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Lawrence Livermore Laboratory

PLUTONIUM SAMPLE EXCHANGE MEETING - Minutes

JOHN C. WALDEN, RONALD J. DUPZYK, AND RILEY D. JARVER

NOVEMBER 17-19, 1976

LAWRENCE LIVERMORE LABORATORY, LIVERMORE, CA 94550

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Nov. 1976

PLUTONIUM SAMPLE EXCHANGE MEETING

Minutes

November 17-19, 1976

Lawrence Livermore Laboratory
Livermore, California
94550

Authors: John C. Walden, Ronald J. Dupzyk
and Riley D. Carver

PLUTONIUM ISOTOPIC STANDARD SAMPLE EXCHANGE MEETING

November 17, 1976

Lawrence Livermore Laboratory
Livermore, California 94550

The chairman, Doug Sellars, opened the meeting. Walter Nervik of LLL gave a welcoming address. Riley Carver was assigned to keep records of the meeting. Attendance was taken by sign up sheet, and everyone introduced himself to the group. (Fig. 1)

Herman Anderson proposed the 1977 exchange sample frequency for each different type of sample. The metal samples are in short supply, but lots of oxide samples are available. (Figs. 2-3)

Liz Baker discussed statistical inferences from the reports. Figure 4 shows the levels and sources of variation. Figure 5, the table of means, shows that small numbers usually have large uncertainties.

Guy Merrill proposed that McClellan, LASL, and LLL run NBS-948 minor isotopes to good precision. Results would be sent to RFP for statistical analysis. McClellan will instigate the effort.

Bob Foster reported three years' results on NBS-948. (Figs. 6-9) The ratio 238/239 is a small number, and some laboratories do not report it at all. The ratio 240/239 was good within laboratories, but there was some spread between laboratories. The other isotope ratios showed good agreement. Some laboratories use this material as a calibration standard.

John Cappis reported on the UK-131 exchange. (Figs. 10-12) When the measurements are viewed as fractional recovery of the two year average value, LLL shows a 0.5% bias on the low side of the 240/239 ratio. The 241/239 ratio shows wide variations within laboratories, implying chemical separation problems. The 242/239 measurements were satisfactory.

Tom Bement reviewed the continuing study of the ²⁴¹Pu half life. (Figs. 13-30) Pooled average from all labs for the UK-131 study is 14.74 years. The value from the NBS-946 study is 14.33 years.

Henry Carden reported on the "R" nitrate exchange. (Figs. 31-34) Mound Laboratory had some variation between exchanges on the 240/239 and was high on all 241/239 measurements.

Wes Efurd discussed the NBS-946 exchange. The laboratories, whose measurements were historically high, still are. The laboratories, whose measurements were historically low, still are. LLL is biased 0.5% low on 240/239.

Evain Ruby reported on the ²³⁸Pu exchange. (Figs. 35-37) All measurements are in agreement, but not as good as four years ago.

PLUTONIUM ISOTOPIC STANDARD SAMPLE EXCHANGE MEETING

Dick Perrin described the LASL mass spectrometer and operating system. Calibration showed a 0.000722 effect for each mass unit. Applying this same correction to measurements from all laboratories reduced most biases to within 0.1% of the overall average. LLL continued to be biased low about 0.5%. (Figs. 38-48).

Bob Foster described the SRP system for measuring plutonium isotopes with gamma spectroscopy. The unit was manufactured by LLL and is used for liquid samples. ^{242}Pu is not measured. Every tenth sample is checked by mass spectroscopy.

Evan Ruby discussed the interfacing of computers with mass spectrometers at RFP. Three systems are now connected: LIS (laboratory information system), NMC (Nuclear materials control), and PDAT (production data analysis and traceability). Three different computers are used. Ultimately, they hope to use microprocessors with paper tape emulators on each instrument.

Ray Newbury presented a paper, "Estimation of Ionization Potentials of Heavy Elements Using a Pulse-Counting Mass Spectrometer. (Appendix I)

There was a discussion about the impact of SAFEGUARDS regulations. All controlled materials inside of laboratories must be accounted for. Each shipment or inventory requires a nondestructive assay. If the current regulations are enforced, there will have to be increases in manpower and expenditures.

The 1977 meeting will be at LASL in the autumn.

Wednesday, November 17, 1976

DELEGATION:

Mound Laboratory

Carden, Henry S.
Goss, Ronald L.
Sellers, Douglas E.

Atlantic Richfield Hanford Co.

Colvin, Curtis A.
Louk, Wayne L.

Rocky Flats Laboratory

Anderson, Herman C.
Baker, Elizabeth L.
Ferris, Yvonne M.
Ruby, Evain D.

Los Alamos Scientific Laboratory

Bement, Thomas R.
Cappis, John H.
Perrin, Richard E.
Zeigler, R. Keith

New Brunswick Laboratory

Pietri, Charles E.

Savannah River Plant

Evans, Thomas C.
Foster, Robert W.

McClellan AFB

Efurd, Deward W.
Merrill, Guy

Lawrence Livermore Laboratory

Carver, Riley D.
Dupzyk, Ronald Jene
Friesen, Richard D.
Magana, Joseph
Mendoza, Benjamin
Newbury, Ray S.
Prindle, Austin L.
Stone, Richard E.

TID L-9 (3)
TIC Oak Ridge, Tn (2)

12/76 NBS 946 (NO_3)
 1/77 238 OXIDE
 2/77 Stream R (NO_3)
 3/77 UK 131 OXIDE
 4/77 NBS 946 (NO_3)
 5/77 Stream R (NO_3)
 6/77 NBS 948 (NO_3)
 7/77 UK 131 OXIDE
 8/77 238 OXIDE
 9/77 NBS 946 (NO_3)
 10/77 STREAM R (NO_3)
 11/77 UK 131 OXIDE
 12/77 NBS 946 (NO_3)

Sample Type	Samples out
NBS 946 (NO_3)	12/76 4/77 9/77 12/77
238 OXIDE	1/77 8/77
STREAM R (NO_3)	2/77 5/77 10/77
UK 131 OXIDE	3/77 7/77 11/77
NBS 948 (NO_3)	6/77

Figure 2

SAMPLE EXCHANGE MATERIAL

November 1976

Metal Exchange	Metal Oxide Isotopic	Grams on Hand	Grams Used/Year	Exchanges Available	Years Supply
S	- -	702 g	560 g	5	1-1/4
U	- -	663 g	560 g	4	1
	R	111 g	1 g		100
	UK-131	6 g	165 mg		5-1/2
	238 OX	60 mg	1.2 mg		50

Figure 3

Standard Deviation x100
Mean

SRP-Oxide

	<u>Scan</u>	<u>Loading</u>	<u>Labs</u>	<u>Mean</u>
239/238	.4%	.3%	.6%	.134
240/238	1.	.9	2.	.015
242/238	.5	.2	.5	.121

METAL R

238/239	6.	8.	41.	.0002
240/239*	.4	.1	.3	.062
241/239	.7	1.	2.	.004
242/239	5.	2.	7.	.0002

UK-131

238/239	3.	1.	5.	.001
240/239	.2	.1	.2	.217
241/239	.5	.4	2.	.025
242/239	1.	.4	.4	.0056

NBS 946

238/239	1.	.6	3.	.0028
240/239	.3	.1	.2	.145
241/239*	.4	.001	.3	.039
242/239	.8	.6	.7	.0067

NBS 948

238/239**	7.	9.	21.	.00014
240/239	.3	.1	.3	.086
241/239	.7	.4	.8	.0046
242/239	4.	2.	4.	.0003

Factor x100

- * F-ratio between filament loadings not statistically significant
 ** F-ratio between filament loadings significant at .05 level

Figure 5

NBS 948 NITRATE RATIO 238/239

Figure 6

NBS 948 NITRATE RATIO 240/239

Figure 7

NBS 948 NITRATE RATIO 241/239

NBS 948 NITRATE RATIO 242/239

Figure 10

Figure 11

Figure 12

TABLE I

241 half-life, UK-131

<u>Lab.</u>	<u>1969</u>	<u>1970</u>	<u>1971</u>	<u>1972</u>	<u>1973</u>	<u>1974</u>	<u>1975</u>	<u>1976</u>	<u>Std. Dev</u>
ARHC	15.61	15.53	15.35	15.02	14.79	14.83	14.82	14.84	.07
SRP	15.63	15.43	15.06	15.06	14.86	14.74	14.49	14.52	.05
RFD	16.02	16.06	15.06	14.57	14.57	14.65	14.80	14.74	.09
LLL	15.16	14.64	15.21	14.89	15.14	15.04	14.94	14.93	.11
LASL	12.59	14.62	14.57	14.50	14.51	14.49	14.16	14.12	.07
ML	11.82	14.05	14.74	15.00	14.88	15.18	15.12	15.24	.06
POOLED					14.80	14.83	14.81	14.74*	.04

241 half-life, NBS-546

<u>Lab.</u>	<u>1975</u>	<u>1976</u>	<u>Std. Dev</u>
ARHC		14.29	.11
SRP		14.13	.10
RFD		14.51	.12
LLL		14.41	.05
LASL		13.73	.08
ML		13.86	.18
USAF		14.97	.17
POOLED	14.31	14.33**	.08

*110 months of data, 177 points

**12 months of data, 72 points

Figure 13

14.7(NBS)

UK-131,241/239,ROBUST

POOLED VALUES

TIME IN MONTHS

10/20/78 10.00.00

Los Alamos
Scientific Laboratory
of the University of California

Figure 14

UK-131,241/239,ROBUST

ARHC

TIME IN MONTHS

01/04/76 17.03.26

Los Alamos
Scientific Laboratory
of the University of California
48
The University of California, Los Angeles, California

Figure 15

UK-131,241/239,ROBUST

TIME IN MONTHS

11/26/76 17.05.00

Los Alamos
Scientific Laboratory
of the University of California

Figure 16

UK-131,241/239,ROBUST

RFD

TIME IN MONTHS

11/08/76 17.05.22

Los Alamos
Scientific Laboratory
of the University of California
An Office of the National Security Agency

Figure 17

UK-131,241/239,ROBUST

TIME IN MONTHS

25/06/78 17.09.78

Los Alamos
Scientific Laboratory
of the University of California
Los Alamos, New Mexico 87545

Figure 18

UK-131,241/239,ROBUST

LASL

TIME IN MONTHS

11/06/78 17.03.28

Figure 19

UK-131,241/239,ROBUST

TIME IN MONTHS

11/06/78 17.09.78

Figure 20

UK-131,241/239,ROBUST

MEAN

TIME IN MONTHS

11/08/76 17.09.29

Figure 21

946,241/239,0676.ROBUST,W/O OUTLIERS

FOOLED VALUES

TIME IN MONTHS

10/29/78 11:46:00

Figure 22

946,241/239,0676,ROBUST,WITH OUTLIERS

POOLED VALUES

TIME IN MONTHS

11/11/78 08:29:08

946,241/239,ROBUST

SRP

TIME IN MONTHS

88/08/78 18.00.48

Los Alamos
Scientific Laboratory
of the University of California
An LLNL Laboratory

Figure 25

946,241/239,ROBUST

RFD

TIME IN MONTHS

98/09/78 10.06.40

946,241/239,ROBUST

LAST

TIME IN MONTHS

23/09/78 10.15. AM

Los Alamos
Scientific Laboratory
of the University of California

Figure 27

946,241/239,ROBUST

USAF

TIME IN MONTHS

05/05/78 10.00.00

Los Alamos
Scientific Laboratory
of the University of California
San Francisco, CA 94305-5080

Figure 29

946,241/239,ROBLST

TIME IN MONTHS

11/01/74 18.04.49

Los Alamos
Scientific Laboratory
of the University of California

Figure 30

R-NITRATE

RATIO 238/239

Figure 31

R-NITRATE RATIO 240/239

Figure 32

R-NITRATE RATIO 241/239

Figure 33

R-NITRATE RATIO 242/239

Figure 34

SRP - OXIDE

239/238

SRP - OXIDE 240/238

SRP - OXIDE

242/238

Figure 37

URANIUM SYSTEMS CALIBRATION

Figure 38

Test Values

UK 131	.21737
R Metal	.06251
NBS 946	.14519
NBS 948	.08642

Figure 40

	SD	MEAN
UK 131	.0376	.9998
R Metal	.0517	.9999
NBS946	.0278	.9998
NBS 948	.0416	1.0006
ALL	.0432	.9999

Figure 41

	SD	MEAN
UK 131	.184	1.0015
R Metal	.153	1.0005
NBS 946	.173	1.0005
NBS 948	.0458	.9993
ALL	.162	1.0006

Figure 42

	SD	MEAN
UK 131	.226	1.0000
R Metal	.226	.9980
NBS946	.196	.9979
NBS 948	.274	.9970
ALL	.227	.9983

Figure 43

	SD	MEAN
UK 131	.141	1.0017
R Metal	.146	1.0008
NBS 946	.101	1.0004
NBS 948	.044	1.0005
ALL	.127	1.0008

Figure 44

	SD	MEAN
UK 131	.176	.9994
R Metal	.724	.9984
NBS946	.037	.9998
NBS 948	.132	1.0037
ALL	.441	.9993

Figure 45

	SD	MEAN
UK 131	-	-
R Metal	.121	1.0002
NBS 946	.177	.9996
NBS 948	.262	1.0002
ALL	.187	1.0004

Figure 46

	SD	MEAN
UK 131	.105	.9936
R Metal	.136	.9944
NBS946	.148	.9953
NBS 948	.015	.9973
ALL	.176	.9949

Figure 47

LAB	ARHC	SRP	RFD	LASL	ML	USAF	LLL
Rel SD	.162	.227	.127	.043	.441	.187	.176
TΣ 95%	.277	.388	.217	.074	.754	.320	.301
Mean	1.0006	.9983	1.0008	.9999	.9993	1.0004	.9949
Bias	+.06	-.17	+.08	-.01	-.07	+.04	-.51

Figure 48

ESTIMATION OF IONIZATION POTENTIALS OF HEAVY ELEMENTS USING A PULSE-COUNTING MASS SPECTROMETER

The Saha-Langmuir equation

The classical application of the S-L equation

A new way to apply the S-L equation

Results

SAHA-LANGMUIR EQUATION

$$\frac{N^+}{N^0} = \frac{g^+}{g^0} \exp \left[\frac{\phi - I}{kT} \right]$$

N^+ $\equiv$ the number of positive ions leaving the hot metal surface per unit area per unit time.

N^0 $\equiv$ the number of neutral atoms leaving the hot metal surface per unit area per unit time.

$\frac{g^+}{g^0}$ $\equiv$ the ratio of the electronic partition functions of the ionic and atomic states.

ϕ $\equiv$ the work function of the hot surface.

I $\equiv$ the ionization potential of the element of interest.

kT $\equiv$ the temperature of the hot metal surface expressed in the same units as ϕ and I .

APPLYING THE S-L EQUATION TWICE FOR SAY U AND P_u ON A
RE IONIZING SURFACE

19

$$U^+/U^0 = \frac{g_U^+}{g_U^0} \exp \left[\frac{\phi_{Re} - I_U}{kT} \right]$$

$$P_u^+/P_u^0 = \frac{g_{P_u}^+}{g_{P_u}^0} \exp \left[\frac{\phi_{Re} - I_{P_u}}{kT} \right]$$

and dividing the first by the second we get

$$U^+/P_u^+ = \frac{U^0}{P_u^0} \frac{g_U^+/g_U^0}{g_{P_u}^+/g_{P_u}^0} \exp \left[\frac{I_{P_u} - I_U}{kT} \right]$$

(notice that ϕ is gone)

which may be plotted as follows in the slope-intercept form

So if I_U is known, I_{P_u} is readily obtainable from the slope.

IONIZING SURFACE TO BE WELL KNOWN AND
 U^0/Pu^0 TO BE CONSTANT.

$N \equiv$ the number of atoms striking the hot metal surface
 per unit area per unit time.

So at equilibrium, $N = N^0 + N^+$.

Since $N^0 \gg N^+$ (limited by the M.P. of the ionizing surface),
 $N \approx N^0$.

So a nice way to keep U^0/Pu^0 constant is to put the uranium
 and plutonium into a Knudsen cell and heat it to some
 convenient temperature. The U/Pu ratio in the molecular
 beam impinging upon the ionizing surface will be invariant
 with time as long as equilibrium is maintained in the cell.

This works and it has been done - but there is another way.

LET'S EXAMINE THE S-L EQUATION AGAIN FOR URANIUM AND PLUTONIUM, FOR EXAMPLE:

$$U^+/Pu^+ = \frac{U^0 g_U^+/g_U^0}{Pu^0 g_{Pu}^+/g_{Pu}^0} \exp \left[\frac{I_{Pu} - I_U}{kT} \right]$$

Substituting U/Pu for U^0/Pu^0 and looking at time increments

$$U_1^+/Pu_1^+ \approx \frac{U_1 g_U^+/g_U^0}{Pu_1 g_{Pu}^+/g_{Pu}^0} \exp \left[\frac{I_{Pu} - I_U}{kT} \right]$$

$$U_2^+/Pu_2^+ \approx \frac{U_2 g_U^+/g_U^0}{Pu_2 g_{Pu}^+/g_{Pu}^0} \exp \left[\frac{I_{Pu} - I_U}{kT} \right]$$

$$U_n^+/Pu_n^+ \approx \frac{U_n g_U^+/g_U^0}{Pu_n g_{Pu}^+/g_{Pu}^0} \exp \left[\frac{I_{Pu} - I_U}{kT} \right]$$

$$\frac{\sum U^+}{\sum Pu^+} \approx \frac{U}{Pu} \frac{g_U^+/g_U^0}{g_{Pu}^+/g_{Pu}^0} \exp \left[\frac{I_{Pu} - I_U}{kT} \right]$$

in which U and Pu are atoms of uranium and atoms of plutonium
 loaded onto the sample filament.

This is done at constant ionizing surface temperature (but not
 at constant sample temperature).

This is true as long as the geometry in the source region of the
 mass spectrometer remains constant.

THIS LAST EQUATION

$$\frac{I}{\sum M^+ / \sum N^+} \cong \frac{M}{N} \frac{g_m^+ / g_m^0}{g_n^+ / g_n^0} \exp \left[\frac{I_n - I_m}{kT} \right]$$

I THINK IS SIGNIFICANT.

It implies that if the ratio of their partition function ratios can be estimated, the *relative ionization potentials* of two elements can be determined simply by measuring the *overall ratio* of their ions counted from an ionizing surface at a constant temperature when the original known sample is evaporated to exhaustion from some source.

USING A TWO-FILAMENT SOURCE WITH Np, U, Pu, AND Am OXIDES
IN KNOWN AMOUNTS LOADED ONTO THE SAMPLE FILAMENT AND
UTILIZING PULSE-COUNTING TECHNIQUES, THE GENERAL
BEHAVIOR ILLUSTRATED BELOW WAS OBSERVED.

Note that the ratios vary widely during the experiment.

But we are interested in the total counts on each species only.

Assuming the ratio of the ratio of the partition functions is unity.

And the hot filament temperature was 2800K.

The ionization potentials calculated for Np, Pu, and Am assuming it is 6.08 eV for U follow.

Ionization potentials of some actinides				
	This work	AIP, 1972	S and H, 1969	Sugar, 1974
Np	6.10 ± 0.06	5.8	6.05 ± 0.06	6.19
U	6.08 (assumed)	6.08	6.08 (assumed)	6.05
Pu	5.91 ± 0.04	5.8	5.60 ± 0.06	6.06
Am	5.89 ± 0.05	6.05	—	5.99

MINUTES
PLUTONIUM METAL SAMPLE EXCHANGE MEETING
LAWRENCE LIVERMORE LABORATORY
LIVERMORE, CALIFORNIA

The annual Plutonium Metal Sample Exchange Meeting was held in the conference room of the Radiochemistry Building on November 18-19, 1976. Chairman of the meeting was D. E. Sellers, Mound Laboratory. Attendees and their laboratory affiliations were:

Rocky Flats Laboratory

Anderson, Herman C.
Baker, Elizabeth L.
Ferris, Yvonne M.
Ruby, Evain D.

New Brunswick Laboratory

Pietri, Charles E.

Los Alamos Scientific Laboratory

Cappis, John H.
Croley, Dale C.
Perrin, Richard E.
Zeigler, R. Keith

Savannah River Plant

Evans, Thomas C.
Foster, Robert W.

Atlantic Richfield Hanford Co

Colvin, Curtis A.
Louk, Wayne L.

Mound Laboratory

Carden, Henry S.
Goss, Ronald L.
Rodenburg, W. William
Sellers, Douglas E.

Lawrence Livermore Laboratory

Blood, Donna
Carver, Riley D.
Dupzyk, Isabelle A.
Dupzyk, Ronald Jene
Magana, Joseph
Meadows, Jesse W.
Mendoza, Benjamin
Newbury, Ray S.
Stone, Richard A.
Walden, John

McClellan AFB

Efurd, Deward W.
Merrill, Guy

After opening remarks by the host personnel, the Chairman started into the major item of business: the discussion of the analysis of the past year's sample exchange. Topics covered, persons presenting information and general comments are as follows:

Determination of Americium, Neptunium, and Uranium - R. W. Foster, SRP

Americium -

- H-Metal - (~800 ppm level) - results from all laboratories checked except for SRP which was 6-12% high when compared to the others.
- R-Metal - (~1000 ppm level) - results from laboratories agree in general. ARHCO and SRP each had one result that was high.
- S and U-Metals - Not enough data available as yet to make meaningful comparisons.

Neptunium -

- H and R Metals - Results from all laboratories check. (~25 ppm and <10 ppm levels respectively)
- S-Metal - (~50 ppm level) - one result from one laboratory was a little high. Other values checked.
- U-Metal - Not enough data as yet.

Uranium -

- U-Metal - (~140 ppm) - Very good agreement for all laboratories.
- R-Metal - (~200 ppm) - Good agreement for all except the Rocky Flats. Spark Source Mass Spec values are lower than chemical values.
- S-Metal - (~40 ppm) - Good agreement for all.
- U-Metal - (~600 ppm) - Not enough data.

Statistical Inferences - Elizabeth Baker, R.F.

An explanation of the statistical treatment given to the reported results was given. This was followed by a review of the informational format presented in the quarterly report of the data from Rocky Flats and the implications of the various values reported.

Plutonium Assay - Wayne Louk, ARHCO

FLATS

H-Metal - Rocky (lats) was over 100% Pu.

RF values ranged from 99.5% to 100.10%

ARHCO averaged the lowest values.

R-Metal - Laboratories agreed

S-Metal - ARHCO had one high value of 100.2%

T-Metal - Not enough data

Mound Laboratory seemed to have been biased low. Mound indicated they had noticed the low bias and had changed their procedure and it looks as though they have corrected for the bias.

There is general agreement that the laboratory to laboratory differences are somewhat greater now than in the past exchanges.

E. Baker had made a comparison of the plutonium assay values as determined by chemistry and an assay value obtained by adding together all detectable impurities and subtracting from 100%. She stated the labs had more consistent values obtained by the 100% technique than by chemical assay.

R. Foster had also looked into similar comparisons. He stated that SRP assay values as determined by 100% method for the H-metal over a period of six quarters were biased 0.06% low when compared to the average for all labs chemical assay values. However, if three chemical values were deleted (these values were over 100%) the SRP values were within 0.02%.

The similar comparison for the R-metal indicated the difference was less than 0.02%.

Carbon and Oxygen Determination - Ronald Goss, Mound

Carbon

S-Metal - No particular problems seemed evident

U-Metal - SRP may be some high and ARHCO low

T-Metal - Not enough data available

R. Foster announced that SRP had replaced the carbon determination apparatus (capillary trap-freezeout) they have used for years with a commercial LECO carbon determinator.

Oxygen

LASL and ML were the only laboratories reporting O_2 and this on only two samples for three quarters. There are no big differences in their values. Mound announced they have disassembled their oxygen apparatus and will no longer report O_2 on any plutonium material.

Trace Metal Impurities TRACE

Two presentations were given on the comparison of trace metal impurities determined by atomic absorption, emission spectroscopy, wet chemistry and spark source mass spectroscopy. Dale Croley, LASL, gave a presentation with emphasis on emission spectroscopic values followed by a similar discussion of results with emphasis on the spark source mass spectrographic values. The following is a somewhat combined summary of both presentations.

Aluminum - In general there was agreement by all laboratories and all methods. No one method nor one laboratory showed a bias.

Chromium - No major differences were evident. ARHCO may be biased low on T and U metals but not much information is available yet on those two metals.

Iron - No one laboratory or method is consistently biased. The chemical values for S-metal varied from ~80 to 220 over three quarters which may indicate bad samples or greatly increased lab-to-lab variation.

Silicon - H-metal had a wide spread of reported values 100 to 350 ppm. The same laboratories that had the highest and lowest reported values this year had the same relative position last year. On the lower silicon values for other metals the laboratories do not show a consistent bias.

Gallium -- ARHCO has a low bias on most metals.

Copper - Level is too low to make a valid comparison except for U metal possibility. There is some spread on the reported values on the copper in the U-metal.

Manganese - Level of manganese is too low to compare on the H, R, and S metals. No laboratory was consistently high or low on the other metals.

Nickel - Good agreement for all method and laboratories except for T metal. The level of nickel at 2000 ppm is beyond the range of most trace methods and no comparisons of any element in this range should be made.

Calorimetry - W. Rodenburg, Mound

The sample sizes being studied for calorimetry are 600 to 1000 grams. When the average values from each laboratory are compared, the standard deviation is 0.11%. Rodenburg considered this to be excellent agreement. However, if the individual laboratories values are compared to similar values from other laboratories, a statistical difference is evident. This would reflect as a shipper-receiver difference.

Mound has been routinely assaying the exchange metals by calorimetry. The R-metal has been measured 22 times with a standard deviation of 0.17%. Similar measurements on the H-metal were 14 replicates and a standard deviation of 0.07%. There is a small but statistical difference between wet chemical assay and calorimetry.

Average results from the past year from the participating laboratories were presented.

Rocky Flats	87.57 % Pu
ARHCO	87.51
Mound	87.52
LLL	87.49 *
Average	87.52 ± 0.04 %

*Used isotopic content obtained by averaging values from other three laboratories.

Rodenburg then stated that he thought the goals of the program had been obtained and recommended the program be terminated. He listed the following reasons to substantiate his recommendation.

1. Heat standards are available that are traceable to the National Bureau of Standards.
2. Isotopic standards are available that are traceable to the National Bureau of Standards.
3. Better methods for calorimeter measurement control are available.
4. No shipper-receiver control by calorimetry in use.
5. Present program does not address bias.
6. An isotopic exchange program exists.
7. Sufficient data has been obtained to demonstrate the capabilities at the present participants.

Y. Ferris, RF, recommends continuing the program because of increasing pressure by ERDA to improve and develop capabilities, especially on shipper-receiver problems. After considerable discussion it was evident that a large majority of the laboratories were in favor of continuing the program. As a consequence of the discussions, two decisions were made. Mound Laboratory, or more specifically, Rodenburg will devise a more meaningful program with additional goals and send an outline to the participants for additional suggestions and/or changes. The program will continue unchanged until a new program from Rodenburg is put into effect.

Miscellaneous

Several subjects were discussed at various times during the presentation of the exchange data. These items are brought together and placed into one general category.

R. Foster, SRP, mentioned the problems they have been having on the determination of trace levels of uranium by a fluorescence procedure. The major problem has been the variable quality of the NaF. ARHCO, LASL, and LLL indicated they have been having the same problem. Most laboratories are now testing several manufacturer's lots of NaF and when they find a good one, buy up all of that lot they can. The variable iron content of reagent grade hydrochloric acid may also be contributing to the problem. C. Colvin, ARHCO, stated they were forced to go to isotope dilution mass spectrometric determination of uranium because of personnel and reagent problems.

H. Anderson (RFP) gave a brief summary of the preparation of the S, T, and U metals that were used in the exchange.

Pu METAL EXCHANGE SAMPLES
S T U
SELECTED RF Pu METAL BUTTONS

Graphite Molds

Residual Oxide removed by wire brushing.

Metal held in crucible at 925°C - 45" before pouring.

Homogenized in mold - 2 hours.

Ingot in casting furnace overnight to eliminate warpage.

Ingot cropped 1/4" all sides - sprue removed.

Machined into 1 1/4" squares - flat sides milled to 0.080"

Machine oil removed by ultrasonic degreasing w/trichlor.

Dried w/isopropanol

1300 + grams of Pu Coupons

The T metal was removed from the exchange since the nickel content of ~2000 ppm is not representative of material handled by any of the laboratories.

W 204mm
H 229mm
TH 32mm
W 8 1/64" X 9 1/64" X 0.110" T 28
H
TH

Anderson also supplied the following key showing which metals had been distributed for each date and sample number.

DATE OF EXCHANGE	METAL SAMPLE			
	1	2	3	4
June 74	R	R	H	H
Sept 74	H	R	R	
Dec. 74	R	H	R	H
Mar 75	H	H	R	R
June 75	R	H	R	H
Sept 75	R	H	R	
Dec 75	S	T	S	T
Mar 76	U	U	S	S
June 76	U	U	S	S
Sept 76	U	S	S	U

One suggestion made was to include more metals in the program so there would be more continuity over a longer period of time. One disadvantage of increasing the number of metals would be less information on each for discussion at the yearly meeting. No other action was taken.

A question of would it be possible to prepare more samples at one time, so the sample would be available for a longer period of time, was asked. Anderson stated there may be a problem of criticality when casting but said he would look into this subject.

C. Pietri, NBL, said they had completed a major part of their move of the plutonium facilities from New Jersey to Argonne, Ill. and they would be actively entering the program in March, 1977. He also said his address is now:

Charles E. Pietri, Bld. 350
New Brunswick Laboratory
9800 South Cass Ave.
Argonne, Ill. 60439

Phone - Lab - 388-2070

Office - 388-5456

Pietri also mentioned the National Bureau of Standards plutonium materials will be stored and shipped from NBL in Argonne instead of by NBS in Gaithersburg, Md.

R. Foster, SRP, said they were interested in storing historical plutonium samples for periods of time up to 20 years. They would prefer to not use glass containers because of breakage problems. He asked if anyone had experience with polystyrene vials. Their experience at SRP indicates much discoloration after 3 to 5 years. Several persons confirmed the discoloration.

The week following the meeting, Walden, LLL, had occasion to observe recovery operations of plutonium stored in both glass and polystyrene vials for 13 years. Plutonium metal stored in glass had oxidized some - <5% estimated. The plutonium in the polystyrene vials had completely converted to oxide, the vials were badly discolored, and fractured and shattered quite easily along the longitudinal axis with only light finger pressure. Anyone having information that could be of help can call Foster directly. His FTS number is 284-4250.

Foster stated that SRP had been requested to routinely prepare plutonium metal that would contain <100 ppm carbon and <125 ppm or preferable <100 ppm of aluminum. Investigations showed that nitric acid used in processing the plutonium contained 2.5 ppm aluminum and this level in the acid added 23 ppm aluminum to the final metal product. The carbon content of the calcium used in the final metal reduction step of the process is variable and the lowest carbon content calcium will introduce enough carbon to raise the final carbon level in the plutonium to greater than the desired 100 ppm. They have not resolved this problem as yet.

A topic, that seems to arise every few years, was discussed at this meeting. What is the purpose of the exchange program, are we working towards the stated goals, is the program worth the considerable effort that is expended etc.? Each laboratory was requested to review the program and its stated goals, which are listed below. The laboratories are then requested to list what they think the goals should be and send them to John Walden, LLL, who will derive a new set of goals from everyone's suggestions and present them at our next meeting. Any other thoughts on the way the program is being run, the format of the annual meeting, etc. could also be included.

GOALS OF SAMPLE EXCHANGE PROGRAM

1. Discuss measurement differences.
2. Review and improve analytical measurements and methods.
3. Discuss new measurement capabilities.
4. Provide data to ERDA on measurement capabilities to evaluate shipper-receiver differences.
5. Provide characterized or standard materials as necessary for exchange participants.
6. Provide a quality control program for plutonium analyses.

Herman Anderson, RF, announced he was retiring and this was his last sample exchange meeting. The mechanics of the program have been handled by Rocky Flats since the inception and the success of the program is due in no small part to his active participation.

The next meeting will be hosted by Los Alamos Scientific Laboratory and will be held November 16, 1977 for the Isotopic meeting and November 17-18, 1977 for the Metal meeting. Lawrence Livermore Laboratory will chair the meeting and prepare the agenda.

John C. Walden
Plutonium Metallurgy
Metals and Ceramics Division

JCW:rjb