed sample flows through a Teflon tube encased in a larger Teflon tube to a bottle mounted outside the box. The bottles are easily changed.

The cleaning sequence is (1) insert aspirator tube and remove solution, (2) add ~ 4 -mL of rinse solution while aspirating, (3) withdraw aspirator tube, (4) add rinse solution to the height of the maximum sample volume, (5) stir, (6) aspirate, (7) withdraw aspirator tube, (8) add a slightly greater volume of rinse solution, (9) stir, (10) aspirate, and (11) withdraw aspirator tube. A detailed description is given as subroutine "rinse" in Fig. E-1.

IV. INSTRUMENT PRECISION

Measurement precision was established by processing weight aliquots, containing ~ 5 mg of plutonium each, of a reference material solution. Eight aliquots per day were processed over a 2-week period. The average within-day RSD for a single determination was 0.082%. Long-term precision, computed using all data, was 0.096% RSD.

ACKNOWLEDGMENTS

Design of the automated controlled-potential coulometer entailed the efforts of many Los Alamos National Laboratory personnel. In particular, credit is

due A. Criscuolo for designing the potentiostat modification, P. Osvath for mechanical drawings, and R. Plehn

for electrical drawings. J. E. Rein provided general guidance and preediting assistance.

APPENDIX A

PREOPERATIONAL INSTRUMENT ADJUSTMENTS AND TESTS

Preoperational adjustments and tests are described for the reagent dispensing and cleaning systems, alignment of the working electrode, and overall operational reliability. The adjustments are made most conveniently by manual control using the calculator special-function keys and the program given in Table E-I.

I. REQUIRED SERVICES

Electrical service requirements are 120 V, 15 A, and 60 Hz. A 2.7-atm inert-gas supply of nitrogen or argon, containing <400 ppm oxygen, is required.

II. REAGENT DISPENSING AND CLEANING SYSTEMS

The dispensed volumes of the electrolyte and phosphate solutions are adjusted by mechanical stop positions on the hydraulic cylinder mechanisms. Just enough electrolyte is added to cover the platinum working electrode. The phosphate solution is dispensed to provide a 1M phosphate concentration in the cell solution, calculated disregarding sample volume. Volumes of rinse solution are controlled by programming the elapsed time of pump operation.

To avoid splashing, the delivery tips dispense all solutions onto the bridge tube of the counter electrode. The tips are cut at 45° and bent slightly toward the bridge tube.

Premature delivery of the phosphate solution is prevented by separating the three dispenser tips by ~1 mm. The electrolyte tip is lowest, the phosphate tip next, and the rinse tip on top, so that any adhering electrolyte and phosphate solutions are removed during the rinse operation.

Delivery rates of the electrolyte and phosphate solution dispensers are adjusted by needle valves in the

hydraulic system to provide smooth motions of the syringe plungers and steady-rate deliveries without splashing. The delivery rate of the rinse pump is adjusted to ~2 mL/s.

III. WORKING ELECTRODE ALIGNMENT

Careful positioning of the working electrode is required for unhindered operation of the cell-raising system and for efficient aspiration. The working electrode bottom must be flat and parallel to the turntable surface. The electrode is positioned centrally in the turntable holes at a height so its bottom is 4.3 cm below the Teflon support. System alignment is attained using a plastic jig counterbored slightly larger than the outer diameter of the working electrode. The jig, positioned on the turntable at the measurement station, is raised manually, and the working electrode wires are adjusted so the electrode is centered in the jig counterbore. The tip of the aspirator tube is positioned at the junction of the working electrode and the cell bottom, so that solution is removed effectively from the working electrode and cell by capillary action.

IV. OVERALL INSTRUMENT RELIABILITY

Overall instrument reliability is evaluated by processing weight aliquots containing ~5 mg of plutonium each, taken from a homogeneous solution. The precision data presented in Sec. IV can serve as the basis of comparison. Causes of poor reproducibility include splashing by the stirrer, faulty solution dispensings, incomplete aspiration, electrode difficulties, and electronic malfunctions. The overall electronic system is tested according to the procedure given in Appendix B.

APPENDIX B

OPERATIONAL PROCEDURE FOR PLUTONIUM DETERMINATION

I. REAGENTS

A. Electrolyte Solution, 5.5M HCl-0.015M Sulfamic Acid

Prepare 5.5M HCl by passing hydrogen chloride into distilled-deionized water, then diluting with the same water as necessary, based on titration with standardized sodium hydroxide. Prepare the electrolyte by dissolving 1.46 g of AR-grade sulfamic acid in the 5.5M HCl to a final volume of 1 liter. This solution is stable for at least 2 months.

B. Phosphate Solution, 5M NaH_2PO_4

Dissolve 690.0 g of $\text{NaH}_2\text{PO}_4 \cdot \text{H}_2\text{O}$ in distilled-deionized water to a final volume of 1 liter. Of several brands evaluated, Mallinckrodt AR-grade has proven reliable.

C. Rinse Solution, 6M HCl

Mix one volume of concentrated AR-grade HCl with one volume of distilled-deionized water.

II. ELECTRICAL TEST OF OVERALL ELECTRONIC SYSTEM

As a minimum, this test is made after a 15-min warmup at the beginning of a day's operation and again at the end. The data obtained are used to eliminate a small day-to-day variance, typically less than 0.04%, caused by slight instability of electronic components.⁴ A high-wattage, 100-ohm resistor is substituted for the cell, and a 1-V potential from the potentiostat is applied for 10 min. The calculator controls the coulometer and the real-time clock, which accurately measures the time interval. (The program flow diagram and program listing for this test are given in Fig. E-2 and Table E-II.) The chemical calibration factor is adjusted by the ratio of the coulombs measured by the coulometer relative to the coulombs calculated from the time-current measurement.

III. CHEMICAL CALIBRATION

As the reference material, NBS SRM 949 plutonium metal is used. For highest reliability, weight portions of a solution prepared with this reference material are processed concurrently with samples, and the sample results are computed relative to the reference material results. A less accurate operational mode is to use a calibration factor established at a previous time relative to NBS SRM 949 and verified within established control limits by the concurrent processing of one or two reference material portions.

IV. BRIDGE TUBE MAINTENANCE

The two bridge tubes are rinsed and refilled with electrolyte solution before starting a day's operation.

V. BLANKS

As a minimum, a blank is processed through the complete electrolysis cycle of reduction, first oxidation, and measurement oxidation at the beginning and end of a day's operation. The accumulated times and coulombs are printed at 1-min intervals for both oxidations. The program flow diagram and program listing are given in Fig. E-3 and Table E-III.

VI. SAMPLES

Samples containing ~5 mg of plutonium in a volume of 2 mL or less are delivered into cells, preferably by weight. The cells are placed on the turntable and the sample designations and turntable positions are logged. The turntable position of the first sample (or blank or reference material) is read in to the calculator. Automated operation is initiated.

The tape printout consists of (1) real time, (2) turntable position, (3) elapsed times for the reduction and two oxidations, and (4) numbers of coulombs for the

reduction and two oxidations. The number of coulombs measured for the blank at elapsed times equal to electrolysis times for the samples are subtracted from the number of coulombs for the sample. The graphic output (Fig. B-1) provides diagnostic information. The program flow diagram and program listing are given in Fig. E-1 and Table E-IV.

Fig. B 1.
Typical graph of plutonium electrolysis.

APPENDIX C

MECHANICAL COMPONENTS

Appendix C consists of reduced copies of mechanical components including construction drawings for the automated controlled potential coulometer. CAPE package number 2669, which consists of working drawings, may be purchased from the Technical Information Center, Oak Ridge, TN 37830.

FIG. 1
PUMP AND ENGINE

Fig. C 2.
Sample transport and cell.

Fig. C-3.
Measurement station.

Fig. C-4.
Reagent dispensers.

Fig. C-6.
Turntable lock.

Fig. C-9
Measurement station supports

Fig. C-10.
Services support panel.

Fig. C 11.
Sample transport components.

APPENDIX D

ELECTRICAL CIRCUITRY

Appendix D consists of reduced copies of details of electrical components including circuit diagrams for the automated controlled potential coulometer. CAPE

package number 2669, which consists of working drawings, may be purchased from the Technical Information Center, Oak Ridge, TN 37830.

Fig. D 2.
Wiring diagram.

Fig. D-3.
Interconnection chassis.

APPENDIX E
SYSTEM SOFTWARE

Appendix E consists of system software for the automated controlled-potential coulometer.

Fig. E-1. (cont)

Fig. E-2.
Electrical test program flow diagram.

Fig. E-3.
Blank program flow diagram.

TABLE E.1

SPECIAL FUNCTION KEYS LISTING

```

f0: *wrt 709,"1";"Clear"
f1: *wrt 709,"10";wait 15000;wrt 709,"1";"Disp electrolyte"
f2: *wrt 709,"11";wait 10000;wrt 709,"1";"Disp phosphate"
f3: *wrt 709,"19";wait 16000;wrt 709,"1";"Rinse solution"
f4: *wrt 709,"13";"Stir"
f5: *wrt 709,"14";wait 30000;wait 20000;wrt 709,"1";"Aspirate"
f6: *wrt 709,"1,12";"Lower cell"
f7: *wrt 709,"1218";"Unlock table"
f8: *wrt 709,"121817";wait 4000;wrt 709,"1,1218";wait 7000;wrt 709,"1,12"
f9: *wrt 709,"16";wait 20;wrt 709,"1";"Reset coulometer"
f10: *wrt 709,"15";"Ext cell"

```

TABLE E.2

ELECTRICAL TEST PROGRAM LISTING

```

*0001 011: *IF5"190H0T2T0";wrt 709,"001";dim A$(10)
1: 1999-1199.48-#16000 A-#wrt 709,"1999"
2: ddr "Electrical Test, 10000";wait 3000
3: ddr "Set to remote" Resistor 1000;str
4: wrt 709,"15";drr "START"0;str
5: wrt 9,"1F";red 9;A#start A#
6: wrt 709,"16";wrt 709,"1,15";red 3;A;Ctit A#0;str +0
7: wrt 9,"1B";str 9;dim 9;"INT"
8: wrt 9,"010600000 UIC"
9: if +1;1;fdr "CAL"
10: wrt 709,"03";frr 722;frr 722;0
11: if 000;Y;150;fdr "OVERLOAD"
12: end -5
13: "INT";red 3;A;C
14: str 1;frr 9;str
15: "CAL";fdr 3;frr "Coulometer";A
16: fdr 5;frr "Roxion C-A";A B;fdr
17: wrt 709,"1";drr "Test complete";str
18: "OVERLOAD";wrt 709,"1";fdr 2;frr "TENTIOSTAT";A "OVERLOAD";fdr 2
19: beerr;wait 200;0+1-0;frr 0 10
20: end
*6001

```

TABLE E-III

BLANK PROGRAM LISTING

```

0: dim A$(5,14):rem 7:wrn 722,"F1F3M3A0H0T3T3":wrn 709,"001"
1: dsn "CPC-Plot, Blank 10wA":wait 5000:dsn "Set to Remote":istr
2: ent "Position No. of First Cell?":P
3: wrn 709,"05":wrn 722,"T3":fnt :red 722,G
4: if G<2:eto +2
5: spc 2:prt " Gas supply", " is off":spc 2:istr :eto +1
6: wrn 709,"14":wait 30000:wait 20000:wrn 709,"1"
7: "GAS":wrn 709,"05":wrn 722,"T3":fnt :red 722,G:cts 1:0+X+C+E
8: if G<2:eto +2
9: spc 2:prt " Gas supply", " is off":spc 2:istr :eto +2
10: wrn 709,"06":wrn 722,"T3":fnt :red 722,L
11: if L>2:eto +2
12: wrn 709,"19":wait 16000:wrn 709,"1":eto "END"
13: wrn 709,"12":wait 10000
14: wrn 709,"03":wrn 722,"T3":fnt :red 722,D
15: if D<2:beep:spc 2:prt " CELL DID", " NOT LOWER":spc 2:istr :eto +1
16: wrn 709,"18":wait 1000
17: wrn 709,"04":wrn 722,"T3":fnt :red 722,T
18: if T>2:beep:spc 2:prt " TABLE DID", " NOT UNLOCK":spc 2:istr :eto +1
19: wrn 709,"17":wait 4000:wrn 709,"1,12,18":wait 8000
20: wrn 709,"1,12":wait 1000
21: wrn 709,"04":wrn 722,"T3":fnt :red 722,T
22: if T<2:beep:beep:spc 2:prt "TABLE NOT LOCKED":spc 2:istr :eto +2
23: wrn 709,"1":wait 10000
24: wrn 709,"03":wrn 722,"T3":fnt :red 722,D
25: if D<2:beep:beep:spc 2:prt "CELL NOT RAISED":spc 2:istr :eto +2
26: wrn 709,"10":wait 14000:wrn 709,"13"
27: wrn 707,"in:rate 400,1500,7400,9000"
28: red 707:sc1 0,2200,-4.5,4.5
29: fxd 0:ixox 0,100:iyax 0,5,-4,4,2
30: rlt 1000,-.35,1:csiz 1.4:lbl "TIME, min"
31: rlt 0,1.2,1:rcalt -3:0
32: csiz 1,1.5,1.5,90:lbl "LOG CURRENT"
33: rlt 400,5,1:csiz 3
34: lbl " BLANK NUMBER ":P:ren
35: for A=1 to 10
36: rlt 0,log(A),-2:rlt 20,log(A),-1
37: next A
38: wrn 709,"1,13"
39: wrn 709,"1250"
40: wrn 709,"16":wait 20:wrn 709,"1,13":wait 20
41: red 3:A,Z:if A#0:eto +1
42: wrn 709,"15"
43: wrn 709,"B":eir 9:ion 9,"INT 1"
44: wrn 9,"UID600000 UID"
45: 1+T:esb "TIME"
46: if fls2=0:spc 2:prt A$(1):spc 2
47: fxd 0:prt "Blank No.":P:P+1+P:if P.24:1+P
48: wrn 709,"00":wrn 722,"R3T3":fnt :red 722,Y
49: if abs(Y)<=.0001:ren:eto +6
50: wrn 709,"02":wrn 722,"R5T3":fnt :red 722,Z
51: if fls1:cts 1:pen:eto +4
52: if abs(Z)>50:eto "OVERLOAD"
53: if Y#0:rlt X,-log(10000*abs(Y)), -2:fld 0:dsn 10000:
54: .315+X+X:eto -6
55: fnt :red 3,r3,A:0:X:iflt 3:prt "Reduction Coul",r3:spc

```


TABLE E-III (CONT)

```

56: 2+T:asb "TIME"
57: wrt 709,"16"iwait 20iwr 709,"1.1315"iwait 20
58: red 3,A,Z:if A#0:sto -1
59: wait 400iwr 9,"B"feir 9:oni 9,"INT 2"
60: wrt 9,"UID60000/UG"
61: wait 200iwr 706,"1570"iwait 200:ert "Oxidation No.1","Min      Coulombs"
62: if f1:icf9 1iwr 9,"UID60000 UIG"
63: wrt 709,20iwr 722,"R3T3"ifmt :red 722,Y
64: if C=2iwr 9,"B"feir:sto +5
65: wrt 709,"02"iwr 722,"R5T3"ifmt :red 722,Z
66: if abs(Z)-50:sto "OVERLOAD"
67: if Y#0:alt X:los 10000abs(Y),-2:if d 0:dsz 10000Y
68: .315+X+X:sto -6
69: 3+T:asb "TIME"
70: wrt 709,"16"iwait 20iwr 709,"1.1315"iwait 20
71: red 3,A,Z:if A#0:sto -1
72: wrt 709,"11"
73: wrt 9,"B"feir 9:oni 9,"INT 3"
74: wrt 9,"UID60000 UIG"
75: wait 10000iwr 709,"1.111315"iwait 200
76: wrt 706,"1680"i:sto :ert "Oxidation No. 2","Min      Coulombs"
77: if +1:icf9 3iwr 9,"UID60000 UIG"
78: wrt 709,"00"iwr 722,"R3T3"ifmt :red 722,Y
79: if B=20iwr 9,"B"feir:sto +5
80: wrt 709,"02"iwr 722,"R5T3"ifmt :red 722,Z
81: if abs(Z)-50:sto "OVERLOAD"
82: if Y#0:alt X:los 10000abs(Y),-2:if d 0:dsz 10000Y
83: .315+X+X:sto -6
84: 4+T:asb "TIME"
85: wrt 709,"1"iwait 5000
86: wrt 709,"14"
87: :rd 3:ert r7-r6:r6-r7:r9-r8:rd 2
88: :lt 200,4,1:csiz 1.4:1b1 A#(1)
89: :rd 3:lt 400,3,8,1:1b1 r7-r6
90: :lt 400,3,6,1:1b1 r8-r7
91: :lt 400,3,4,1:1b1 r9-r8
92: :enfiwait 30000
93: "PINSE"iwr 709,"13"iwait 2000iwr 709,"1,14"iwait 10000
94: wrt 709,"1,19"iwait 15000iwr 709,"1,13"iwait 15000
95: wrt 709,"1"iwait 5000
96: wrt 709,"14"iwait 30000iwait 20000
97: wrt 709,"1,19"iwait 16000iwr 709,"1,13"iwait 15000
98: wrt 709,"1"iwait 5000
99: wrt 709,"14"iwait 30000iwait 20000iwr 709,"1"
100: :s 2:sto "GAS"
101: "TIME"iwr 9,"R"i:red 9,A#(T)
102: 60val:A#(T,7))+val:A#(T,10))+val:A#(T,13): 60-r:T+5:
103: ret
104: "INT 1":sfa 1feir 9:iret
105: "INT 2":fnt :red 3,A,Z:1+C+C
106: fnt +2.0:re14.3iwr 16,C+A
107: :sfa 1feir 9:iret
108: "INT 3":fnt :red 3,A,Z:1+B+B
109: fnt +2.0:re14.3iwr 16,B+A
110: :sfa 3feir 9:iret
111: "OVERLOAD"iwr 709,"1"i:sto 2:ert " POTENTIALSTAT"," OVERLOAD"i:sto 2
112: beepiwait 2000+1+0:imp 0.10
113: "END":end
+28856

```

TABLE E-IV

SAMPLE PROGRAM LISTING

```

0: dim A$(5,14);rem 7;wrt 722,"FIR3M2A0H0T3T3";wrt 709,"00"
1: dse "CFC-Plot, Sample 100mA";wait 5000;dse "Set to Remote" ;wrt
2: ent "Position No. of First Cell?";P
3: wrt 709,"05";wrt 722,"T3";ired 722,G
4: if G<2;goto +2
5: sdc 2;irt " Gas supply," is off";sdc 2;sta irta -2
6: wrt 709,"14";wait 30000;wait 20000;wrt 709,"1"
7: "GR3";wrt 709,"05";wrt 722,"T3";ired 722,G;cta 1;0+
8: if G<2;goto +2
9: sdc 2;irt " Gas supply," is off";sdc 2;sta irta -2
10: wrt 709,"05";wrt 722,"T3";ired 722,L
11: if L<2;goto +2
12: wrt 709,"19";wait 16000;wrt 709,"1";irta END
13: wrt 709,"12";wait 10000
14: wrt 709,"03";wrt 722,"T3";ired 722,D
15: if D<2;beep;sdc 2;irt " CELL DID," NOT LOWER ;sdc 2;sta irta -2
16: wrt 709,"10";wait 1000
17: wrt 709,"04";wrt 722,"T3";ired 722,T
18: if T<2;beep;sdc 2;irt " TAELE DID," NOT UNLOCK ;sdc 2;sta irta -2
19: wrt 709,"17";wait 4000;wrt 709,"1,1218";wait 8000
20: wrt 709,"1,12";wait 1000
21: wrt 709,"04";wrt 722,"T3";ired 722,T
22: if T<2;beep;beep;sdc 2;irt "TAELE NOT LOCKED";sdc 2;sta irta -2
23: wrt 709,"1";wait 10000
24: wrt 709,"03";wrt 722,"T3";ired 722,D
25: if D<2;beep;beep;sdc 2;irt "CELL NOT RAISED ;sdc 2;sta irta -2
26: wrt 709,"10";wait 14000;wrt 709,"13"
27: wrt 707,"1";scl 400,1500,7400,9000"
28: rsc 707;scl 0,2200,-4.5,4.5
29: fcd 0;ico 0,100;ico 0,1.5,-4,4,3
30: plt 1000,-.35,1;icsiz 1.4;lbl "TIME, min"
31: plt 0,1,2,1;icplt -3,0
32: csiz 1,1,5,1,5,90;lbl "LOG CURRENT"
33: plt 400,5,1;icsiz 3
34: lbl "SAMPLE NUMBER ";P;ken
35: for A=1 to 10
36: plt 0,log(A),-2;plt 20,log(A),-1
37: next A
38: wrt 709,"1,13"
39: wrt 706,"1250"
40: wrt 709,"16";wait 20;wrt 709,"1,13";wait 20
41: red 3,A,2;if A#0;goto -1
42: wrt 709,"15"
43: wrt 709,"B";feir 9;oni 9,"INT 1"
44: wrt 9,"UID1000000/UID"
45: 1+T;esb "TIME"
46: if f;g;goto "RINSE"
47: wrt 709,"00";wrt 722,"R3T3";ired 722,Y
48: if abs(Y)<=.0005;pen;goto +5
49: wrt 709,"02";wrt 722,"R5T3";ired 722,Z
50: if abs(Z)>50;goto "OVERLOAD"
51: if Y#0;plt X,-log(10000*abs(Y)), -2;if d 0;dse 100000Y
52: .315*X+X;goto -6
53: red 3,r3,A;0+X
54: 2+T;esb "TIME"
55: wrt 709,"16";wait 20;wrt 709,"1,1315";wait 20
56: red 3,A,2;if A#0;goto -1
57: wait 400;wrt 9,"B";feir 9;oni 9,"INT 2"
58: wrt 9,"UID900000/UID"
59: wrt 706,"1570";wait 200
60: if f;g;goto "RINSE"

```

TABLE E IV (CONT)

[illegible]

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