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MND-3607-173-1

SNAP 19 RADIATION MEASUREMENTS REPORT

APPENDIX C

MARTIN MARIETTA

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NOVEMBER 1967
MND-3607-173-1

CONTRACT AT(30-1)-3607

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FOREWORD

This report is submitted by Martin Marietta Corporation, Nuclear Division, in support of United States Atomic Energy Commission Contract AT(30-1)-3607. This classified volume contains Appendix C to the unclassified SNAP 19 Radiation Measurements Report (MND-3607-173). This, in conjunction with the unclassified volume, constitutes the final report for Item 1(c) of the Experimental Safety Program for SNAP 19, dated January 1966.

NEUTRON SOURCE CHARACTERIZATION

Investigations and measurement of neutron emission spectra from isotope-fueled heat sources, no matter how accurate and detailed, do not provide the radiation analyst with a basic starting point, the neutron source spectra. A complete analysis should include a detailed description of the magnitude, energy dependence and production modes of the neutrons born within the fuel material. The objective of the study reported in this section is such a description. Results are derived in part from the analysis of a SNAP 19 capsule and subsystem neutron emission spectra measurements. The capsule measurements were performed by Mound Laboratory on System No. 6 capsules. The subsystem measurements are those performed by Martin Marietta and reported in Section V of the unclassified part of this report (MND-3607-173). Attention is directed to the influence on neutron source spectra of specific isotopes of oxygen in the plutonium oxide fuel so as to define the benefit which may be obtained in reduction of neutron radiation by selective isotope reductions.

Neutron production in plutonium and plutonium dioxide fuel forms is mainly due to (α, n) reactions and fission of Pu-238. Mound Laboratory (Ref. C-1) reports a neutron count rate for small samples of the metal close to that of the Pu-238 spontaneous fission release rate of 3.5×10^3 n/sec-gm of Pu-238 of Segre (Ref. C-2). Apparently Pu-238 metal, with impurities, air and cladding material absent, would emit only fission neutrons. Use of the oxide fuel form introduces neutrons from the O-17 (α, n) Ne-20 and O-18 (α, n) Ne-21 reactions. Neutrons from the O-17 (α, n) Ne-20 reaction are insignificant since O-17 is present in only very minute quantities (isotopic abundance of 0.04% or less) and the (α, n) cross section for O-17 over the alpha energy range of interest is approximately half that of O-18 (Ref. C-3). However, O-18 is present in natural oxygen with an abundance of 0.2% and would emit at least 10 times the number of neutrons per unit volume as the O-17 reaction.

Recent measurements by Mound Laboratory (Ref. C-1) on small samples of the oxide fuel form depleted in O-18 indicate a linear reduction of neutron production with decreasing O-18 content. A maximum of 1.5×10^4 n/sec-gm of Pu-238 was observed with the material abundance of 0.2% O-18. For small samples, no energy degradation within the sample may be safely assumed or, equivalently, the absolute magnitude of the neutron source is equal to the total emission rate. Hence, these experimental data indicate a neutron source for small samples prepared using natural oxygen consisting of approximately 1.15×10^4 n/sec-gm of Pu-238 from the O-18 (α, n) Ne-21 reaction and 0.35×10^4 n/sec-gm of Pu-238 from spontaneous fission of Pu-238 for a total of 1.5×10^4 n/sec-gm of Pu-238.

The results of this experiment must be contrasted to measurements of total emission rates for oxide fuel systems (Refs. C-4, C-5 and C-6) which vary from 2.3×10^4 to 2.9×10^4 n/sec-gm Pu-238. Point kernel calculations over the actual geometrical source configuration (SNAP 19 capsule) using neutron removal cross sections indicate a neutron degradation or loss within the source of 25%. That is, the approximate actual neutron source is 1.33 times the measured emission rate or 3.33×10^4 n/sec-gm Pu-238.

A study was undertaken with the objective of determining the absolute magnitude and detailed energy spectra of the individual components of the spectra for the oxide fuel system. This study combined theoretical calculations and analysis of emission spectra for the fuel capsules and subsystem. The first step was to compute the spectrum of neutrons from O-18 (α, n) Ne-21 reaction according to the method of

Hess (Ref. C-7), using an energy dependent cross section derived from measured values (Ref. C-3) and an excitation scheme in which, after neutron emission from the compound nucleus Ne-22, 55% of the residual Ne-21 nucleus is left in the ground state, 35% in the first excited state (0.35 Mev) and 10% in the second excited state (1.73 Mev) (Ref. C-8). The computed spectrum exhibits a peak at approximately 3.0 Mev which compares favorably with measured neutron spectra. In addition, the oxygen calculations show an integrated neutron source of 1.28×10^4 n/sec-gm of Pu-238 which compares very favorably with the value derived from the small sample experiment. Secondly, analysis of measured spectra indicates a much higher magnitude of fission neutrons than that component due to spontaneous Pu-238 fission.

The end result of the study was the postulation of spectral components which appear consistent with both the theoretical and experimental evidence. In magnitude, this spectrum consists of the following:

2.0×10^4 n/sec-gm Pu-238 from O-18 (α, n) Ne-21 and other (α, n) reactions in impurities

1.33×10^4 n/sec-gm Pu-238 from spontaneous and induced fission of Pu-238.

Inherent in these numbers is the 33% difference between production rates and emission rates; i.e., the corresponding emission rates for the heat source would be 1.5×10^4 n/sec-gm Pu-238 for (α, n) reactions and 1.0×10^4 n/sec-gm Pu-238 for fission neutrons. It is significant to note that the postulation of a nearly threefold increase in fission neutrons requires verification and that if this increase is, indeed, the case, the reduction of O-18 during fuel production would not only reduce the O-18 component but would significantly reduce the induced fission component.

This energy-dependent production spectrum is presented in Table C-1. The spectrum of all (α, n) neutrons was assumed to be that calculated for O-18 and the U-235 fission spectrum shape was assumed for Pu-238 fission neutrons. Note that the values in the table are point values in terms of lethargy (n/sec-gm Pu-238) or, equivalently, energy per unit energy (Mev/sec-Mev-gm Pu-238). For comparison, the source spectrum is shown in Fig. C-1 with a typical SNAP 19 capsule measured emission flux spectrum. Figure C-1 presents the spectrum in units of n/sec-Mev-gm Pu-238. The conversion from lethargy units to energy spectrum for each point i is I/E_i; i.e., the values of Table C-1 have been divided by the appropriate energy in Mev. The emission flux has corresponding flux units per unit energy but the magnitude is relative.

The data in Fig. C-1 demonstrate the expected differences between the neutron source and emission spectra. The removal theory prediction of an effective 25% loss of neutrons within the source is seen as an increase in neutrons of lower energy (below 3 Mev) and a corresponding loss of neutrons above this energy. This loss is not a true absorption, but a combination of absorption and scattering of neutrons to lower energies. This combination results in a 25% reduction in neutron dose rates (the basis for removal cross sections). Scattering and absorption accounts for the observed shift of the peak of 2.8 Mev (source spectrum) to 2.3 Mev (emission spectrum). Figure C-1 also shows the expected near-agreement in shape of the two spectra at higher neutron energies (greater than 40 Mev).

A preliminary verification of the postulated, induced fission neutron source was accomplished by performing an integral over the computed space averaged neutron flux spectrum for a small spherical source of plutonium dioxide (radius = 4.4 cm) and the energy-dependent fast fission cross section of Pu-238 (Ref. C-9) to determine

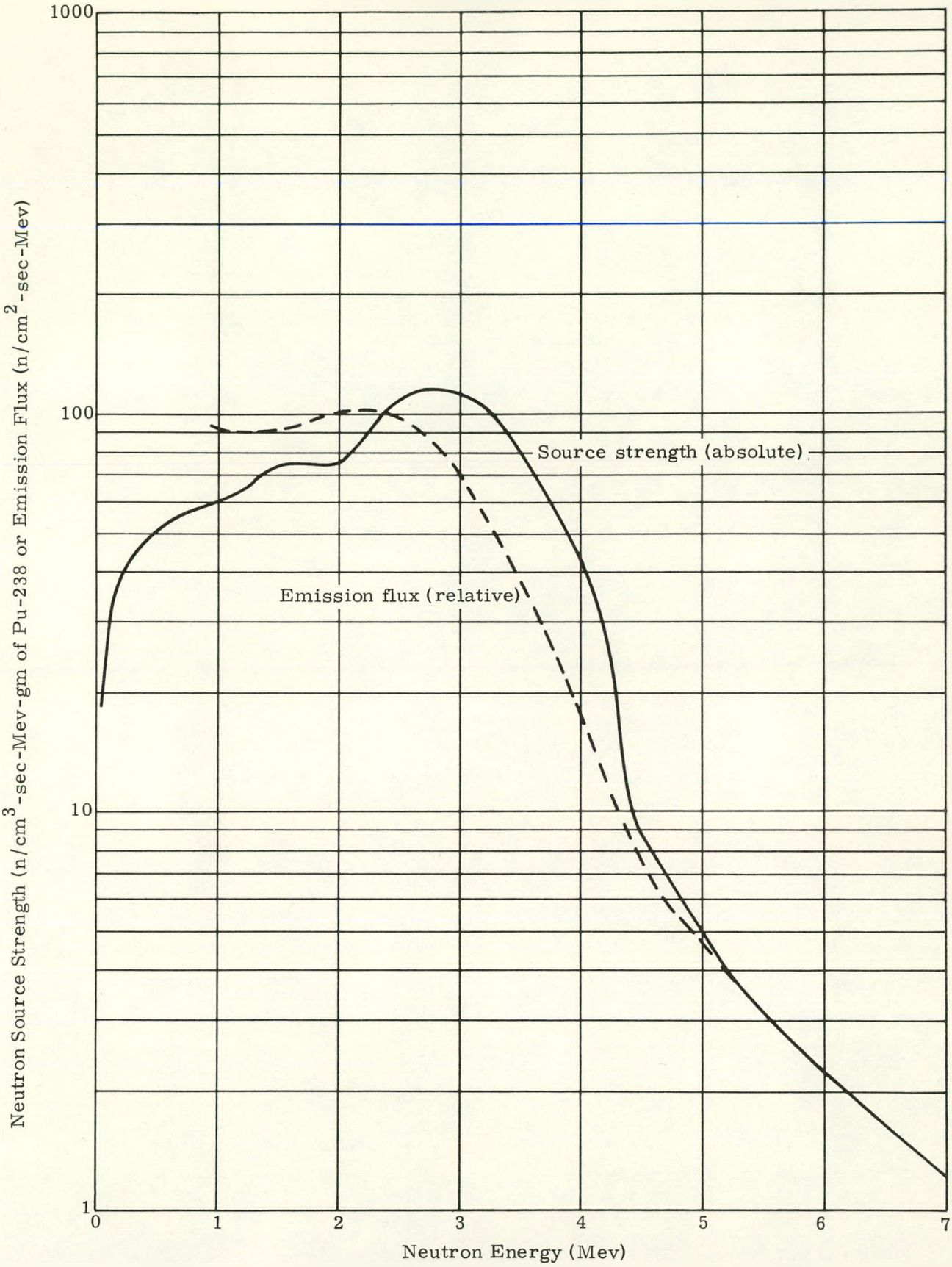

Fig. C-1. Comparison of Neutron Source Spectrum and Measured Emission Flux Spectrum

the magnitude of the induced fissions. The result of the integration was an average of 1.94×10^4 fissions/cm^3-sec over the volume of the source. Assuming 2.3 emitted neutrons per fission, the resultant neutron source from induced fission is 4.46 n/cm^3-sec or, equivalently, 8.44×10^3 n/sec-gm Pu-238. This result is within 15% of the postulated induced fission source of 9.80×10^3 n/sec-gm of Pu-238 (i.e., 1.33×10^4 n/sec-gm of Pu-238 minus the spontaneous fission component of 0.35×10^4 n/sec-gm of Pu-238). The real significance of this result is that since most of the neutrons which produce this induced fission component are born in O-18 (α, n) reactions, the production of fuel depleted in O-18 could possibly result in a nearly six-fold reduction in the magnitude of the neutron source. Since gammas are emitted in the fission process, a reduction in gamma source strengths would also be effected.

TABLE C-1

Neutron Source Spectra

Point Index, i	Energy (Mev)	Lethargy[1] U	A (α, n) Neutrons (n/sec-gm of Pu-238)	B Fission Neutrons	Composite Spectrum A + B
1	14.09	0	0	5.50×10^0	5.50×10^0
2	10.97	0.25	0	5.49×10^1	5.49×10^1
3	8.55	0.50	0	2.97×10^2	2.97×10^2
4	6.66	0.75	0	9.76×10^2	9.76×10^2
5	5.18	1.00	0	2.05×10^3	2.05×10^3
6	4.46	1.15	0.93×10^3	3.12×10^3	4.05×10^3
7	4.04	1.25	1.19×10^4	0.37×10^4	1.56×10^4
8	3.14	1.50	2.85×10^4	0.52×10^4	3.37×10^4
9	2.45	1.75	2.10×10^4	0.60×10^4	2.70×10^4
10	1.91	2.00	0.80×10^4	0.62×10^4	1.42×10^4
11	1.49	2.25	0.56×10^4	0.57×10^4	1.13×10^4
12	1.16	2.50	2.43×10^3	4.99×10^3	7.42×10^3
13	0.900	2.75	1.32×10^3	4.09×10^3	5.41×10^3
14	0.702	3.00	0.71×10^3	3.23×10^3	3.94×10^3
15	0.546	3.25	0.39×10^3	2.47×10^3	2.86×10^3
16	0.331	3.75	0.18×10^3	1.35×10^3	1.53×10^3
17	0.201	4.25	0.80×10^2	6.94×10^2	7.74×10^2
18	0.122	4.75	0.31×10^2	3.44×10^2	3.75×10^2
19	0.0449	5.75	0	8.10×10^1	8.10×10^2

[1] Lethargy $U = \ln (E_o/E)$ where $E_o = 14.09$ Mev.

This after-the-fact result remains to be verified by the more elaborate neutron multiplication transport calculation which utilizes fission cross sections and iterates to a converged source. The initial source for this type calculation would be the known (α, n) and spontaneous fission components. Indeed, parametric calculations of this type could be run to investigate the effect of varying source geometries. Results, combined with investigations of (α, n) prediction in impurities and quantitative measurements of gamma emission for small samples depleted in O-18 (similar to those performed for neutrons), would furnish better understanding of the mechanisms of radiation production. This knowledge could be used in design and production of a generator heat source geometry and fuel specification which produces a *minimum of* emitted radiation.

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