ORNL-5215
Special

Development of Krypton-85 Sources for the Bureau of Mines Coal Dust Combustion Analyzer

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Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
Price: Printed Copy [redacted]; Microfiche $2.25

ORNL-5215
Special

Contract No. W-7405-eng-26

Radioisotope Department

DEVELOPMENT OF KRYPTON-85 SOURCES FOR THE BUREAU OF MINES COAL DUST COMBUSTION ANALYZER

W. C. Bradley, F. N. Case, N. H. Cutshall, K. W. Haff

OPERATIONS DIVISION

Final Report

Prepared for

UNITED STATES DEPARTMENT OF THE INTERIOR
BUREAU OF MINES

under Contract No. DOM H0232024

(ERDA 40-400-73)

Manuscript Completed June 1976

Date Published: September 1976

Prepared by
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

REPORT DOCUMENTATION PAGE

1. Report No.	2.	3. Recipient's Accession No.
4. Title and Subtitle Development of Krypton-85 Sources for the Bureau of Mines Coal Dust Combustion Analyzer		5. Report Date June 1976
		6.
7. Author(s) W. C. Bradley, F. N. Case, N. H. Cutshall, K. W. Haff		8. Performing Organization Report No. ORNL/5215
9. Performing Organization Name and Address Oak Ridge National Laboratory Post Office Box X Oak Ridge, Tennessee 37830		10. Project/Task/Work Unit No. ERDA 40-400-73
		11. Contract or Grant No. H0232024
12. Sponsoring Organization Name and Address Office of the Assistant Director-Mining Bureau of Mines Department of the Interior Washington, D.C. 20241		13. Type of Report Final
		14.
15. Supplementary Notes		

16. Abstract
Americium-241 used as a radiation source in a field instrument for measuring coal concentrations in mine dust presents a contamination hazard if the instrument is damaged. Alternative source designs based on ^{85}Kr were developed and evaluated. Krypton-85 beta particles were used to excite x-rays from a target and the x-rays were used in place of the ^{241}Am gamma radiation. Output of sources using ^{85}Kr foils was not high enough because the concentration of ^{85}Kr in the foils was too low. Output of sources using gaseous ^{85}Kr, encapsulated in tantalum was adequate for the coal dust analyzer although the associated radiation field from ^{85}Kr gamma rays make these sources inappropriate for application in a portable instrument.

17. Originator's Key Words Coal dust analyzer; krypton-85; x-rays; backscatter; americium-241	18. Availability Statement
19. U.S. Security Classif. of the Report Unclassified	20. U.S. Security Classif. of This Page Unclassified
21. No. of Pages 21	22. Price

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FOREWORD

This report was prepared by Oak Ridge National Laboratory, Operations Division, Oak Ridge, Tennessee, under USBM Contract Number H0232024. The contract was initiated under the Coal Mine Health and Safety Program. It was administered under the technical direction of PM&SRC with Mr. J. Edmund Hay acting as the Technical Project Officer. Mr. Dean Priddy was the contract administrator for the Bureau of Mines.

This report is a summary of the work recently completed as part of this contract during the period May 10, 1973 to June 30, 1976. This report was submitted by the authors on June 30, 1976.

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INTRODUCTION

Dust explosions in coal mines are a severe hazard when the amount of coal dust mixed with rock dust reaches critical levels. The U. S. Bureau of Mines has developed a portable instrument for measuring the abundance of coal dust in mine dust samples.[1] With this instrument coal dust abundance is estimated by measuring the scattering of X-ray beams directed at the sample. For energy levels of less than 100 keV, high angle scattering is greater by coal dust than by rock dust since photoelectric absorption exceeds scattering and the electron density of rock dust is higher than that of coal dust.

The present instrument uses two 15 mCi ^{241}Am sources which nominally produce 5×10^6 60-keV photons/sec in a beam collimated within an 8° angle. Because of the potential contamination problem from alpha emitting ^{241}Am should the source capsules be broken, an alternative x-ray source would be desirable. Krypton-85 would be a logical candidate for this application because it would be dispersed rapidly in the event of source rupture and its 10.7-year half-life would provide a useful service life.

The photon emission from ^{85}Kr is too energetic (514 keV) and of relatively low yield (0.41%); however, the 0.67 MeV β-particles emitted with 100% yield can be used to excite secondary x-rays in suitable target materials. Furthermore, since different target materials can be chosen, the primary energy of the emitted x-rays can be selected from the range of 20 to 60 keV.

This report describes the production and testing of several prototype ^{85}Kr excited x-ray sources fabricated from two basic designs. One design used ^{85}Kr impregnated foils, sandwiched between foils of the target element whose x-rays are excited. Various numbers and thicknesses of target foils were investigated. The other design used enriched gaseous ^{85}Kr encapsulated in a container made of or lined with the target element. Several sources of different output were fabricated and tested.

FOIL SANDWICH SOURCE DEVELOPMENT

Krypton-85 impregnated foils are produced by impingement of ^{85}Kr ions on an aluminum foil. Although foils containing up to 1.0 mCi/cm^2 of ^{85}Kr have been made, limitations on production of foils impregnated with higher concentrations of ^{85}Kr are encountered when the ion beams are of non-uniform intensity and locally high ion intensity causes melting of the foils. Once produced, however, the foils are quite stable, and although some radioactivity may be removed from the foils by abrasion, ^{85}Kr does not appreciably diffuse out of the foils at room temperature and atmospheric pressure.

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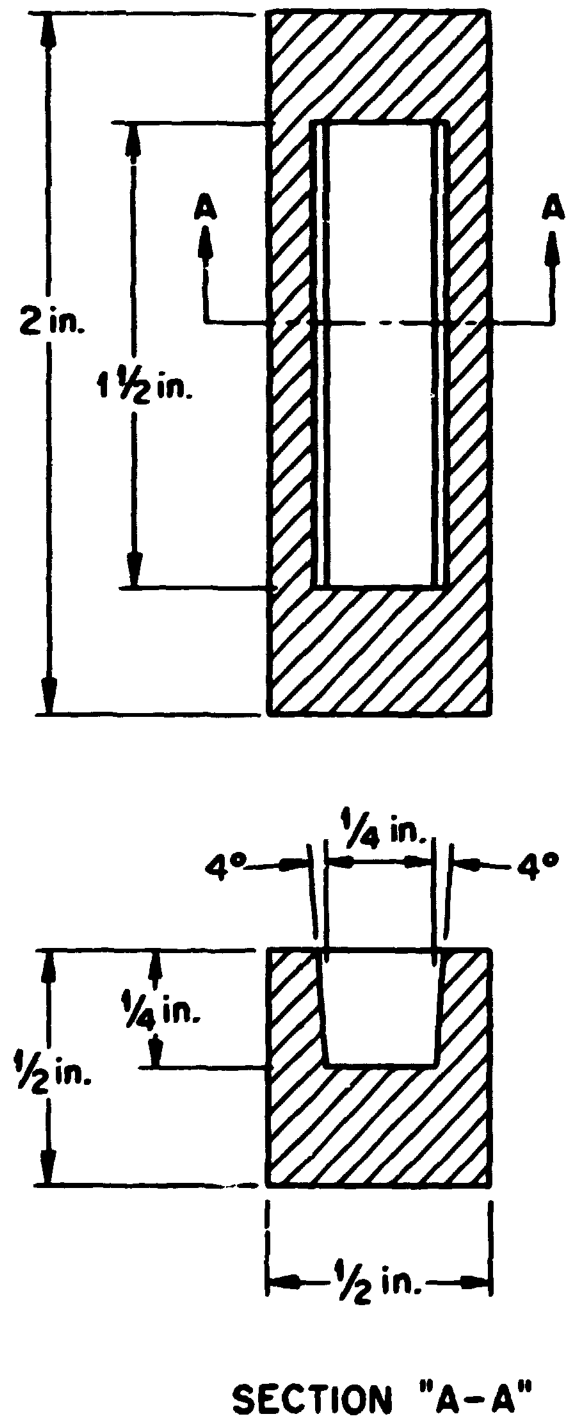

Fig. 1. Aluminum Holder for Krypton-85 Source

Sandwich type x-ray sources were fabricated at first by placing krypton impregnated foil into an aluminum holder (Figure 1) and covering the holder with a metal target. Beta particles are absorbed by the target and the resulting x-rays exit through the window. The photon spectrum for each fabricated source was measured using Ge(Li), Si(Li), or NaI(Tl) detectors in conjunction with a Nuclear Data Model 4410 multichannel analyzer system. The quantity of ^{85}Kr contained in the source was determined by counting the output of 514 keV gamma rays on detectors that had been calibrated using the 511 keV annihilation radiation from an IAEA ^{22}Na standard. The source outputs of x-rays were measured on detectors calibrated with an IAEA ^{241}Am standard which has photopeaks at 13.9, 17.8, 20.8, 26.3 and 59.5 keV.

Single Layer Foil Sources

Three sources were fabricated using single layers of target element foil to cover ^{85}Kr impregnated aluminum foil. Target elements used in these sources were molybdenum, cadmium, and tungsten. In the cases of molybdenum and cadmium, 0.006-in.-thick ^{85}Kr foils were placed in an aluminum tube and the tube was covered by the target element foil. In the case of tungsten the aluminum tube was replaced by a holder fabricated from Hevimet, a tungsten-copper alloy (Figure 1). In each case the 360° yields of characteristic K x-rays (summed K_α and K_β x-rays) and of photons with energies in the x-ray range were determined. In addition the effective flux of x-rays in a beam collimated to an 8° angle was also determined. Results (Table 1) are expressed as absolute yields, as ratios of K x-ray yields to total photon yields and as the efficiency of K x-ray production with respect to beta activity in the source foil.

Table 1. Output Characteristics of Single Layer Foil Sources

Target Element and Thickness (inches)	X-ray Energies (keV)		^{85}Kr in capsule (mCi)	X-ray Yield (photons/sec)		Ratio of K X-rays to 8-80 keV photons	K X-ray Production Efficiency Based on β decay rate (%)
	K_α	K_β		360°	8° Beam		
Molybdenum 0.006	17	20	21.7	7.5×10^6	1.7×10^5	0.28	0.94
Cadmium 0.005	23	26	67	8.75×10^6	1.9×10^6	0.33	0.35
Tungsten 0.002	59	68	23	2.62×10^6	5.8×10^4	0.048	0.31

The single layer source outputs are approximately 20 to 80 times too low to replace the presently used ^{241}Am sources. In order to increase output, without making the source larger, multiple layers of target and ^{85}Kr foils are necessary.

Tantalum Multilayer Sources

A series of sources each containing 38 mCi of ^{85}Kr in aluminum foils 1/8-in. wide by 1.5-in. long and 0.002-in.-thick, was constructed. These foils were sandwiched between 1/2-in. by 2-in. pieces of 0.001-in.-thick tantalum foil. The number of tantalum foil layers was varied to vary the target thickness. Energy spectra for this source series were analyzed using a 3-in. by 3-in. NaI(Tl) detector. Energies of the K_α and K_β x-rays of tantalum are 57 and 66 keV respectively.

As for the single layer sources, K x-ray photon yield for 360° emission, the K x-ray production efficiency relative to the total β-decay rate and the ratio of K x-ray yield to the total photon yield in the 35 to 80 keV energy range were determined (Table 2).

Table 2. Tantalum Multilayer Source Output Characteristics

Tantalum foil thickness (in.)	K X-ray 360° yield (photons/sec)	Ratio of K X-rays to 35-80 keV photons	K X-ray Production efficiency (%)
0.001	2.0×10^4	0.35	1.4
0.002	2.8×10^4	0.47	2.0
0.003	3.0×10^4	0.58	2.1
0.004	2.9×10^4	0.65	2.0

X-ray production efficiency does not vary significantly for target layer thicknesses of 0.002 to 0.004 in. but production from 0.001-in. foils is lower indicating that some beta-particles penetrate the 0.001-in. foils and are lost. The K x-ray-to-total-photon ratio increases monotonically as the number of foil layers is increased from 1 to 4. This effect is the result of attenuation of lower energy photons due to their absorption in the tantalum and, although it appears to continue as the number of target foils is increased, eventually the absorbtion of K x-rays would diminish production efficiency.

A second series of multilayered tantalum sources was constructed in different layered configurations but again with the same quantity of ^{85}Kr in each source. Six aluminum foils, 0.25-in. wide by 1.5-in. long by 0.002-in. thick, containing a total of 4.6 mCi of ^{85}Kr were used in each source. The tantalum foils were 0.5-in. by 2.0-in. long by 0.001-in. thick. A convention was developed for source designations to indicate the total layer thickness in thousanths of an inch of the foil layers. The outer layers are always Ta so that 4 6 2 6 4 means 4 layers Ta (0.004 in.), 3 layers Kr/Al (0.006 in.), 2 layers Ta (0.002 in.), 3 layers Kr/Al (0.006 in), and 4 layers Ta (0.004 in.).

As usual the K x-ray output for 360° emission, the K x-ray production efficiency and the ratio of K x-ray yield to the total photon yield for the 30 to 90 keV energy range were determined (Table 3).

Table 3. Tantalum Multilayer Source Output

Source configuration	K x-ray 360° yield (photons/sec)	Ratio of K X-rays to 30-90 keV photons	K X-ray production efficiency based on β decay rate (%)
4 4 1 4 1 4 4	1.1×10^6	0.59	0.63
5 4 1 4 1 4 5	9.9×10^5	0.62	0.58
4 6 2 6 4	9.9×10^5	0.58	0.58
4 6 4 6 4	9.2×10^5	0.59	0.54
5 6 4 6 5	8.5×10^5	0.63	0.50
5 6 2 6 5	9.4×10^5	0.62	0.55
5 12 5	8.1×10^5	0.58	0.48

Except for the 5 12 5 configuration, sources using 0.005-in. of Ta on the outside consistently yielded a higher K x-ray to total photon output as would have been expected from the previous study. Production efficiencies of the multilayer sources were all substantially lower than those of the single layer sources tested previously, however, indicating that the optimum total sandwich thickness had been exceeded. Thus a compromise must be made between production efficiency and spectral purity of the x-rays.

The last foil sandwich source tested was a multilayer source of a 5 8 2 8 5 configuration. This source was built so that the collimated output could be directly measured. It contained 3.45 mCi of ^{85}Kr on eight aluminum foils, 0.187-in. by 1.375-in. by 0.002-in. thick. The tantalum foils were 0.375-in. by 2.00-in. by 0.001-in. thick. Measured total K x-ray output was 6.7×10^5 photons/sec and production efficiency was 0.52%. A 2-in. cube of lead was drilled to hold the source and slotted so as to produce a beam collimated to 13° angle. The measured K x-ray yield in the collimated beam was only 1.3×10^4 photons/sec and was near the maximum attainable yield for the multilayer source design using available foils. At least 600 mCi of ^{85}Kr would be required in order to produce the required low energy x-ray beam of 5×10^6 photons/sec. Since the kryptonated aluminum foils presently available contain a maximum of 1 mCi/cm^2 of ^{85}Kr and an average of ~0.26 mCi/cm^2, it appears that the desired x-ray output cannot be produced from a source of reasonable physical dimensions made with presently available ^{85}Kr-impregnated foils.

GASEOUS KRYPTON-85 SOURCE DESIGN

Two types of gaseous krypton sources were designed to produce the desired low energy x-ray beam with an output of 5×10^6 photons/sec. Source capsules of 1/4-in. inside diameter and 1 1/2-in. internal length were used. An all tantalum capsule and an aluminum capsule with a tantalum inner liner were designed and fabricated for evaluation (Figures 2 and 3). The tantalum capsule wall thickness was 0.009-in. This capsule should withstand an internal pressure of 775 psig based on a 45,000 psi ultimate strength. The aluminum capsule wall

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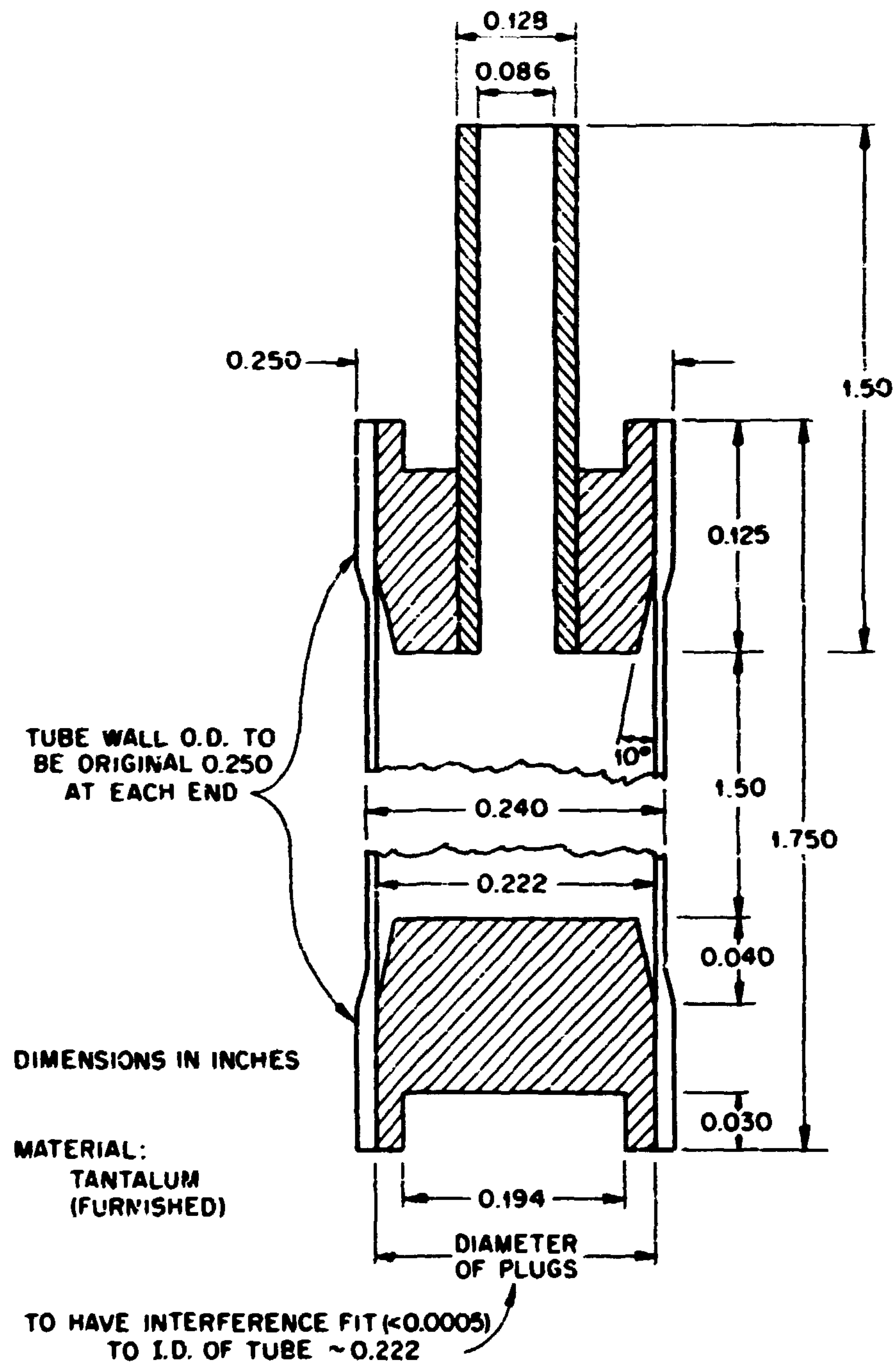

Fig. 2. Tantalum Krypton Source Holder

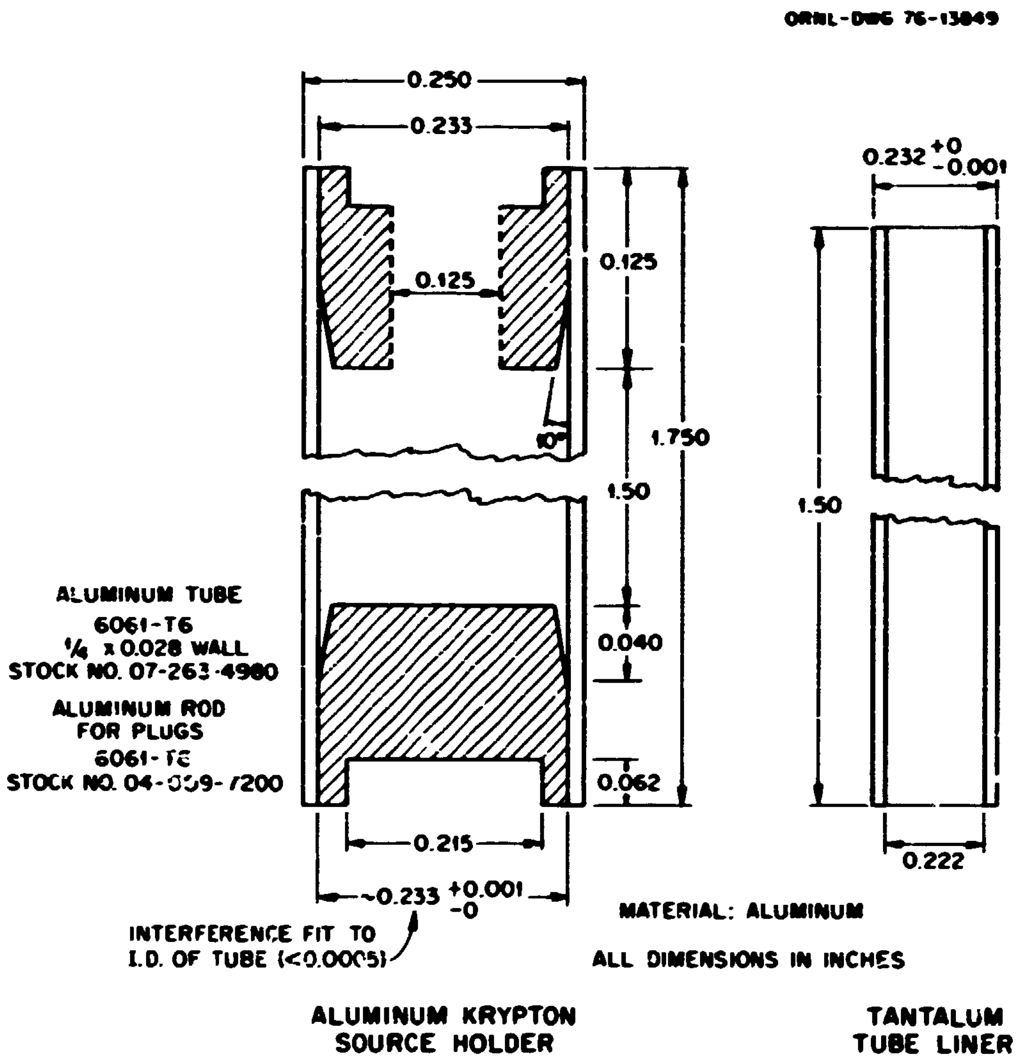

Fig. 3. Aluminum Krypton Source Holder
Tantalum Tube Liner

thickness was 0.0085-in. and should withstand an internal pressure of 390 psig based on a yield strength of 6000 psi. The calculated thickness of tantalum liner for optimum x-ray production was 0.005-in. The internal volume of each capsule is about 1 cm^3. Based upon the requirement of 600 mCi of ^{85}Kr, the internal gas pressure using 10% enriched ^{85}Kr would be 50 psig.

Krypton-85 Gas Source Fabrication

The tantalum and aluminum capsules (Figures 2 and 3) were welded in an inert atmosphere glove box using inert gas tungsten arc gas (GTA) welder. After the capsules were submerged in alcohol and pressure-leak checked they were loaded with ^{85}Kr as follows:

1. The specified volume of 10% ^{85}Kr was frozen into an evacuated flask of known volume.

2. The flask was warmed and the pressure checked.

3. The source capsule was connected to a manifold which contained a vacuum supply, a helium gas supply, and the premeasured krypton charge.

4. The source capsule was evacuated overnight.

5. The source capsule was cooled with liquid nitrogen in a chill block.

6. The vacuum was closed off.

7. The krypton was released from the flask and frozen out in the source.

8. Helium gas was allowed to fill the source up to approximately atmospheric pressure.

9. The 1/8-in. diameter fill tube was pinched off.

10. A GTA weld fused the fill tube to seal the capsule.

11. The source was placed into a sealed container for several hours after which the container air was sampled and counted for evidence of a possible leak.

Two tantalum encapsulated sources and one aluminum encapsulated source were completed. Outputs of the sources were then measured as for the previously tested sources (Table 4). In all cases an ^{241}Am IAEA calibrated source was counted each time a source or group of sources were analyzed. A ^{22}Na IAEA calibrated source was used to calibrate the Ge(Li) detector used to measure the ^{85}Kr 514 keV γ-rays.

Table 4. Output Characteristics of Gaseous Krypton-85 Source

Source	^{85}Kr (Ci)	Calculated capsule pressure (psig)	K X-ray Yields (photons/sec) 360°	8° beam	Ratio of K X-rays based on β decay rate
Ta source #1	0.585	77	9.3×10^7	2×10^6	0.44
Ta source #2	1.837	222	2.9×10^8	6.4×10^6	0.43
Al/Ta source	1.902	230	3.8×10^8	8.4×10^6	0.54

The aluminum encapsulated source lined with 0.005-in. of tantalum produced about 1.3 times the K x-ray output of tantalum source No. 2 even though both sources contained roughly the same amount of ^{85}Kr. This greater production efficiency for the aluminum encapsulated source is probably due to the lower thickness of the tantalum liner (0.005 in.) compared to the tantalum encapsulated source (0.009 in.). Production efficiencies are comparable to those of the multilayer sandwich sources and total outputs meet the desired level.

Calculated capsule pressures include the helium gas which was added to prevent contamination leakage of air into the capsules prior to seal welding the filler tube. These pressures are below the calculated maximum capsule pressures of 775 psig for the tantalum capsule and 390 psig for the aluminum capsule.

Photon distribution for the sources for energies of 5 through 80 keV were determined for several ranges (Table 5).

Table 5. Photon Distribution from Gaseous Krypton-85 Source

Source	Percent photons in energy range 5-20 keV	20-70 keV	70-80 keV	Ratio of K X-rays to total photon yield in ranges of 5-80 keV	55-70 keV
Tantalum Source 1	43.0%	51.1%	5.9%	0.136	0.450
Tantalum Source 2	44.2%	48.9%	6.7%	0.109	0.378
Aluminum/ Tantalum 1	43.1%	50.8%	6.0%	0.099	0.373

The ratios of K x-rays to total photons for the gaseous sources are lower than for the sandwich-type sources. It is not clear why this is so but the observation is consistent for all three gaseous sources.

Application of Gaseous Krypton-85 Sources

Since x-ray output of the gaseous sources appeared high enough, it was decided to test them in the Bureau of Mines instrument. Tantalum sources 1 and 2 were placed in shields fixed with collimators (Figure 4) and tested in a coal dust analyzer (Figure 5). In addition, radiation dose rates outside the analyzer were measured (Figure 6).

Radiation dose rates were highest at contact, 120 mr/hr, and were appreciable at 12 inches, 5 mr/hr. These rates are intolerable and shielding would have to be increased. When the unit was tested using coal dust and rock dust the results (Table 6) reflected the relatively high radiation environment in the analyzer.

Table 6. Count Rates Using Gaseous Krypton-85 in Coal Dust Analyzer (Net Count Rates in Parentheses)

Sample	Counting Rates (cts/sec)		
	20-77 keV	20-60 keV	5-20 keV
Background	160	115	376
100% Rock Dust	181 (21)	132 (17)	398 (22)
100% Coal Dust	193 (33)	144 (29)	402 (26)

The background count rates in the three energy ranges tested are all excessive compared to the backscatter count rate. The operating range for the instrument is the difference between the 100% coal dust and 100% rock dust or 12, 12 and 4 counts/second respectively for 20-77, 20-60 and 5-20 keV ranges. This means that counting times of 1-2 minutes (as are used with the ^{241}Am sources[1]) would yield a coefficient of variation of higher than 10% using the gaseous ^{85}Kr source. Either the counting times would have to be increased unreasonably or else the background count rate would have to be reduced by shielding the source on all sides except the window.

SUMMARY AND CONCLUSIONS

Several radiation sources, based on ^{85}Kr were evaluated as substitutes for ^{241}Am in the U. S. Bureau of Mines coal dust analyzer. Sources based on ^{85}Kr impregnated aluminum foils cannot yield an adequate x-ray output using presently available foils. At least a 30-fold increase in ^{85}Kr loading of the foils would be necessary. While there may be other applications for x-ray sources made from ^{85}Kr foils sandwiched between target element foils (molybdenum, cadmium, tungsten), such foils are unlikely to replace ^{241}Am in the coal dust analyzer.

Sources using enriched gaseous ^{85}Kr encapsulated in containers made from or lined with a target element, such as tantalum, have much higher x-ray yields approaching those of the currently used ^{241}Am sources. Unfortunately these

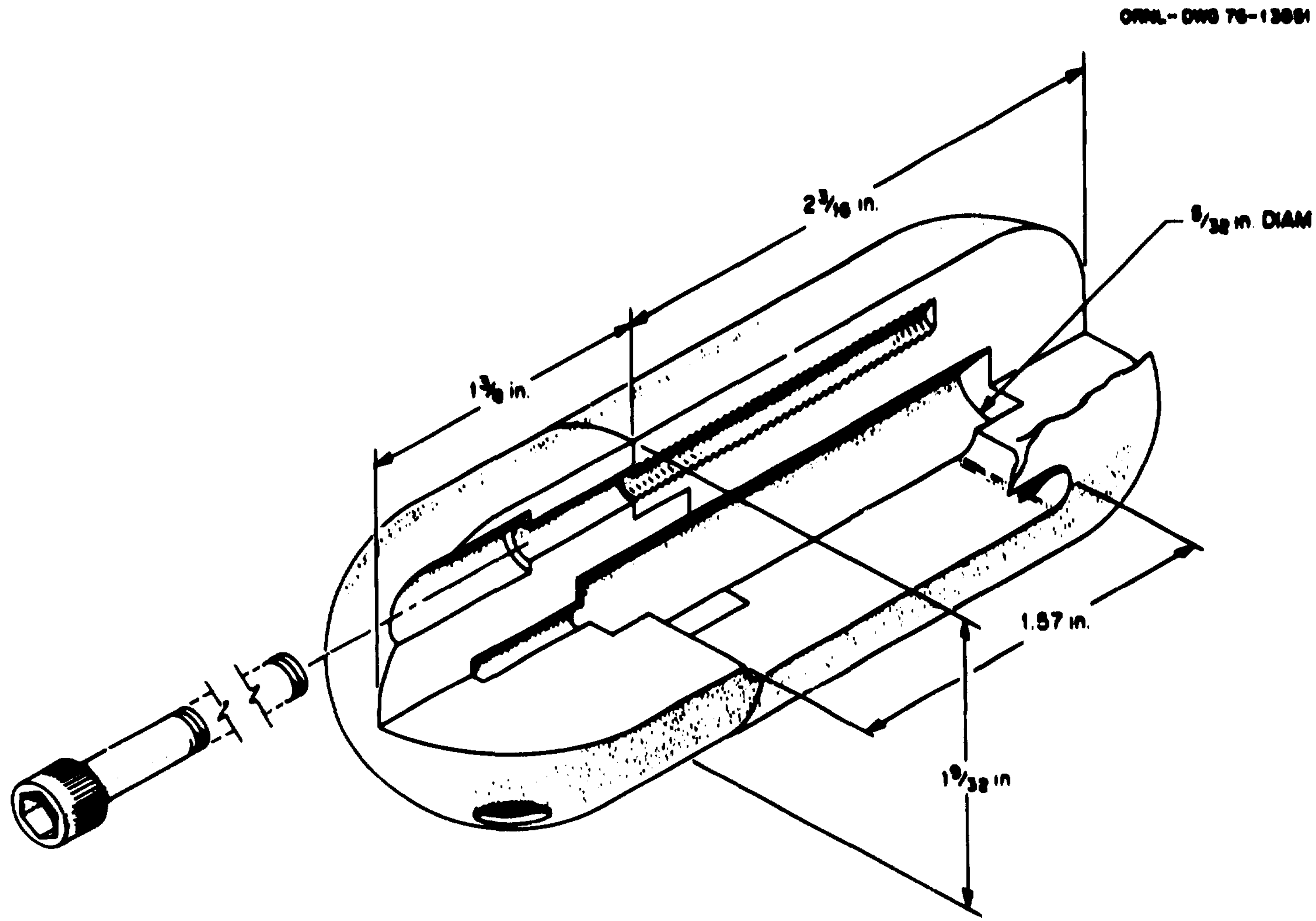

Fig. 4. Krypton Source Collimator

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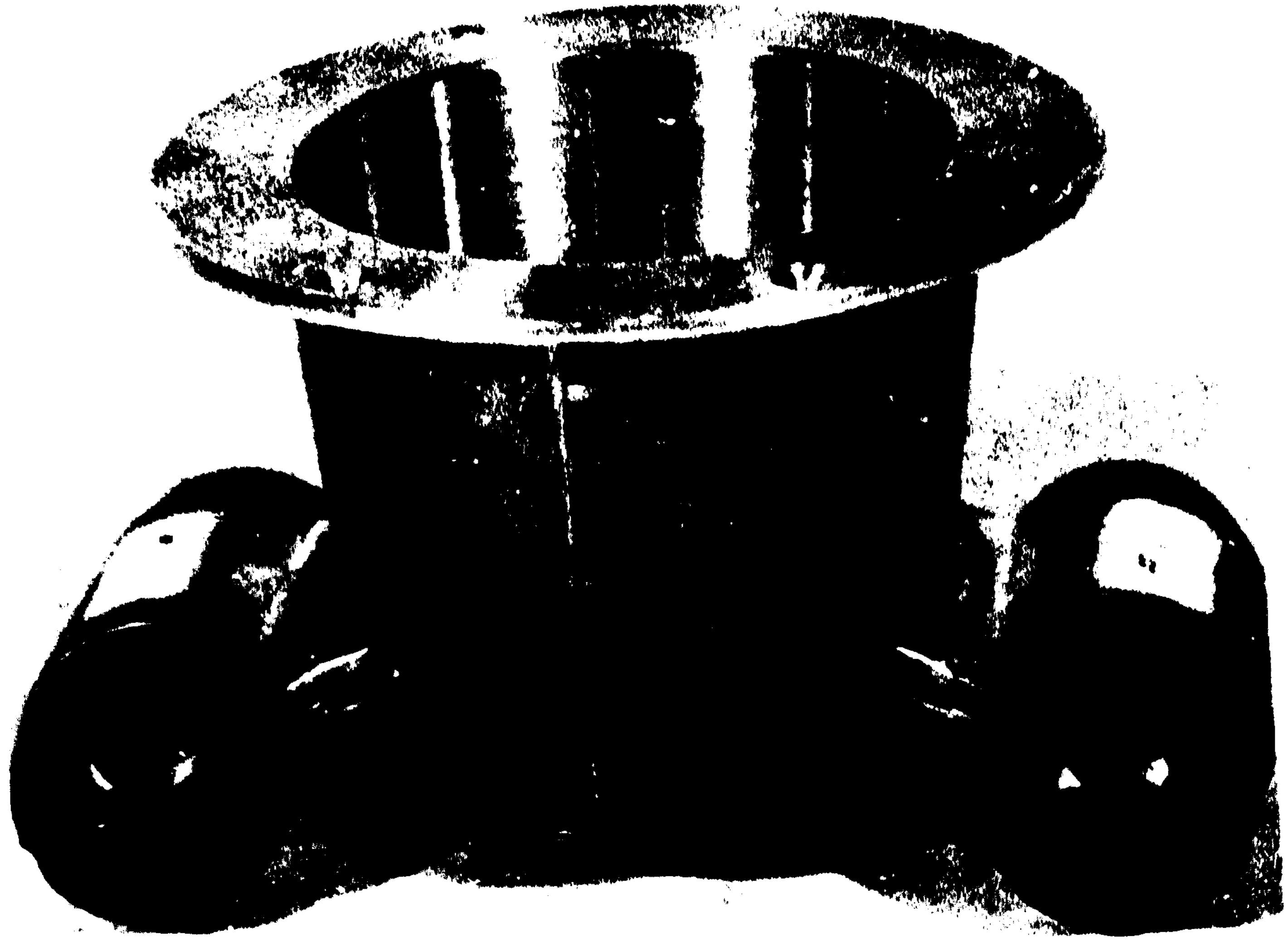

Fig. 5. Coal Dust Analyzer

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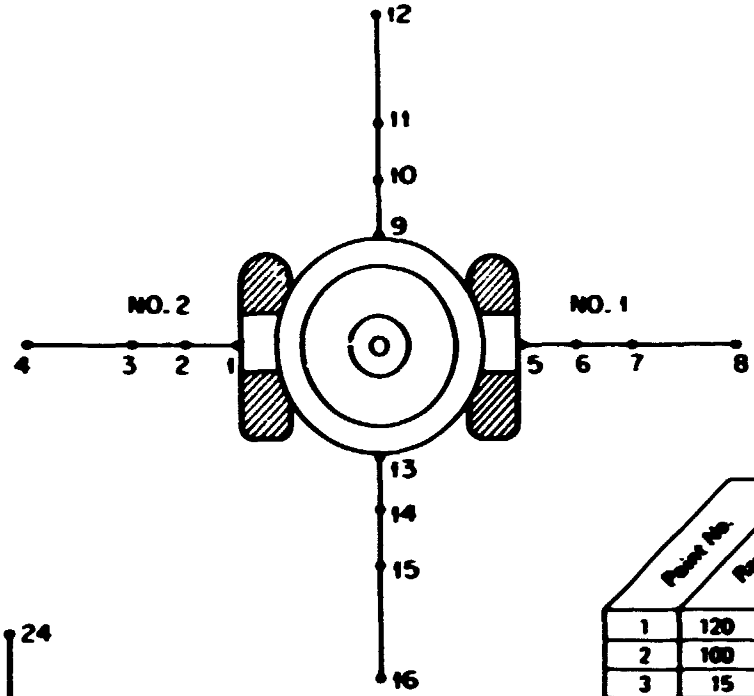

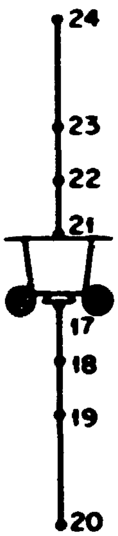

Point No.	Radiation Mr/hr	Distance from source
1	120	Con.
2	100	3 in.
3	15	6 in.
4	4	12 in.
5	50	Con.
6	12	3 in.
7	5	6 in.
8	1.8	12 in.
9	74	Con.
10	9	3 in.
11	4	6 in.
12	3.2	12 in.
13	84	Con.
14	15	3 in.
15	7	6 in.
16	3	12 in.
17	110	Con.
18	30	3 in.
19	15	6 in.
20	5	12 in.
21	50	Con.
22	20	3 in.
23	12	6 in.
24	5	12 in.

Fig. 6. Radiation Dose Rates (outside analyzer)

sources also have high radiation emission of relatively penetrating gamma rays (0.51 MeV) from ^{85}Kr decay. This radiation would not only be hazardous for the operator of the analyzer using present shields but also it severely hampers the measurement of scattered radiation from the sample because of the excessive background count rates. If shielding were increased for operator protection and for the lowering of background count rates, weight of the instrument would probably increase beyond practical limits. It therefore seems unlikely that the gaseous ^{85}Kr source will be useful as a replacement for ^{241}Am sources in the coal dust analyzer. Perhaps other applications for these gaseous ^{85}Kr sources in non-portable configurations where shield weight is not a factor may be found.

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