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INTERIM REPORT #5 - PART I
POSSIBILITIES FOR REDUCING NPR GAMMA DOSE RAT[ES]

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Z PLANT RADIATION STUDY
INTERIM REPORT #5 - PART I
POSSIBILITIES FOR REDUCING NPR GAMMA DOSE RATES

by

H. A. Moulthrop
Radiological Development
Facilities Engineering
Chemical Processing Department

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Z PLANT RADIATION STUDY
INTERIM REPORT #5 - PART I
POSSIBILITIES FOR REDUCING NPR GAMMA DOSE RATES

INTRODUCTION

Previous interim reports (1,2,3) have presented data on radiation aspects of Z Plant processes including isotopic content, neutron energy and flux variations between plutonium compounds, gamma growth rate of daughter products and other factors of influence on personnel exposure rates.

Interim report #4 (4) substantiated the importance of clean hood interiors as a primary approach to maintaining low gamma radiation levels. Subsequent operating experience has emphasized the extreme difficulty of maintaining clean hood interiors.

The resultant high radiation levels near process hoods has led to a need for further definition of present and projected shielding requirements. The high cost of shielding the surface of large hoods together with the high radiation to which hands are still exposed during glove operation has further led to considering the possibility of eliminating hoods wherever feasible with reliance on direct confinement within the process equipment.

The previously unpublished gamma radiation shielding data as presented in Part II(5) of this interim report has been treated to permit its application not only to conventional process hoods, but also to localized shielding of unhooded process equipment.

SUMMARY

Three series of samples consisting of various plutonium compounds, as well as samples of plutonium metal were selected from the Z Plant process stream. The samples representing different process conditions were studied to determine radiation data of importance to present and future radiological design. The report relates data on gamma spectrum as observed with a 256-channel analyzer through various shielding to data on dose rate attenuation of the samples using conventional survey instruments.

The gamma radiation is extrapolated by energy group for application to process material being anticipated from the new production reactor (NPR).

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This Part I of the report presents an analysis of the data for application to present and future processes within the Z Plant. Tabulated unclassified data upon which this Interim Report No. 5 is based is presented in Part II.

Figures IA through IF show for lead and for lead glass the basic relationship between total gamma dose rate and increasing thicknesses of shield. Figures IA and IB present graphically the variation in NPR shielding requirements for the three basic types of Z Plant process material, as categorized by gamma shielding requirements. These are: 1) product having a low fission product content and a relatively low rate of neutron emission; 2) product with high fission product content and a relatively low rate of neutron emission; and 3) product with a high rate of neutron emission from alpha-neutron reaction without regard for fission product content.

These charts also demonstrate the advantage of minimizing fission product content. As may be seen from Figure IA, the difference between low and high fission products in reducing NPR gamma radiation at 4-inches to 1.0 mr/hour is the difference between 0.6 and 1.5-inches of lead.

For Purex production of which the study samples are representative zirconium-niobium is the principal fission product impurity. Ruthenium-rhodium in the nitrate feed was not apparent in subsequent process steps.

Plutonium fluoride which has a high neutron flux from alpha-neutron reaction was found to be by far the most difficult process material in Z Plant to shield for gamma radiation. Measurements of dose rates with routine survey instruments showed twice as much lead to be required to reduce gamma radiation from fluorides than was required for any other plutonium compounds including the metal. An explanation of this behavior is presented in Figure IF. It shows that with fluorides gamma reduction is governed by attenuation of 1.27 mev, and the 2.1 mev components of the fluoride gamma energy spectrum. These two high gamma energies were observed in all fluoride, but in no other samples.

CONCLUSIONS

Analysis of the study data has resulted in the following conclusions:

1. One-inch thickness of lead over ½-inch of steel reduces the gamma radiation dose rate associated with NPR plutonium isotopes to approximately 0.1 mr/hour at 4-inches from the steel equipment walls.

2. For an NPR Z Plant process performed within equipment having localized shielding of 1½-inches of lead over ½-inch of steel gamma radiation penetrating the shield would be limited to that produced either by fission products or by gamma produced as a result of neutron reactions.

3. Even with only negligible gamma from all plutonium isotopes, it would still be possible, within the present acceptability limits for fission products in Z Plant feed, to receive a dose rate of 2 to 3 mr/hour

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through the $1\frac{1}{2}$-inch lead shield at a distance of 4-inches.

4. Ruthenium is appreciably removed between the nitrate and oxalate steps in the Z Plant processes, but zirconium-niobium is not. Consistent and effective removal of Zr-Nb from the product stream within the separations plants will greatly simplify Z Plant shielding and reduce radiation exposure.

5. Even with a product that was relatively low in fission products, the containment of plutonium flroride could, because of gamma radiation produced from reaction by emitted neutrons, result in gamma dose rates of 5 to 10 mr/hour at a distance of 4-inches through a $1\frac{1}{2}$-inch lead shield. (See Fig. IA, Area 3)

6. The high gamma radiation associated with a well shielded plutonium fluoride process can be attributed to 1.27 mev gamma produced by Sodium$_{22}$ when it decays to Neon$_{22}$. The Na$_{22}$ is formed from the fluorine atom when it absorbs an alpha particle and produces a neutron. The 2.1 mev gamma also noted only in the fluoride samples is attributable to neutron captures by hydrogen to form deuterium.

7. For conventional process hoods, lead glass of 6.2 Sp. Gr. will result in the following approximate NPR gamma dose rates, expressed in mr/hour at one foot from the hood wall.

Lead Glass	Process Condition		
	Low	High	High
6.2 Sp. G	F.P.	F.P.	Neutron
1-Inch	1	1-2	2-5
2-Inch	0.1	0.5	0.5-2

8. For nitrate solution of 200 Gr. Pu/l and low fission product content, 1-inch of 6.2 Sp. G. lead glass or $\frac{1}{2}$-inch of lead is required to reduce NPR gamma radiation to 2 mr/hour at 4-inches from a $\frac{1}{4}$-inch tank wall.

9. For fabrication hoods thicknesses of 4.8 Sp. G. lead glass of from 1-inch to 3-inches are required to reduce the gamma radiation to 1.0 mr/hour at 4-inches from the hood wall. Thickness varies with distance of metal from the hood wall. Data is based on shapes considered to be contemporary with NPR processing. For opaque sections of fabrication hoods where shielding is achieved by lead cladding over $\frac{1}{4}$-inch steel walls, NPR gamma dose rates of 1 mr/hour at 4-inches will require a lead thickness of from $\frac{1}{4}$-inch to 1-inch of lead, depending on distance of the source from the hood wall.

10. Reductions in dose rate by as much as 1 mr/hour at 4-inches from a $\frac{1}{4}$-inch thick lead or lead glass surface are obtained by covering the surface with a plastic film only 0.020-inches thick. Thin steel is equally effective. This reduction is approximately 0.2 mr/hour as observed at 12-inches from the lead shielded samples of 400 Gr. Pu, which were used as sources.

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This production of radiation at the outer surface of the lead decreases with intensity of the gamma field penetrating the shield.

11. The multi-channel gamma analyzers now available in process control laboratories are an effective instrument for evaluating relative concentrations of troublesome fission products in a process stream. The ease of analysis through observation of spectrum data has potential for simplified and more effective control of the process as regards high radiation impurities.

12. With additional fission product control of the Z Plant feed material to minimize the presence of zirconium-niobium, and with the elimination of plutonium fluoride as a process material, it is feasible with localized shielding of equipment, to operate an NPR button line with a total gamma radiation field of only 0.1 mr/hour at a distance of 4-inches from the process equipment.

RECOMMENDATIONS

The conclusions derived from data developed and presented in this interim report support the following recommendations concerning the Z Plant process and associated equipment design.

1. The fluoride process should be eliminated or replaced in the Z Plant operation as soon as it is feasible to do so. This action, quite aside from direct neutron radiation exposure reduction, will greatly simplify gamma shielding since, without plutonium fluoride as a process material, 1.27 mev gamma would be eliminated and 2.1 mev gamma would be minimized.

2. Attention should be directed in the separation plants to the specific reduction of zirconium-95 and niobium-95 as identified by the 0.750 mev energy group in the gamma spectrum.

3. Consideration should be given by the separation plants to simplified and more direct control of fission products through greater in-line application of gamma analyzer data. This should include direct comparison of observed 0.400 and 0.750 mev energy peaks as a fundamental specification criteria.

4. Development of the localized shielding and unhooded equipment concept should be expedited to permit the use of 1½-inches lead shield, thus eliminating Pu gamma radiation exposure as a factor in the scheduling of either maintenance or production personnel within the button line operation.

5. To permit gamma field readings of less than 1 mr/hour near fabrication hoods in which work with larger and heavier metal shapes, as currently projected, will be processed, the following minimum shielding thicknesses are recommended:

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a. ½-inch of lead on ½-inch steel walls for opaque surfaces; and

b. 1-inch of 6.2 Sp. G. lead glass over 3/8" lucite for transparent surfaces.

In addition hood frames should be constructed to permit: (1) a cladding of up to one-inch of lead where the source is within six-inches of the hood wall and no use is made of supplemental shields inside the hood; and (2) a window shield of two-inches of 6.2 Sp.G. lead glass for windows where the source may be within six-inches of the window surface.

6. The minimum shielding of ½-inch of lead for opaque surfaces and 1-inch of 6.2 Sp.G. lead glass, as recommended for fabrication hoods, should also be used on existing hoods scheduled for NPR process material, wherever the hood construction permits such supplemental shielding.

7. A suitable lightweight covering for lead and lead glass outer surfaces should be developed for absorbing the secondary electrons originating at the lead surface. The surface should be such as to permit visibility when applied over lead glass. The covering should also provide a corrosion and contamination protective coating. The covering should be of sufficient integrity and intactness to insure reclamation of all lead metal used in process shielding wherever equipment is retired from use.

DISCUSSION

Division of Study into Part I and Part II

A discussion of the data supporting the conclusions, as presented herein, is contained in Part II of this report which is published as a separate and unclassified document. The data for the most part is presented in graphic and tabular forms in eight appendix sections each covering a phase of the study. Appendix I of Part II is also included in Part I. Reference to these Part II graphs of the shielding dose rate relationship, as calculated for Z Plant processes with NPR process material will be helpful in visualizing the summary data and conclusions. Reference to the supporting Appendices I through VIII as contained in Part II is recommended.

Gamma Increase Factors

One point of discussion not contained in Part II for reasons of classification is the derivation of factors used in extrapolating the gamma radiation from that of the samples as used to that expected from plutonium produced in the NPR. This is a function of isotope distribution which in turn is based on the megawatt days per ton in the reactors, MWD/T. Calculations were made in HW-60695 (6) of the factor of increase for each gamma energy group when the plutonium is irradiated to 2,000 MWD instead of the 650 MWD. These factors together with an

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upward approximation of them for the 575 MWD samples are shown below:

Increase Factors per Energy Group for Gamma Dose Rates from Indicated MWD/T to 2,000 MWD/T

MEV	.017	.060	.100	.200	.380	.750	1.27	2.1
575 MWD/T	1.5	7.0	7.0	7.0	2.5	1.0	1.0	1.0
650 MWD/T	1.3	6.2	6.3	6.5	2.3	1.0	1.0	1.0

As shown in the Part II Appendix of this report, gamma photons of 0.500, 0.660, 0.750, 1.27 and 2.1 mev energy were also present in the gamma spectrum of the samples. The 0.500 and 0.600 mev energies are attributed to ruthenium-rhodium and 0.750 mev energy to zirconium-niobium impurities. Since decontamination is a function of the separations plants with no apparent relation to plutonium isotope distribution, it is assumed to be constant with change in MWD/T of the plutonium. An increase factor of 1.0 is therefore used in extrapolating gamma dose rates to 2,000 MWD/T for fission products in NPR plutonium.

The same increase factor of 1.0 has been used for extrapolating the 1.27 and 2.1 mev gamma dose rates. Gamma of these two energies, actually, is likely to increase by a factor of 1.1 for fluorides from NPR plutonium produced at 2,000 MWD/T. This is based on the assumption that the 1.27 mev gamma may result from Na^{22} which is produced from the fluorine atom in proportion to the alpha-emission rate. Since more than 99% of the total neutron flux from plutonium fluoride is attributable to the alpha-neutron reaction, this 1.1 factor also applies to the 2.1 mev gamma from hydrogen capture.

Variation of Dose Rates with Source Strength

Base Source Strength

One of the difficult decisions in calculating shielding thicknesses for a hood is the determination of the source strength to be used as the base. The volume of a hood and the multiplicity of operations and geometrical arrangements possible within it requires some assumptions to be made. Since a finite dose rate is known to exist from the specific samples used in this radiation study, an assumed base source strength for calculating purposes has been taken as a multiple of the dose rate from an average study sample element.

Average Study Sample Elements

The following average sample elements have been used in this interim report.

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Physical Nature of the Plutonium Source	Actual Plutonium Compounds	Type of Container	Weight of Plutonium in Source
Aqueous Solution	Nitrate	Plastic liter bottle	200 grams in one liter of solution
Dry or nearly dry powder	Oxalate, oxide, fluoride	1 pint cardboard carton	400 Grams
Buttons, castings	Metal	Plastic bag	1,000 Grams

In the calculations, as presented in the Part II Appendix, the base source strength as selected for shielding determination is three times that of the study sample element as used for all forms of plutonium except the metal.

Factors for Base Source Strengths

For the metal a factor of five is used, but this applies to the larger and heavier metal shapes which are currently projected for fabrication.

The factor of three, as used for solutions and powders, is applied to both conventional process hoods and to confinement within unhooded process equipment. In Table IVC of Appendix IV in Part II, it is shown that the maximum dose rate at the center of a tube, 39-inches in length, would be 4 times the dose rate of the average study sample element. A factor of 3 rather than 4 was used to allow for the uncorrected effect of self-shielding by adjacent source material.

The same factor of three was also used in increasing the study sample dose rate to obtain base hood dose rate for solutions and powders. This factor was selected after considering the collective effect of: (1) probability for dispersement within a hood or vessel; (2) limitations for concentration imposed by critical mass considerations; (3) the hardening effect of self-shielding within bulk sources; and (4) the decreased rate of dose rate reduction with distance as the source area within a hood increases. Although arbitrary the factor of three is considered to be a reasonable increase over the dose rate of the average study sample for calculating base source strength.

Factors for Upper Limits of Source Strength

In Figure IA of Part I a probable upper limit for source strength is shown by dotted curves. This upper limit is taken as twice the base limit and

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recognizes the possibility that any elongated tube containing the plutonium powdered or liquid compound may be completely full rather than only half full as assumed in the calculations.

It was shown in Interim Report #4 of the Z Plant Radiation Study that radioactive residues in a hood have a direct bearing on dose rate. In Figure IB a factor of three over the base dose rate has been selected for assigning a maximum dose rate resulting from radioactive residues.

Distance Factors

For some fabrication and inspection hoods where large concentrated metal shapes are the primary sources, distance of the source from the wall of hood becomes a major consideration in determining body dose rates to personnel. For this reason Figures ID and IE which give dose rates for plutonium metal include values for distances ranging from 4-inches to 20-inches. The 4-inch distance applies for sources close to the hood wall and the 20-inch distance for those at arm's length within the hood.

Importance of Reducing Fission Product Content

The relative difficulty among the various study samples in shielding gamma radiation is a function not only of the compound, as in the case of the fluoride, and of sample size because of self-shielding, but also it is a function of the fission product content of the sample.

Data is presented in Fig. IA for gamma radiation from the plutonium isotopes only of the two samples highest in fission products; namely, the oxide and fluoride of Series III. This data compares very closely with the total gamma from the sample lowest in fission products; namely, the oxalate of Series III.

As may be seen from Fig. IA, a ten-fold difference in NPR gamma radiation through 1-inch of lead exists between the oxalate of Series I and the oxide of Series III. The oxalate representing feed that had one of the lowest fission product contents ever received in Z Plant from Purex shows 0.2 mr/hour at 4-inches through one-inch of lead; whereas, the oxide of Series III representing the highest acceptable fission product from Purex shows 2.0 mr/hour through the one-inch of lead.

A maximum effort in reducing the fission products in Z Plant feed is thus capable of reducing shielded Z Plant gamma dose rates by a factor of 10.

Penetrating Gamma From Plutonium Fluoride

In Figure IF the relationship of dose rate shielding studies, as presented in Appendix VIII of Part II is compared with gamma spectrum attenuation through shielding, as presented in Appendix III of Part II. As shown, all

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plutonium compounds other than the fluoride of those studied fall between .380 mev as a lower limit of attenuation and 0.750 as an upper limit. With low Zr-Nb content the .380 mev gamma attenuation curve is approached as a basis for shielding requirement. As may be seen, the fission product content, as represented by Zr-Nb, governs in all cases with the one exception of the fluoride.

In the case of plutonium fluoride, it becomes evident that the effective gamma energy is not 0.750 mev. Rather, as shown in Figure IF, attenuation of the fluoride lies between that of the 1.27 mev and 2.1 mev gamma energies. These were observed with the multi-channel gamma analyzer as occurring in the fluoride samples. As may be seen from the data presented in Figure IIA, Part II of this report, the 1.27 and the 2.1 mev energies were not observed in any of the other plutonium samples that were studied.

Importance of Eliminating the Fluoride Process to Reduce Gamma

As shown in Figure IA, the gamma from fluoride of Series I will read 7 mr/hour at 4-inches through one-inch of lead, as compared to 2.5 mr/hour from the oxide of Series III. This increase by three in penetrating gamma radiation exists even though the oxide has ten times more fission products in it than the fluoride. (See Table IIA of Part II).

Although the primary advantage in eliminating plutonium fluoride as a process material has been and still is the major reduction in neutron radiation, it is now evident that its elimination is very much in order also as a means of reducing requirements for gamma shielding.

Homer Moulthrop
Radiological Development
Facilities Engineering
Chemical Processing Department

HA Moulthrop

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2. HW-55463, "Z Plant Radiation Study - Special Sampling Program - Interim Report No. 2," HA Moulthrop, March 27, 1958 (Secret).

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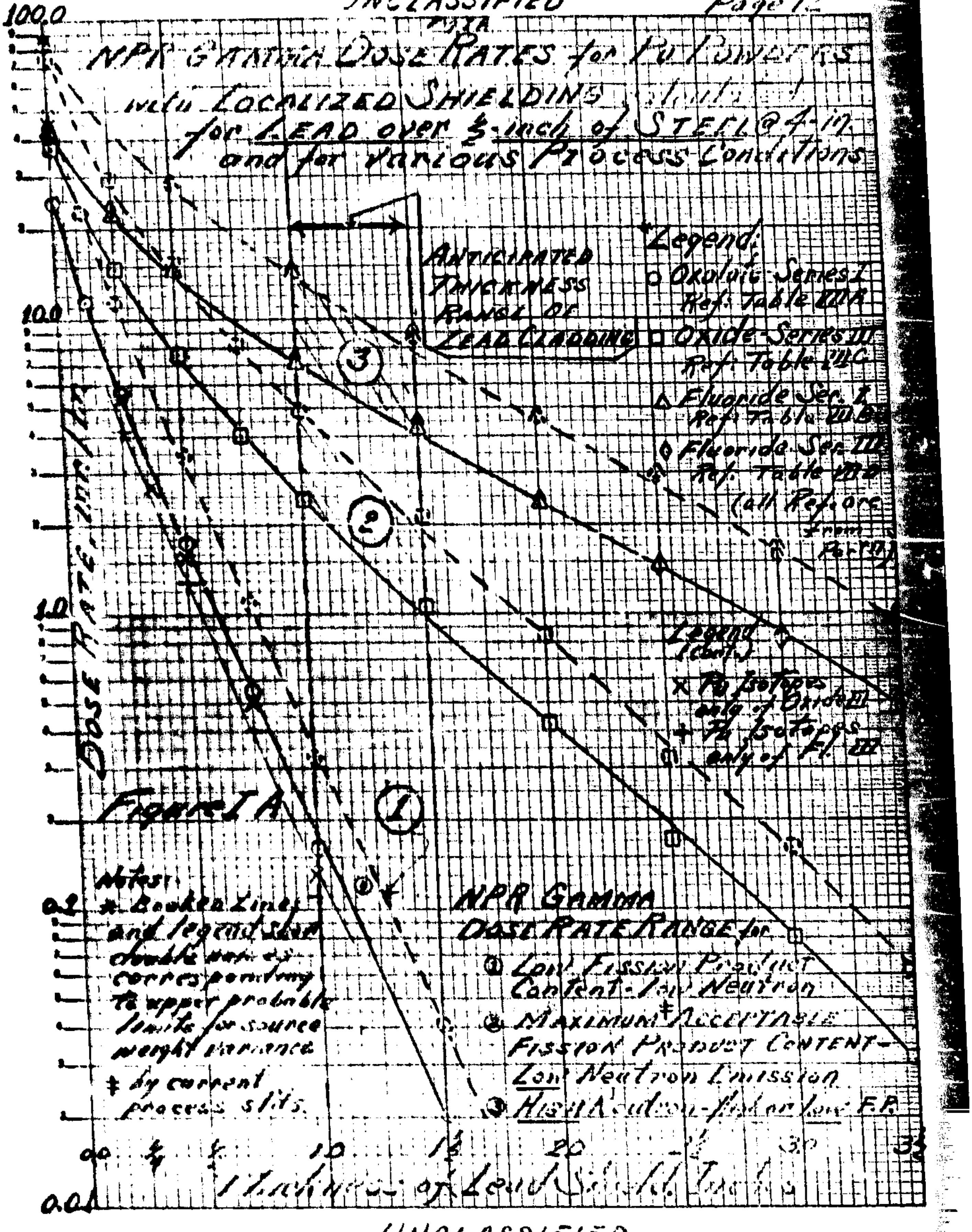
NPR GAMMA DOSE RATES for Pu POWDERS
with LOCALIZED SHIELDING
for LEAD over ½-inch of STEEL @ 4-in.
and for Various Process Conditions
ANTICIPATED THICKNESS RANGE OF LEAD CLADDING
Legend:
○ Oxalate Series I Ref. Table IIIA
□ Oxide Series III Ref. Table IIIC
△ Fluoride Ser. I Ref. Table IIIB
◇ Fluoride Ser. III Ref. Table IIID
(all Ref. are from ...)
Legend (Cont.)
× Pu Isotopes only of Oxide III
+ Pu Isotopes only of Fl. III
DOSE RATE, mr/hr
Figure IA
Notes:
* Broken lines and legend show double values corresponding to upper probable limits for source weight variance
‡ by current process specs.
NPR GAMMA DOSE RATE RANGE for
① Low Fission Product Content - Low Neutron
② MAXIMUM‡ ACCEPTABLE FISSION PRODUCT CONTENT - Low Neutron Emission
③ High Neutron - ... F.P.
100.0
10.0
1.0
0.1
0.01
00
¼
½
10
1½
20
30
Thickness of Lead Shield

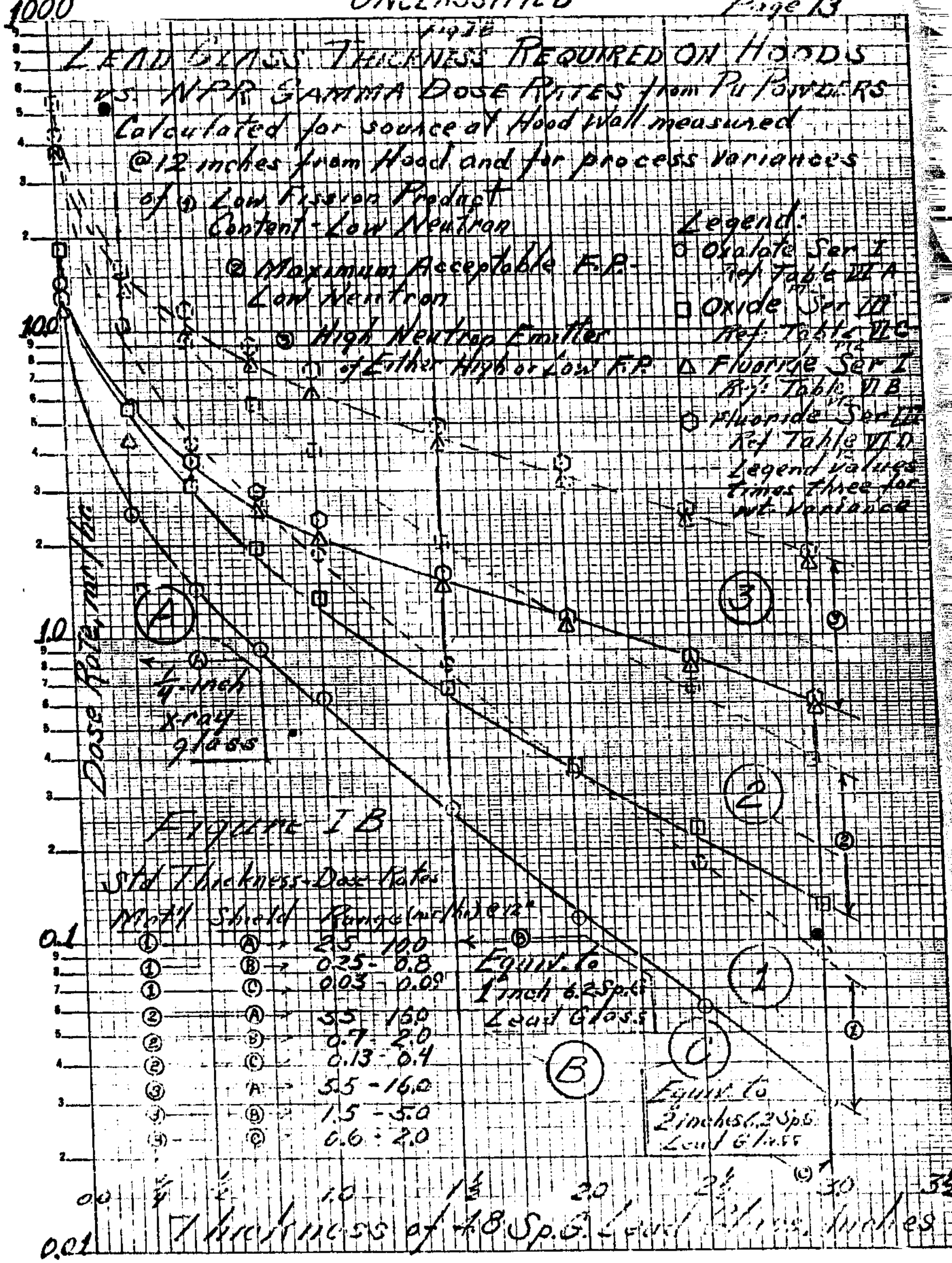
Fig IB
LEAD GLASS THICKNESS REQUIRED ON HOODS
vs. NPR GAMMA DOSE RATES from Pu POWDERS
Calculated for source at Hood Wall measured
@12 inches from Hood and for process Variances
of (1) Low Fission Product
Content - Low Neutron
(2) Maximum Acceptable F.P. - Low Neutron
(3) High Neutron Emitter of Either High or Low F.P.
Legend:
○ Oxalate Ser I Ref Table VI A
□ Oxide Ser III Ref Table VI C
△ Fluoride Ser I Ref Table VI B
⬡ Fluoride Ser II Ref Table VI D
Legend values times three for wt. variance
Dose Rate, mr/hr
1/4 inch X-ray glass
Figure IB
Std Thickness-Dose Rates
Mat'l Shield Range (mr/hr) @12"
① Ⓐ 2.5 - 10.0
① Ⓑ 0.25 - 0.8
① Ⓒ 0.03 - 0.09
② Ⓐ 5.5 - 15.0
② Ⓑ 0.7 - 2.0
② Ⓒ 0.13 - 0.4
③ Ⓐ 5.5 - 16.0
③ Ⓑ 1.5 - 5.0
③ Ⓒ 0.6 - 2.0
Equiv. to 1 inch 6.2 Sp.G. Lead Glass
Equiv. to 2 inches 6.2 Sp.G. Lead Glass
Thickness of 4.8 Sp.G. Lead Glass, Inches

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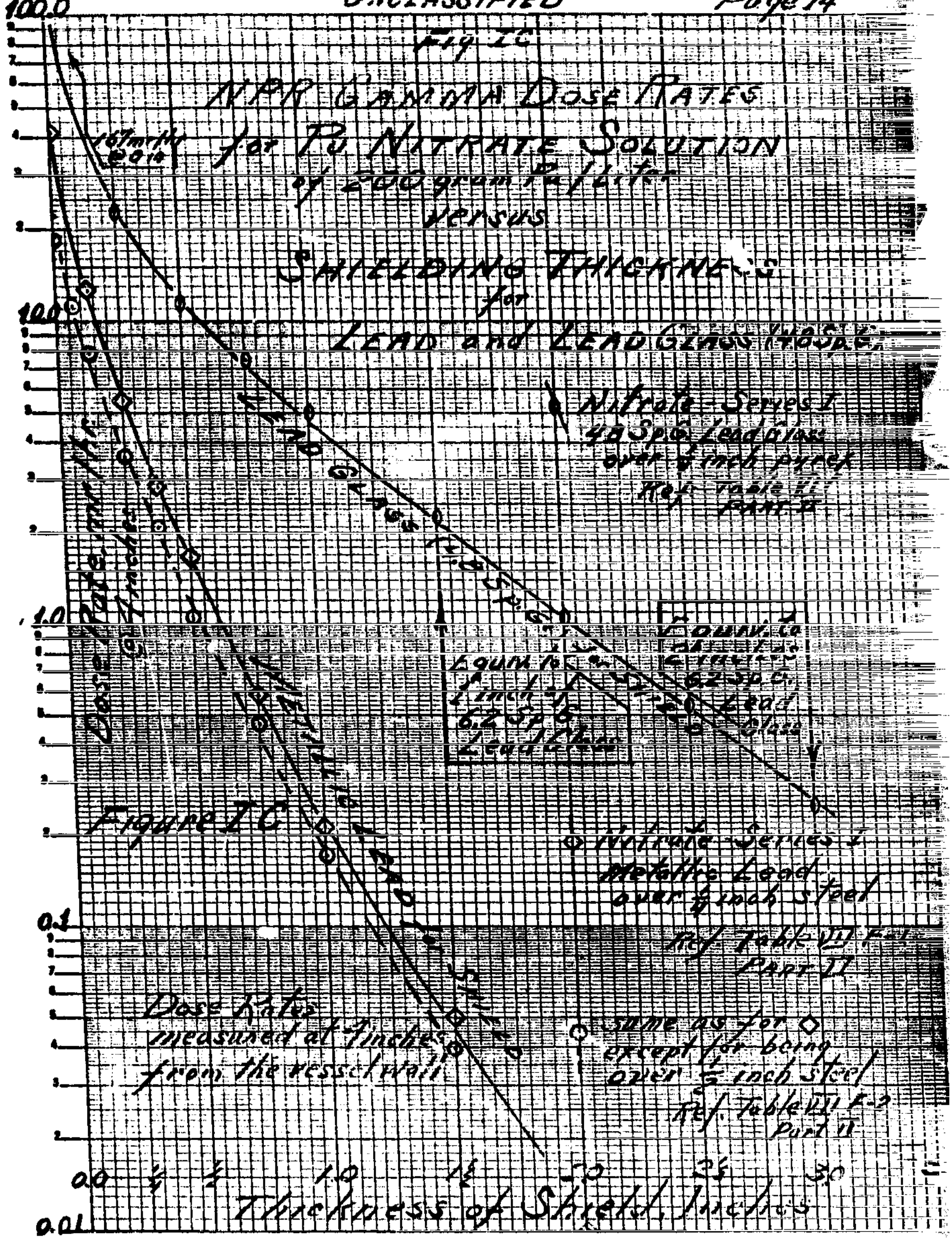

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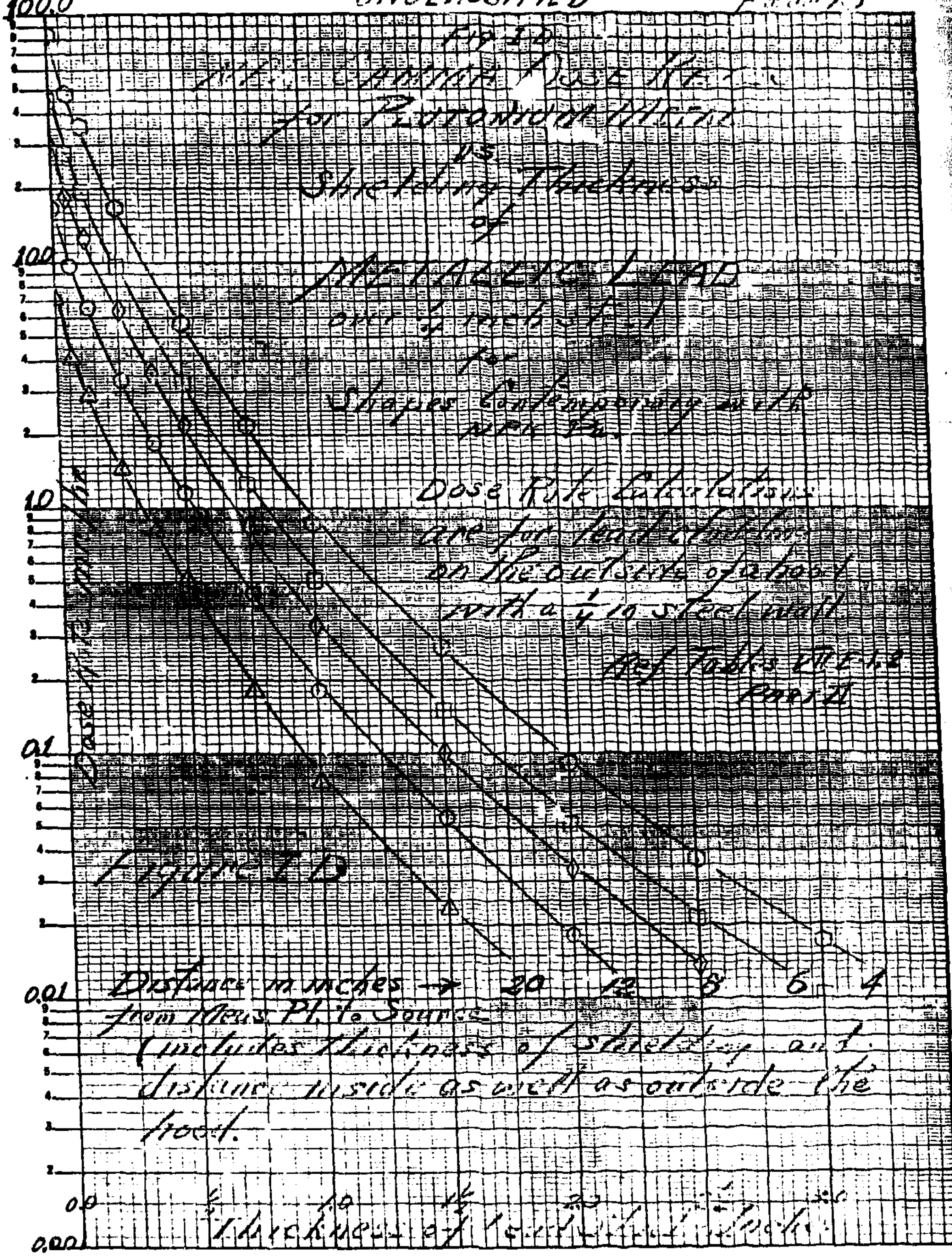
vs
Shielding Thickness
of
Dose Rate Calculations
on the outside of a hood
with a 1/4 in steel wall
Figure I D
Distance in inches → 20 12 8 6 4
from Meas. Pt. to Source
(includes thickness of shielding and distance inside as well as outside the hood.
100.0
10
0.1
0.01

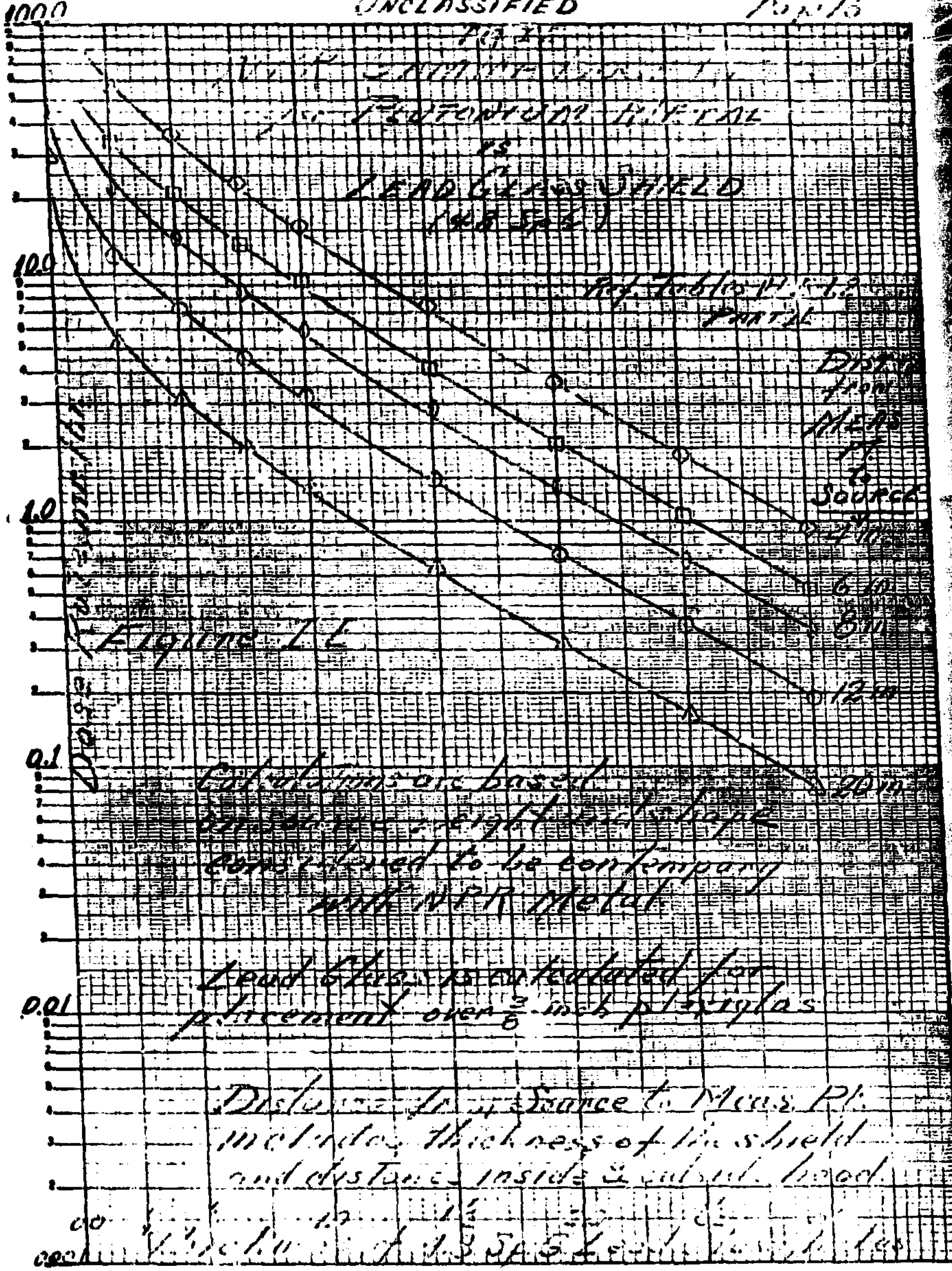
1000
100
1.0
0.1
0.01
Ref Table VI
Dist from Meas Pt to Source
12 in
20 in
Figure II
Calculations are based on ...
Lead Glass is calculated for placement over 3/8 inch plexiglas
Distance from Source to Meas Pt includes thickness of the shield and distance inside hood.

Figure I F
ATTENUATION COMPARISON
in LEAD
for
Measured Gamma Dose Rates of Samples
vs.
Percent Photon Transmission by Energy