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TITLE: COMPARISON OF THE SAVANNAH RIVER SITE BILLET ACTIVE WELL COINCIDENCE COUNTER AND TWO CALIFORNIUM SHUFFLERS

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COMPARISON OF THE SAVANNAH RIVER SITE BILLET ACTIVE WELL COINCIDENCE COUNTER AND TWO CALIFORNIUM SHUFFLERS

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ABSTRACT

A Scrap Californium Shuffler at the Savannah River Site (SRS) was calibrated to assay the U-Al cores of billets (an intermediate step in the SRS reactor fuel fabrication cycle). The precision of the Scrap Shuffler over several years has been approximately 0.50%. A typical total uncertainty for the assay of a core on the Scrap Shuffler is approximately 0.33% for a twelve minute assay. The precision over several months and a typical total uncertainty for the Billet Active Well (neutron) Coincidence Counter (BAWCC) are approximately 1.0% and 1.9%, respectively, for a fifteen minute assay. A new Billet Californium Shuffler specifically designed for assaying SRS billets has yielded precision (over one month) and total uncertainty results of 0.40% and 0.69%, respectively, for an eight minute assay. The introduction of a measurement point into the fuel fabrication cycle to replace estimates based upon material weight will greatly enhance material and process control in the Reactor Materials area of SRS. The use of all three instruments provides a comparison of the relative merits of Active Well (neutron) Coincidence Counters (AWCCs) and shufflers for the assay of homogeneous and geometrically simple material containing ^{235}U. The measurement precisions, systematic and random uncertainties, as well as the procurement and operation of each instrument will be compared.

INTRODUCTION

The Reactor Materials area of the Savannah River Site (SRS) manufactures Li-Al and U-Al tubes for the SRS reactors. Process control and DOE Order 5633.3 require the measurement of the ^{235}U content of items produced in the area.

A billet is an intermediate stage in the fuel tube manufacturing process and is described in References 1 and 2. Three solutions to the problem of measuring the ^{235}U content of billets have been proposed. First, in approximately 1985, the procurement of a Billet Californium Shuffler (BS) was begun. The BS arrived at SRS at the end of January 1991. Also in 1985, a program was begun that would utilize the existing Scrap Californium Shuffler (SS) to assay the U-Al cores of the billets for ^{235}U before the cores were sealed into aluminum containers. This program was completed in Fall 1990. The third solution, proposed in 1988 and received in Summer 1990, was the Billet Active Well (neutron) Coincidence Counter (BAWCC)

STANDARDS

To calibrate these three instruments, 30 U-Al cores varying in ^{235}U mass, enrichment, and geometry (two different lengths and diameters) were cast from virgin uranium, i.e., uranium that has not been irradiated in a reactor. The concentrations of uranium isotopes in four samples drilled out of each U-Al core were determined by New Brunswick Laboratory (NBL). These concentrations were then used to assign the mass of ^{235}U to the standard cores. The precision of the NBL replicate analyses were very good, but the overall scatter of the results from each core yielded final uncertainties in the mass of ^{235}U in the standard cores of approximately 1% to 3%.

The analyses of several samples of two cores varied a great deal more than the samples from the other cores, so these two cores were not used in the calibration of the instruments. Also, the four cores fabricated at 44.5 weight percent (wt%) ^{235}U were finished improperly and could not be used in the current calibration measurements. Efforts are proceeding to make these cores useable for future calibrations.

SCRAP CALIFORNIUM SHUFFLER (SS)

References 1 and 2 do not discuss this shuffler, so a brief description of the SS, pictured schematically in Figure 1, will be given here. Reference 3 gives a much more detailed description of the SS and the initial efforts to calibrate it. The SS was the first shuffler built for use outside of Los Alamos National Laboratory (LANL). It was delivered to SRS in 1979. It assays primarily scrap material, but recently has been calibrated to assay billet cores.

During an assay, the ^{252}Cf neutron source follows a path that is perpendicular to the height axis of the sample. The source neutrons pass through approximately 3.8 cm of tungsten and 8.9 cm of nickel before reaching the sample. This spectrum tailoring lowers the source neutron energy somewhat to reduce fissioning of the even isotopes of uranium

The calibration curve for assaying cores in the SS is shown in Figure 2. For this and all following figures, the error bars reflect both the uncertainty in the instrument calibration measurement and in the mass of ^{235}U in the standard cores. The curve is a straight line with y-intercept equal to zero and no enrichment dependence over the range of 50 wt% to 80 wt%. In addition, both types of cores, though geometrically different, lie on the same curve. The short term precision for 10 runs of each standard during the calibration measurements was on average 0.25%. The long term precision measured by assaying a check standard whenever items are assayed is approximately 0.50%.

The total uncertainty is determined by combining in quadrature the systematic uncertainty with the random uncertainty of a single assay. The systematic uncertainty in the calibration coefficient was calculated by the regression routine to be 0.21%. A typical random uncertainty for a single 12-minute assay as calculated by the SS software is 0.26%. Therefore, a typical one sigma value for the total uncertainty is 0.33%.

While the results from the SS are very good, the operation and maintenance of this 12-year-old instrument are very difficult. No user-friendly interface exists, and the computer needs to be reset frequently to clear errors. The operator exposure limit when the SS was built was 5 mrem/hr which is an order of magnitude higher than the current limit. As for maintenance, the ^{252}Cf neutron source is replaced every five years, after which time the 887 microgram (2.0 x 10^9 n/s, 480 mCi) source has decayed to 239 micrograms (5.5 x 10^8 n/s, 130 mCi). This replacement poses a radiation hazard to the individuals facilitating the change. Also, the instrument hardware and software are so intimately connected that the original operating system on the PDP-11/23 computer can not be upgraded. Troubleshooting operating problems is very complex. Hardware upgrades also can not be instituted.

BILLET ACTIVE WELL COINCIDENCE COUNTER (BAWCC)

The discussion of the design and testing of this instrument is given in Reference 1.

The calibration curves for assaying billets in the BAWCC are shown in Figures 3 and 4. As was the case for the SS, the neutron count rate does not depend upon the enrichment of the uranium. This is the result of the moderation of the AmLi source neutrons by approximately 1.7 cm of high density polyethylene. This moderation also reduces the neutron penetrability of the billets.

In contrast to the SS, the two types of billets lie on different calibration curves. As shown in the figures, the final calibration curves are straight lines with non-zero y-intercepts. Several other types of curves were attempted in the fitting process, and all yielded approximately the same results.

The measured precision typically agrees with the precision calculated through propagation of errors for the observed count rates. The average precision of five consecutive assays during a calibration measurement was 0.90%. Two standards have been used as check standards for approximately six months. The precision of these measurements is 1.0%. A typical total uncertainty for a single assay is 1.9%, of which the systematic uncertainty in the calibration equation is 0.60%, and the random uncertainty is 1.8%. This fairly large random uncertainty is caused by the high neutron background in the BAWCC from counting the AmLi source neutrons concurrently with the coincident fission neutrons produced in the sample.

Operation of the BAWCC is very simple. A computer is convenient, but not required. The shift register electronics can accumulate data in a stand-alone mode, although a short, simple computer code eases analysis of the data. The code is also operator-friendly since it requires only the billet identification, number of cycles to run, and whether to report the data as a count rate or a gram value. The radiation dose rate from the BAWCC can not be measured above room background (<2mrem/hr). This operator protection is accomplished without massive amounts of shielding since the AmLi neutron activity (1.0×10^5 n/s) is relatively low and the many gamma rays (2300 mCi) from the ^{241}Am are blocked by tungsten source holders.

The BAWCC is also simple to maintain. The single moving part is the elevator mechanism for loading/unloading the billets, and it is controlled by individual "UP" and "DOWN" pushbuttons. Troubleshooting is eased by not relying on a computer to operate the BAWCC. The ^{3}He detectors, amplifiers, single channel analyzers, and shift register are all commercially available and can be replaced in the field very quickly. While the useful life of the AmLi (^{241}Am half life = 433 years) neutron sources is not known, they should not require replacement very often.

BILLET CALIFORNIUM SHUFFLER (BS)

The discussion of the design and testing of this instrument is given in Reference 2.

The calibration curves for assaying billets in the BS are shown in Figure 5. A count rate dependence upon enrichment, but not billet geometry, is evident. This dependence is the result of some fissioning of the even isotopes of uranium since the ^{252}Cf source neutrons are not moderated. However, this lack of moderation also provides greater neutron penetrability of the billets which allows less sensitivity to any possible inhomogeneities in the billet core.

Within each enrichment, a straight line calibration with a non-zero y-intercept fits the data well. The BS computer stores a separate calibration for each enrichment. The software interpolates between the two calibration enrichments that straddle the value for the enrichment of the sample billet which is input by the operator. The average precision of five consecutive runs during a calibration measurement was 0.23%. The same two check standards as for the BAWCC have been assayed in the BS for one month. The precision of these measurements is 0.40%. The typical total uncertainty for a single assay is 0.74%, of which the systematic uncertainties due to the calibration curves and various correction factors are 0.69%, and the random uncertainty of a single measurement is 0.28%.

Through the computer interface, the BS is easy to operate; however, if the computer is inoperable for any reason, the instrument can not function. Due to the complexity of the BS, several menu options are devoted to safety and performance checks. The computer does allow automatic operation and analysis of the performance check data to determine whether the BS is operating in a statistically acceptable manner. In a regular assay mode, the operator is required to input only the billet identification and its ^{235}U enrichment. Due to the large amount of neutron shielding on the BS, the radiation dose rate from the instrument can not be measured above room background (<2mrem/hr).

The BS has not been in use long enough to determine the maintenance requirements of the instrument. However, the ^{252}Cf neutron source will be changed after approximately three years. Source removal and installation are easy, and the BS requires a relatively small 60 microgram (1.4×10^8 n/s, 32 mCi) ^{252}Cf source, so the radiation exposure to personnel will be small when the neutron source must be changed. The source transfer system and very large door are the moving pieces of the BS.

The BS is a technically complex instrument due to the software intensiveness of both the safety monitors and the cycle of irradiation, delayed neutron counting, and analysis. This complexity will probably make diagnoses and repairs of some problems challenging, but possible.

COMPARISON

Table 1 lists various attributes of the three instruments described above. While the Scrap Californium Shuffler (SS) has the very appealing calibration of a straight line with y-intercept equal to zero, there is nothing intrinsically incorrect about the calibration curves for the other two instruments. The calibrations for the Billet Californium Shuffler (BS) do include an enrichment dependence. The enrichment dependence is not desirable, but it is also not catastrophic as long as the production billets to be assayed on the BS have similar isotopic contents to the standard billets. The hardness of the source neutron spectrum that results in the enrichment dependence also provides the best neutron penetrability for the BS.

Both the short-term and long-term precisions of the BS are the best of the three instruments despite the fact that it requires the shortest assay time. The SS has the next best precision, which indicates that four orders of magnitude greater source neutron emission can outweigh the geometrical advantages of the Billet Active Well Coincidence Counter (BAWCC). For the BAWCC and the BS, the short-term precision is the average relative standard deviation of five calibration runs for each of the 30 standard billets. For the SS, the short-term precision is the average relative standard deviation of 10 calibration runs for each of the 30 standard billet cores. For all three instruments, the long-term precision is the relative standard deviation for a check standard that was assayed each day before beginning measurements. The SS check standard has been assayed over a period of several years, while the check standards for the BAWCC and BS have been assayed for only six months and one month, respectively.

The typical random uncertainty for a single assay is lowest for the SS, with the BS the next lowest. For equal assay times, the contribution of the random uncertainty to the total uncertainty would be equal for the BS and the SS. The BAWCC has a much larger random uncertainty associated with it than the shufflers. The systematic uncertainty for the BS is larger than for the SS and the BAWCC. Each enrichment curve for the BS has fewer standards available to determine it which results in a larger systematic uncertainty. Despite this, the total uncertainty for the BS is less than 1%, while the total uncertainty for the BAWCC is nearly 2%. With the current length of assay times, the best total uncertainty is obtained by the SS.

Computer operation is required for the two shufflers, but billets can be assayed in the BAWCC even if its computer becomes inoperable. The BAWCC code is simple enough that the small executable file and two small data files could be transported easily to an operating computer. That computer would then merely need to be connected to the BAWCC shift register via a single RS232 cable. This portability is not possible on the SS due to the interconnection of software and hardware. The BS code can be transported, but employing a different computer would require much more time and effort to affect the transfer due to the large number of files (approx. 80) and external wiring involved in the operation of the BS.

Maintenance and troubleshooting of each system varies in much the same manner as the computer systems of the instruments. The hardware/software interconnection in the SS makes replacement of malfunctioning or old items nearly impossible. In contrast, the BAWCC and the BS are new and designed for easy replacement of parts. Troubleshooting the BAWCC should be easier than the BS due to the simpler data analysis. However, observing the results from individual detector banks is nearly impossible in the BAWCC since the data from all banks is summed before entering the shift register. External lights can indicate gross problems with individual banks, but to see fine problems the BAWCC must be opened. On the other hand, the BS computer monitors each bank individually. Computer maintenance and troubleshooting is much easier for the BAWCC than the BS due to the vast difference in the computer requirements for each instrument.

The final four items in Table 1 illustrate non-technical concerns. The indicated sizes include the assay instruments, electronics racks, computers, terminals, printers, and tables for all of the computer peripherals. The SS already existed and required a minor additional piece of equipment to assay the billet cores. As can be seen in the table, the excellent precision of the BS did not come without a price. The shielding required to protect personnel from the ^{252}Cf neutron source is much greater than the shielding required in the BAWCC. Also, since the BS was custom-made by LANL and is more complex than an AWCC, the procurement time was much longer and the cost was much higher for it than for the BAWCC. The $650,000 in Table 1 for the BS was the SRS contribution; DOE provided additional funds. As more shufflers are produced, the development cost associated with each one will obviously be reduced, but the total cost will still be higher than the cost for AWCCs.

FUTURE

The SS is currently performing accountability measurements on U-Al scrap material and billet cores. As stated previously, upgrades are not feasible, so the SS will not be enhanced in the future. A new SS, primarily for assaying scrap, but also capable of assaying billet cores is being planned to replace the current SS. It will not arrive at SRS for several years.

In an attempt to alleviate the enrichment dependence of the BS, polyethylene sleeves of several thicknesses that will fit between the ^{252}Cf neutron source and the billets are being fabricated. The desire is to moderate the source neutrons so that they will be less sensitive to the even uranium isotopes in the billets. To investigate the effect of the polyethylene on neutron transmission through the billets, pieces of U-Al will be placed on the outside of the billets. If the moderated neutrons still uniformly interrogate all portions of the billets, the BS will measure a total ^{235}U equal to the sum of the ^{235}U in the billet and the ^{235}U in the extra pieces. Similar tests are planned for the BAWCC as well.

SUMMARY

The three instruments, a Scrap Californium Shuffler (SS), a Billet Active Well (neutron) Coincidence Counter (BAWCC), and a Billet Californium Shuffler (BS), have all performed well in assaying U-Al for ^{235}U. Each instrument exhibits various strengths and weaknesses. Both shufflers yielded better precisions than the BAWCC, with the assay time of the BS being approximately one half of the assay time of the BAWCC. The total uncertainty of the SS is lower than the total uncertainty of the BAWCC and the BS. The total uncertainty of the BAWCC is dominated by the random uncertainty of the instrument resulting from the relatively low AmLi neutron emission rate and high background. The dominant factor in the total uncertainty of the BS is the systematic uncertainty of the calibration equations since the billet standards must be separated according to enrichment.

The enrichment dependence of the BS complicates matters in a production environment where the isotopic concentrations of the feed material can vary. Presently, the isotopic concentrations in the production uranium are close to those of the standard billets, so the increased penetrability of the BS neutrons over the BAWCC neutrons will result in less sensitivity to any possible inhomogeneities in the billets.

Some advantages of the BAWCC over the BS are its relative simplicity which allows its use without a computer, easy maintenance and troubleshooting, compact size, shorter procurement time, and lower cost.

The decision on whether to purchase an AWCC or a californium shuffler must be based on competing forces. Shufflers assay material with better precision than AWCCs. Therefore, if the content of the calibration standards is reasonably well known, a shuffler will yield a smaller total uncertainty than an AWCC. However, if the content of the calibration standards is not known very well, a high precision instrument will not be fully utilized until high accuracy standards or many lower accuracy standards can be obtained. AWCCs have maintenance and procurement advantages over the shufflers. The user must decide whether the higher cost in time and money is worth the higher precision of a shuffler.

ACKNOWLEDGEMENTS

The authors are indebted to F. G. Armstrong and E. W. Jenkins for their statistical work on this project.

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2. P. M. Rinard, K. Kroncke, C. M. Schneider, R. D. Biddle, E. T. Sadowski, and R. V. Studley, "A Shuffler for Uranium Billets," these proceedings.

3. T. W. Crane, "Test and Evaluation Results of the ^{252}Cf Shuffler at the Savannah River Plant," Los Alamos Scientific Laboratory Report, LA-8755-MS, (March 1981).

CAPTIONS

1. Schematic diagram of the SRS Scrap Californium Shuffler from Reference 3.

2. Calibration curve for assaying billet cores in the Scrap Californium Shuffler.

3. Calibration curve for assaying BI billets in the Billet Active Well Coincidence Counter.

4. Calibration curve for assaying BO billets in the Billet ^ 'ive Well Coincidence Counter.

5. Calibration curve for assaying billets in the Billet Californium Shuffler.

KEY WORDS

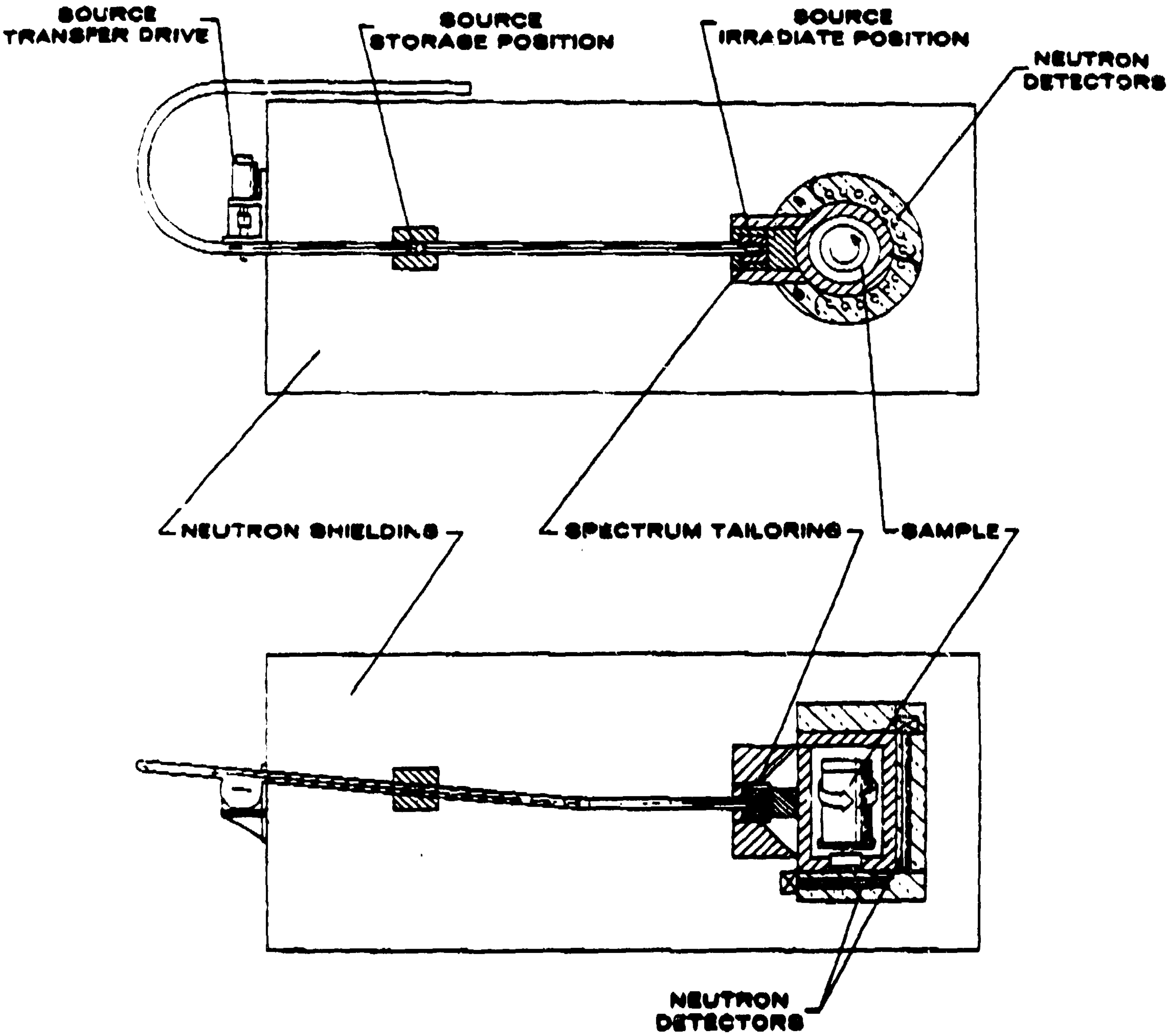
Measurement Technology, Neutron Assay, Active Well (neutron) Coincidence Counter, Californium Shuffler

TABLE 1
Comparison of Attributes

Attribute	SS	BAWCC	BS
Calibration	y = mx	y = mx + b	y = mx + b
Enrichment dependence	no	no	yes
Neutron pe[illegible]t.ability	intermediate	worst	best
Assay time	12 mins	15 mins	8 mins
Precision (short-term)	0.25%	0.90%	0.23%
Precision (long-term)	0.50%	1.0%	0.40%
Random uncertainty	0.26%	1.8%	0.28%
Systematic uncert. (number of standards per calibration curve)	0.21% (24)	0.6[illegible]% (15 BI, [illegible] BO)	0.69% (4 to 7)
Total uncertainty	0.33%	1.9%	0.74%
Operation without computer	none	laborious	none
Operation with computer	not simple	simple	simple
Maintenance/ Troubleshooting	most difficult	easiest	intermediate

TABLE 1 (cont.)
Comparison of Attributes

Attribute	SS	BAWCC	BS
Required floorspace	4.5m x 1.5m	2.2m x 0.9m	5.3m x 2.2m
Funding/ Specifications	-	8 mos.	33 mos.
Design/ Fabrication	2 mos. (adaptation)	9 mos.	27 mos.
Cost	-	$150,000	$650,000+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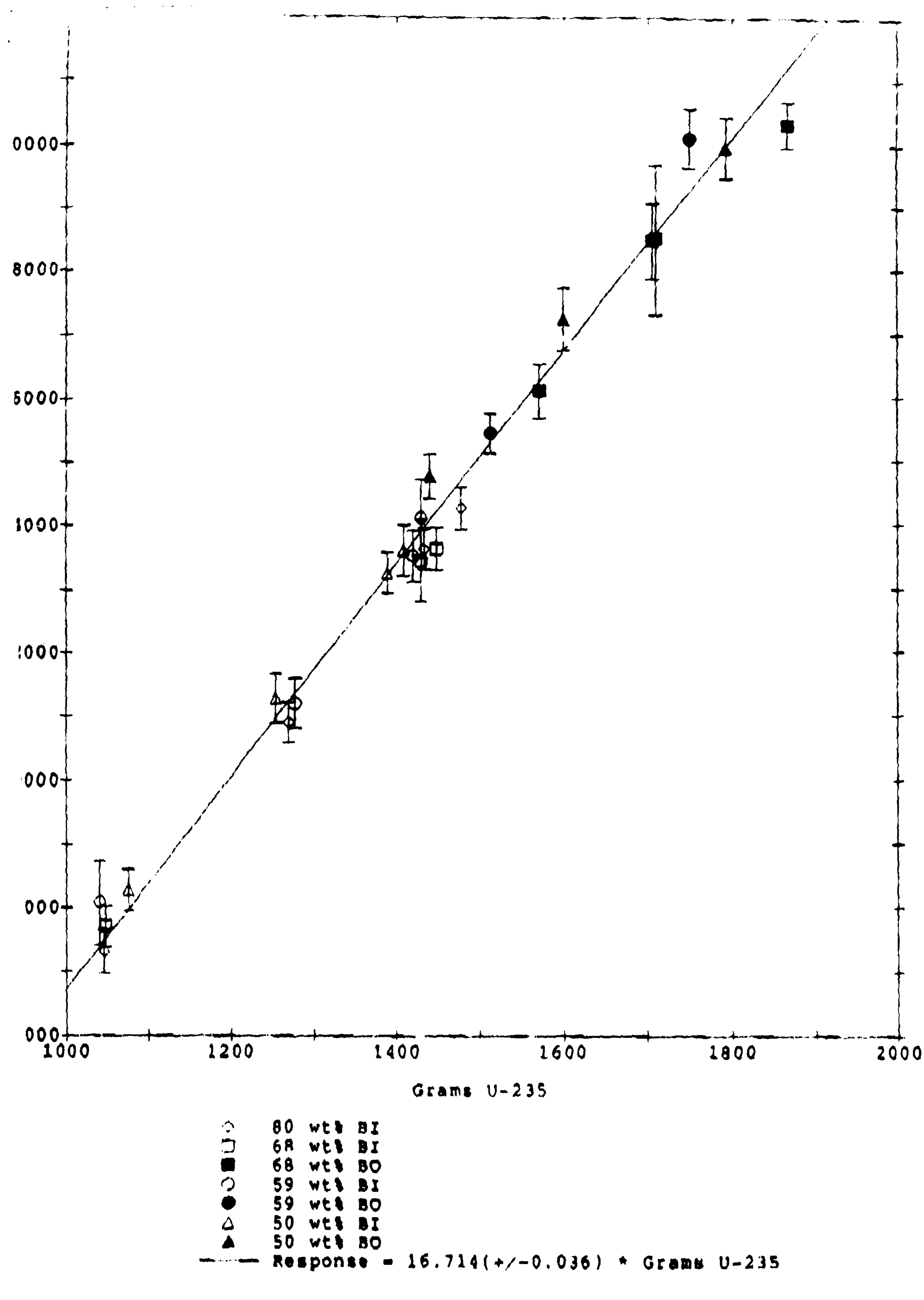
DETECTORS



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8000
6000
4000
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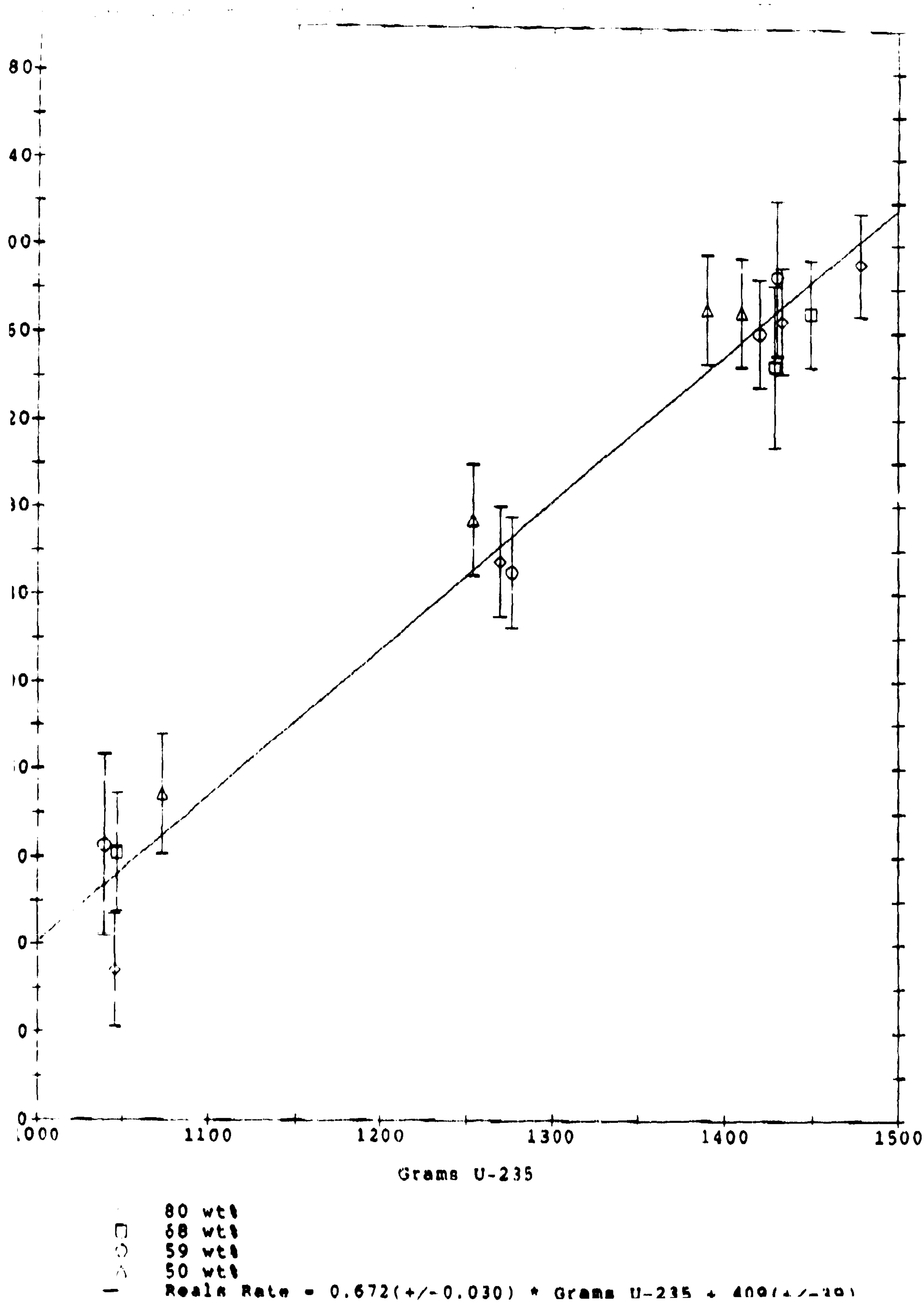
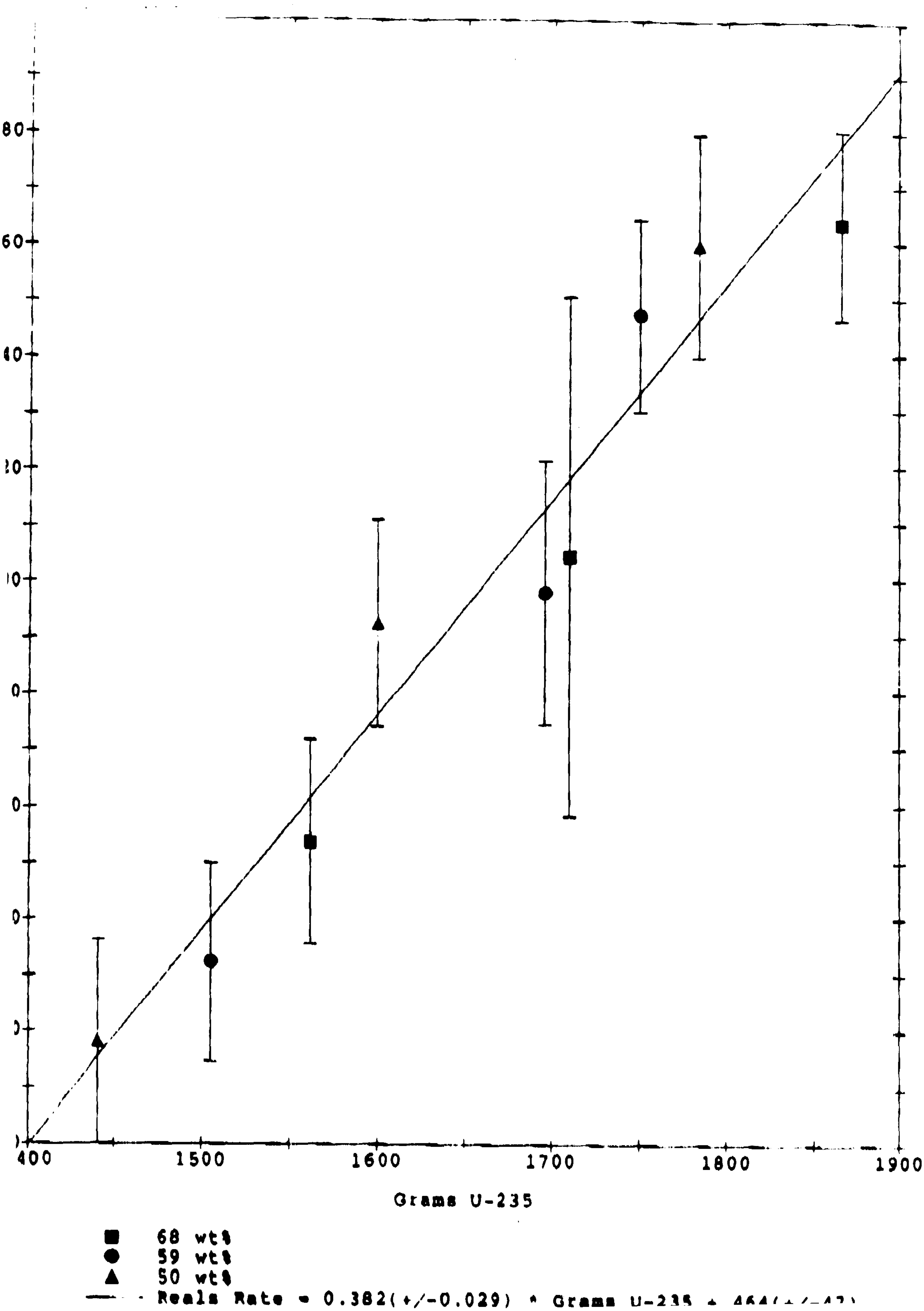
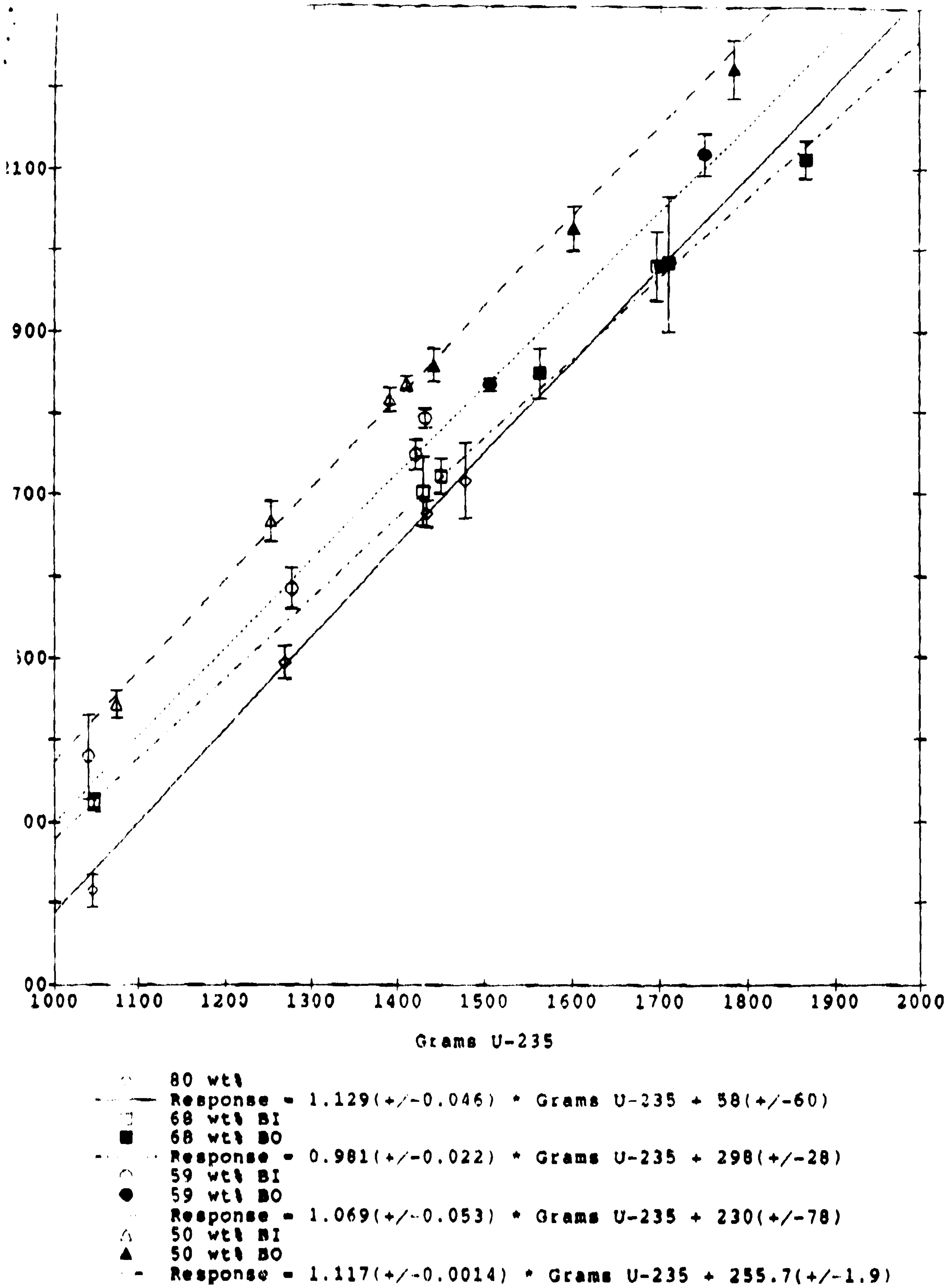
1000
1200
1400
1600
1800
2000
Grams U-235
80 wt% BI
68 wt% BI
68 wt% BO
59 wt% BI
59 wt% BO
50 wt% BI
50 wt% BO
Response = 16.714(+/-0.036) * Grams U-235



80
40
00
50
20
30
10
0
1000
1100
1200
1300
1400
1500
Grams U-235
80 wt%
68 wt%
59 wt%
50 wt%
Reals Rate = 0.672(+/-0.030) * Grams U-235 + 409(+/-



80
60
40
400
1500
1600
1700
1800
1900
Grams U-235
68 wt%
59 wt%
50 wt%
Reals Rate = 0.382(+/-0.029) * Grams U-235 + 464(+/-47)


900
700
1000
1100
1200
1300
1400
1500
1600
1700
1800
1900
2000
Grams U-235
80 wt%
Response = 1.129(+/-0.046) * Grams U-235 + 58(+/-60)
68 wt% BI
68 wt% BO
Response = 0.981(+/-0.022) * Grams U-235 + 298(+/-28)
59 wt% BI
59 wt% BO
Response = 1.069(+/-0.053) * Grams U-235 + 230(+/-78)
50 wt% BI
50 wt% BO
Response = 1.117(+/-0.0014) * Grams U-235 + 255.7(+/-1.9)