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APPLICATION OF A150-PLASTIC EQUIVALENT GASES IN
MICRODOSIMETRIC MEASUREMENTS[‡]

by

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ABSTRACT

Neutron dosimetry measurements with ionization chambers, for the most part, employ tissue equivalent plastic-walled cavities (Shonka A150) filled with either air or a methane-base "tissue-like" gas. The atomic composition of TE-gas and A150 plastic are not matched and are quite dissimilar from muscle. Awschalom and Attix (1980) have partially resolved the problem by formulating a novel A150-plastic equivalent gas. This establishes a homogeneous wall-gas cavity dosimeter for neutron measurements and confines the necessary corrections to the application of kerma ratios. In this report, we present measurements of applications of two A150-plastic equivalent gases in a low pressure spherical proportional counter. Gas gains and alpha-particle resolutions were determined. For these A150-mixtures as well as a methane-based TE-gas and an Ar-CO₂ mixture, we report measurements of event size distributions from exposure to a beam of 14.8 MeV neutrons.

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INTRODUCTION

A continuing difficulty with the dosimetry of fast neutrons as deduced from cavity chamber measurements results from the mismatch in atomic composition between the cavity wall and the filling gas. In particular, the dose contribution due to neutron interactions with the filling gas varies considerably with the neutron energy and even at 15 MeV is ~15% (Dennis, 1980). This gas contribution is cavity size (or more precisely, cavity thickness) dependent. Tissue dose determinations are achieved after correcting for stopping power differences between the gas and wall. Contributions due to gas effects, e.g., direct field interactions and W-value differences, are ignored.

Of course the ideal situation would be an identical match between wall, gas and tissue. Even for this case, differences between gas and solid-phase stopping powers as well as variations in W-values between recoil electrons and heavier charged particles must still be corrected for. Al50-plastic (Shonka, 1950) ionization chambers filled with a methane-based tissue-equivalent gas (Failla, 1959) are widely used for neutron dosimetry. For this combination, neither the wall nor gas in fact match tissue, differing significantly in their oxygen and carbon components. TE-gas and Al50-plastic do not match each other with respect to oxygen and carbon content. In lieu of an attempt to provide a complete tissue equivalent system, Awschalom and Attix (1980) proposed several gas mixtures which are at least Al50 plastic equivalent. Tissue dose determinations would then reduce to correcting for the variation in neutron kerma between tissue and Al50 plastic.

In this report, we present results of an investigation of the applicability of one of the Al50 mixtures proposed by Awschalom and Attix as well as an additional recently developed mixture for miniature proportional counter use.

MATERIALS AND METHODS

Table I summarizes the atomic composition of ICRU muscle (1962), Al50 plastic (Smathers, 1977), TE-gas, Al50 (Mix 1) (Awschalom and Attix, 1980) and Al50 (Mix 5)). The close match between Al50-plastic and mixes 1 and 5 is apparent. In Table II the various constituent gas component weight percentages for TE-gas and Mixes 1 and 5 are listed. It should be noted that these values are determined by an analysis of the actual gas mixtures employed. Thus, the derived weight fraction by element (Table I) differs slightly from the nominal values recommended in the literature for TE-gas (Failla, 1956). This has a negligible effect upon the stopping power and kerma values (Table III). For Mix 5, the oxygen content is somewhat higher than in Al50-plastic, but the stopping powers and kerma values for various neutron and photon beams are essentially identical to Al50 plastic (e.g. Table III).

In a previous report (DeLuca, Attix, Pearson and Awschalom, 1980) we presented measurements of the behavior of Mix 1 in ionization chambers of various volumes and geometries. An estimate of W for exposure to 14.8 MeV neutrons and ^{60}Co photons was given in that report. During these efforts it was noted that Mix 1 exhibited greater recombination losses than either air or TE gas. A possible explanation of this effect would be increased electronegativity due quite probably to the Freon-14 component in Mix 1 (Table II). A degree of electronegativity could severely inhibit the application of this gas for proportional counter purposes. Subsequently, a Freon-free mixture, Mix 5, was developed. Freon was eliminated and CO was replaced by CO_2 . The new mixture retains a close match in atomic composition, stopping powers and interaction coefficients with Al50-plastic (Tables I, III).

The revised mixture, Mix 5, the original mixture, Mix 1, and the methane-based TE-gas were compared for low pressure miniature proportional counter applications. The counter is of commercial design (Far West Technology), constructed of Al50-plastic, and contains a ^{244}Cm α -particle calibration source. Standard nuclear instrumentation was employed for data acquisition. Following analog-to-digital

conversion, proportional counter pulses were transferred to a DEC 11/34 computer for storage and display. Final analysis was performed on a DEC VAX 780 computer. Details of this acquisition and storage procedures can be found in USDOE Report No. DOE/EV/01105-272.

Relative gas gain and resolution were studied as a function of filling gas pressure and applied bias. The internal ^{244}Cm source was employed during these experiments. The results are presented below. Beyond this work, the various gas mixtures were employed in the measurement of the neutron dose in a 14.8 MeV beam of neutrons. This fast neutron source has been previously described in some detail (DOE/EV/01105-272,1980). Results of these experiments are discussed below.

During these measurements, the A150 gases were thoroughly remixed before filling the counter. A heater placed at the bottom of the gas storage vessels was used to raise the gas temperature to $40^{\circ}\text{C} - 50^{\circ}\text{C}$ for 1 hour before filling the counter. The gas handling manifold and counter were evacuated to a pressure of 6 mPa for 1 hour before filling. Several flushings of the chamber and manifold preceded final pressurization.

RESULTS

Gas Gain and Resolution

Proportional counter resolution and gas gain were measured as a function of applied bias at filling pressures of 4, 8 and 16 kPa. At these pressures, the corresponding cavity target thicknesses were in the range of $72-360 \mu\text{g}/\text{cm}^2$. The relative gain values were normalized to the α -particle energy loss at a given gas thickness and type mixture. The voltage range studied had as an upper bound that voltage just below spontaneous discharge, and as a lower bound that voltage when the signal-to-noise ratio was ~ 2 .

The gas gains for either A150 mixture and TE-gas were quite similar. Figures 1 and 2 show results for A150 (Mix 5) and TE-gas, respectively.

Campion (1971) has proposed that the gain of proportional counters of this type can be expressed as:

$$\frac{\ln G}{P} = \frac{A V/P}{B \ln(b/a)} \left[\exp\left[\frac{-aBP}{V} \ln(b/a)\right] - \exp\left[\frac{-bBP}{V} \ln(b/a)\right] \right], \quad (1)$$

where A and B are constants depending upon gas mixture, V is the applied bias, in volts, P is the filling gas pressure in Torr (1 Torr=0.133 kPa), 'a' is the anode radius in cm, 'b' is the cathode radius in cm, and G is the relative gain. Previous applications of this expression to proportional counter data have yielded inconsistent results from laboratory to laboratory (Campion, 1980; Eichel and Booz, 1976; Herskind and Junen, 1976). However for a specific geometry and experimental configuration, this expression permits comparison of gas gain for different mixtures over a considerable range of applied bias and filling pressure. Figures 3 and 4 depict the curves resulting from fitting equ. (1) to gas gain data for Mix 5 and TE-gas. The relative gas gain for TE-gas was under all conditions greater than Mix 5. Gain measurements for Mix 1 (not shown) yield results essentially identical to Mix 5. For any gas mixture, the gain decreased with increasing pressure.

Alpha-particle resolution measurements for Mix 5 and TE-gas are shown in Figs. 5 and 6, respectively. The resolution values are deduced from the FWHM of the α -particle event distributions. No corrections for the statistical fluctuations of the energy loss process in a thin absorber were made, nor was the finite thickness of the α -particle calibration source accounted for. The measured resolutions for any gas showed a similar dependence upon bias and pressure. At 4kPa, resolutions were typically ~30% decreasing to ~20% at the higher pressures (8 kPa and 16 kPa). Resolution at any pressure deteriorated at low bias and reached a broad maximum between 400 and 600 V.

Absorbed Dose Determinations

Tissue equivalent proportional counters have infrequently been employed to measure absolute neutron dose (see e.g. Stinchcomb, 1980, Weaver, 1977). As these devices employ physically small chambers operated at low pressures, the difficulties a wall-gas mismatch impose upon the validity of Bragg-Gray Cavity Theory are significantly reduced. Energy deposition events are recorded on an event-by-event basis. Any gas-mixture dependent interaction differences are readily discernable. We have measured event spectra for each gas mixture in a collimated beam of 14.8 MeV neutrons. For absorbed dose comparisons an Al50-plastic ionization chamber and neutron insensitive GM counter were operated as a paired dosimeter system under experimental conditions identical to the proportional counter measurements. These dosimeter measurements were used to unfold the inherent ~3% photon dose component in the neutron beam. Proportional counter event spectra data were taken with filling gases of Al50 Mix 1, Al50 Mix 5, TE-gas and Ar-CO₂. The ArCO₂ mixture consists of 94.3% Ar and 5.79% CO₂ by weight. This mixture is frequently employed in our graphite proportional counter because of its very low inherent neutron sensitivity.

Photon events in the proportional counter event spectra are resolved by using a ⁶⁰Co "calibration spectrum" of known dose in conjunction with the unfolded photon dose from the paired dosimeters. The resulting "neutron-only" event spectrum, therefore, does rely upon the photon dose deduced from the paired dosimeters. As the inherent photon dose component in the neutron beam is only ~3%, only a negligible uncertainty results from this spectrum stripping process.

The effect, if any, of a wall-gas mismatch upon the proportional counter data can be observed by comparing the dose distributions vs. event size for different gas mixtures. To assist this comparison, spectra for each gas were normalized to the stopping power of Al50-plastic. For either Al50 mixture and TE-gas, the resultant spectra were identical. However, for ArCO₂ gas, there is a distinct shift in the location in event size of the recoil protons relative to

heavier charged particles (Fig. 7). Recalling that "event spectra" are in fact ionization yield spectra, gas-dependent variations in yield due to charged particle velocity dependence in the differential W-values might be anticipated. Undoubtedly, the principal effect is due to the difference in the stopping power velocity dependence between Ar-CO₂ and the other very similar mixtures. The importance of averaging the wall-gas stopping power ratio over the complete recoil spectrum is evident for the case of a mismatched wall-gas interface.

The results of the absorbed dose measurements are summarized in Table IV. Dose values have been normalized to a common neutron fluence via a separate monitor ionization chamber. Dose values determined from the proportional counter show no observable dependence upon counting gas. The proportional counter dose for any gas was systematically greater than the ionization-chamber- deduced dose values. Whether this represents a true variance or systematic bias is difficult to ascertain. For example, we have naively assumed that the "absolute" dose per event calibration deduced from the calibration α -particle source applies to all measured events. Bichsel (1974) and Booz (1980) have discussed the distortion in measured spectra resulting from the variation in W-values between the α -particle source and the actual secondary charged particle spectrum. Reported measurements of W for TE-gas give a value of 30.91 eV/ip for the calibration α -particles, while that value for 3 MeV protons is 30.33 eV/ip (Kohrig and Collvett, 1978). We estimate the effect of the W-value variation to be ~2%, albeit in the "correct" direction. A further difficulty concerns the actual volume of the proportional counter. In this case, we have used the manufacturers value for the cavity dimension, 1.27 cm. Finally, the effective energy of the α -particle source enters through the value of the stopping power for the filling gas. Of these effects, the volume correction and effective energy of the calibration source are the most suspect.

CONCLUSIONS

Two gas mixtures which are Al50-plastic equivalent were tested in a miniature proportional counter. The measured gas gains and α -particle resolutions were found to be comparable to those of the methane-based TE-gas mixture frequently employed in ionization chamber measurements. No untoward gas handling procedures were found to be necessary. The 1.7% by weight component of Fluorine in Mix 1, which was eliminated in Mix 5, did not seriously degrade the gas gain or resolution. The charge collection losses observed for Mix 1 during ionization chamber work (DeLuca et.al., 1980) were most probably due to a reduction in mean electron velocity rather than electronegativity induced by the presence of CF_4 in the mixture.

Beyond these efforts, we have compared microdosimetric event spectra taken with Mix 1 and Mix 5 to that acquired with TE-gas as the filling mixture. In each case the spectra were identical. However, when the filling gas was an Ar- CO_2 mixture, significant differences in the event spectra were observed. As the charged particle spectrum from the wall is independent of filling gas to an excellent approximation, the spectrum in the cavity can be assumed to be identical for any gas at these low pressures. The observed differences are due to variations in ion yield per particle resulting from velocity dependent effects in the various stopping powers and W-values.

Finally absorbed tissue doses from exposure to a beam of collimated 14.8 MeV neutrons were determined with the proportional counter and compared to those deduced from ionization chamber measurements. Dose values for filling gases of TE-gas, Al50 Mix 1 and Mix 5 and Ar- CO_2 varied less than $\pm 3\%$ but were uniformly greater than the ionization chamber results ($\sim 14\%$).

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FIGURE CAPTIONS

- Fig. 1 Relative gas gain for Al50 (Mix 5) gas at several pressures plotted vs. applied bias.
- Fig. 2 Relative gas gain for TE-gas at several pressures plotted vs. applied bias.
- Fig. 3 Relative gas gain for Al50 (Mix 5) gas plotted vs. pressure normalized bias. The curve represents a fit of equ. (1) to the data. Values of A and B result from the fit.
- Fig. 4 Relative gas gain for TE-Methane gas plotted vs. pressure normalized bias. The curve represents a fit of equ. (1) to the data. Values of A and B result from the fit.
- Fig. 5 Resolution for Al50 (Mix 5) gas at several pressures plotted vs. applied bias. Resolution values are determined at full-width-at-half-maximum (FWHM) for the calibration α -particle.
- Fig. 6 Resolution for TE-Methane gas at several pressures plotted vs. applied bias. Resolution values are determined at full-width-at-half-maximum (FWHM) for the calibration α -particle.
- Fig. 7 Plots of the event-size-weighted fractional dose as a function of event-size for exposure to 14.8 MeV neutrons. Curves are shown for Al50 (Mix 5) gas and Ar-CO₂ gas. Data are normalized to the stopping power of Al50-plastic.

TABLE I

WEIGHT PERCENTAGES BY ELEMENT

Element	Muscle ¹	A150-Plastic ²	TE-Gas ³	A150 (Mix 1) ³	A150 (Mix 5) ³
H	10.2	10.2	9.97	10.2	10.2
C	12.3	76.8	45.10	76.8	76.8
N	3.5	3.6	3.89	3.6	3.6
O	72.9	5.9	41.04	5.9	9.0
F	---	1.7	---	1.7	---
Ar	---	---	---	1.8	0.4
Ca	---	1.8	---	---	---
other	1.1	---	---	---	---

1. ICRU Report 10b(1962)

2. Smathers, et.al. (1977)

3. Analysed gas mixtures employed in this work.

TABLE II

WEIGHT PERCENTAGES BY COMPONENT*

Component Gas	TE-Gas	A150 (Mix 1)	A150 (Mix 5)
Methane, CH ₄	39.70	---	---
Ethylene, C ₂ H ₄	---	44.5	41.5
Propadiene, C ₃ H ₄	---	37.8	42.1
Nitrogen, N ₂	3.89	3.6	3.6
Carbon Monoxide, CO	---	10.3	---
Carbon Dioxide, CO ₂	56.40	---	12.4
Freon-14, CF ₄	---	2.0	---
Argon, Ar	---	1.8	0.4

*Gases mixtures and analysis obtained from Mathison Gas Products, Inc.

TABLE III

PHYSICAL DATA FOR A150-PLASTIC, TE-GAS,
A150 MIX 1 AND A150 MIX 2

	A150 Plastic	TE-Gas	A150 (Mix 1)	A150 (Mix 2)
Density(Mg/cm ³)	1126	1.149	1.442	1.518
Sp(MeVcm ² /g)	121.021	119.18	120.88	121.43
S _e (MeVcm ² /g)	2.375	2.370	2.374	2.380
μ _{en} /ρ(cm ² /g)	.0293	.0293	.0293	0.0294
k(rad.cm ²)	.704*10 ⁻⁸	.681*10 ⁻⁸	.704*10 ⁻⁸	.704*10 ⁻⁸

*Density values are for STP conditions. Stopping powers are evaluated at 3 MeV proton energy and 300 keV electron energy using the tabulations of Anderson and Ziegler (1977) and Berger and Seltzer (1964,1966) for neutron and photon exposure, respectively. Mass energy absorption coefficients (μ_{en}/ρ) are taken from the data of Hubble (1969) and evaluated at 1.25 MeV. Kerma values are interpolated from the data of Caswell (1980) and evaluated at 14.8 MeV neutron energy.

TABLE IV

COMPARISON OF NEUTRON DOSES DEDUCED FROM ION CHAMBER (IC),
AND PROPORTIONAL COUNTER (PC) MEASUREMENTS

Gas	D ⁺ (IC)	D ⁺ (PC)	PC/IC
TE-Gas	8.95	10.15	1.13
A150 (Mix 1)	8.95	9.98	1.11
A150 (Mix 5)	8.95	10.52	1.17
Ar-CO ₂	8.95	10.30	1.15
	8.95±0.18	10.24±0.23	<1.14±0.03>

+Tissue dose per monitor chamber nC.

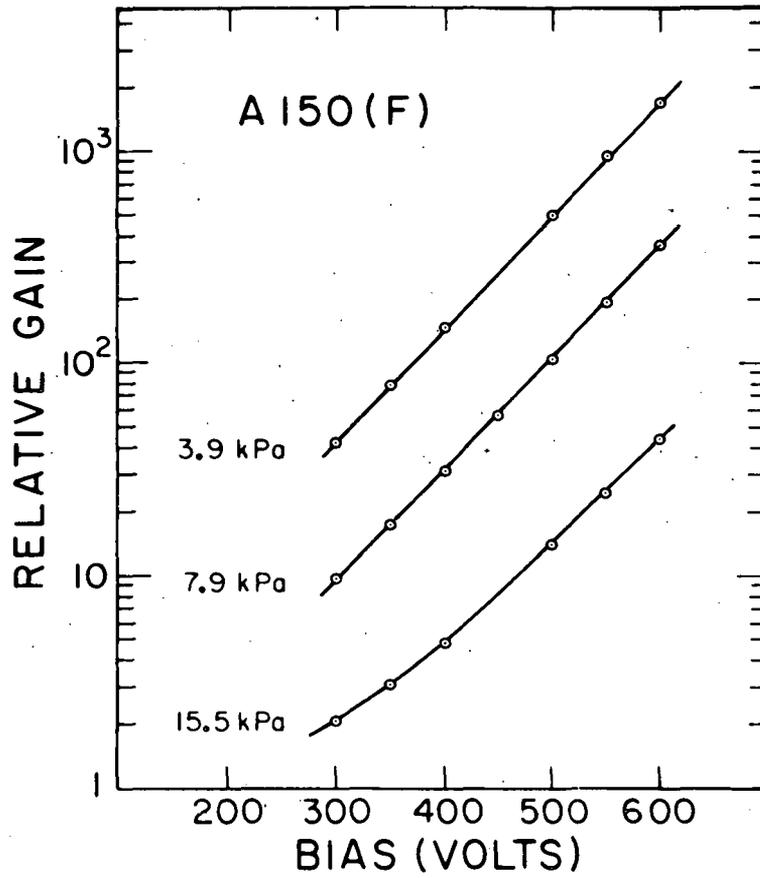


Fig. 1 Relative gas gain for A150 (Mix 5) gas at several pressures plotted vs. applied bias.

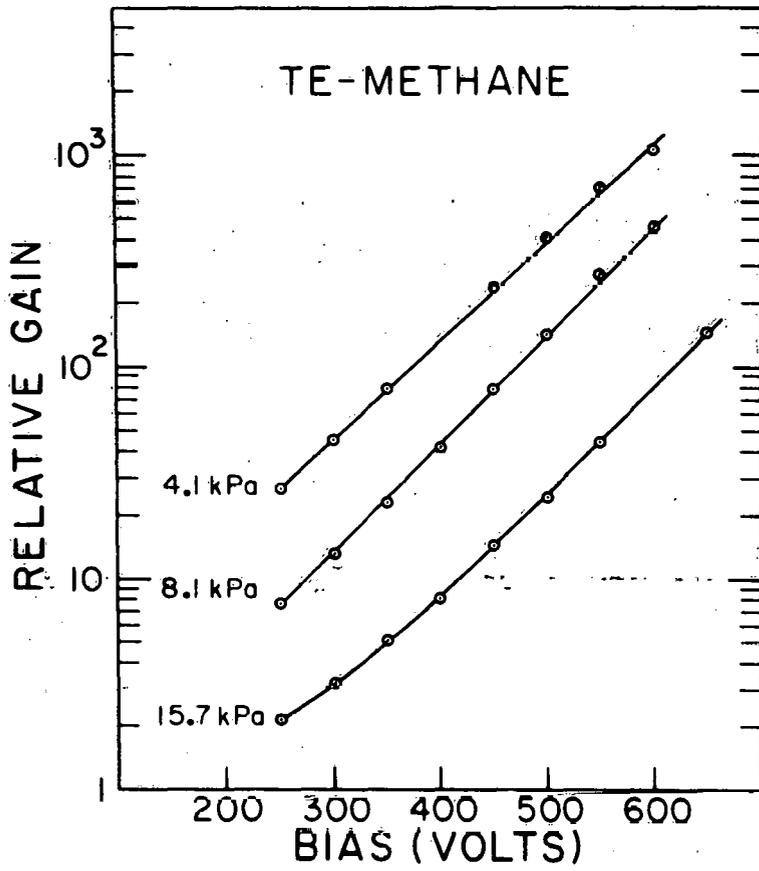


Fig. 2 Relative gas gain for TE-gas at several pressures plotted vs. applied bias.

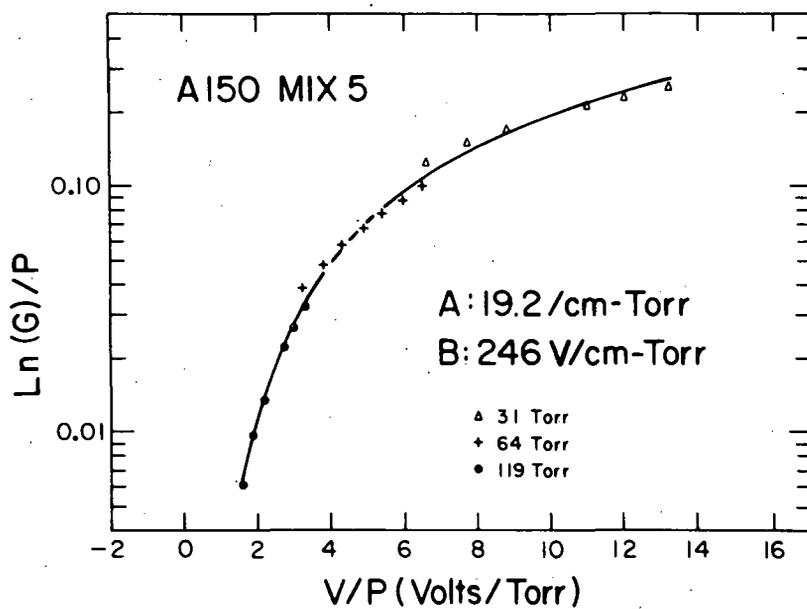


Fig. 3 Relative gas gain for A150 (Mix 5) gas plotted vs. pressure normalized bias. The curve represents a fit of equ. (1) to the data. Values of A and B result from the fit.

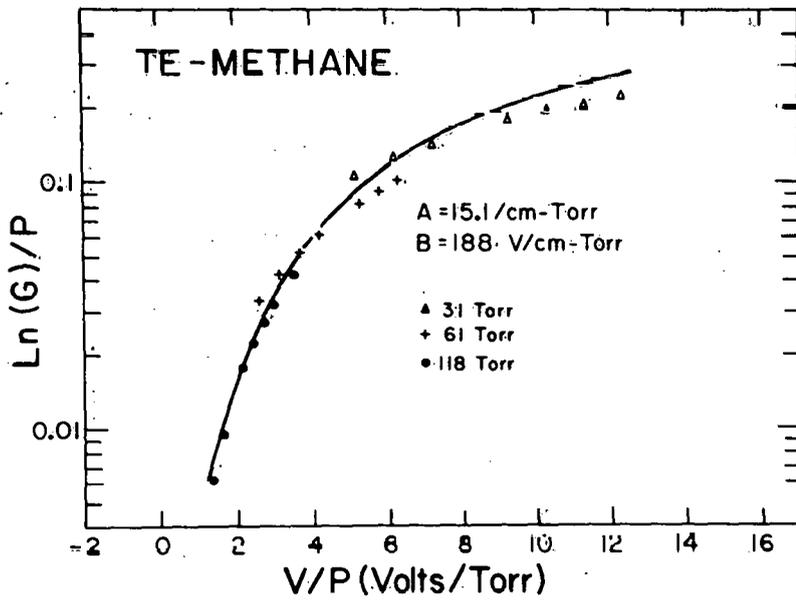


Fig. 4 Relative gas gain for TE-Methane gas plotted vs. pressure normalized bias. The curve represents a fit of equ. (1) to the data. Values of A and B result from the fit.

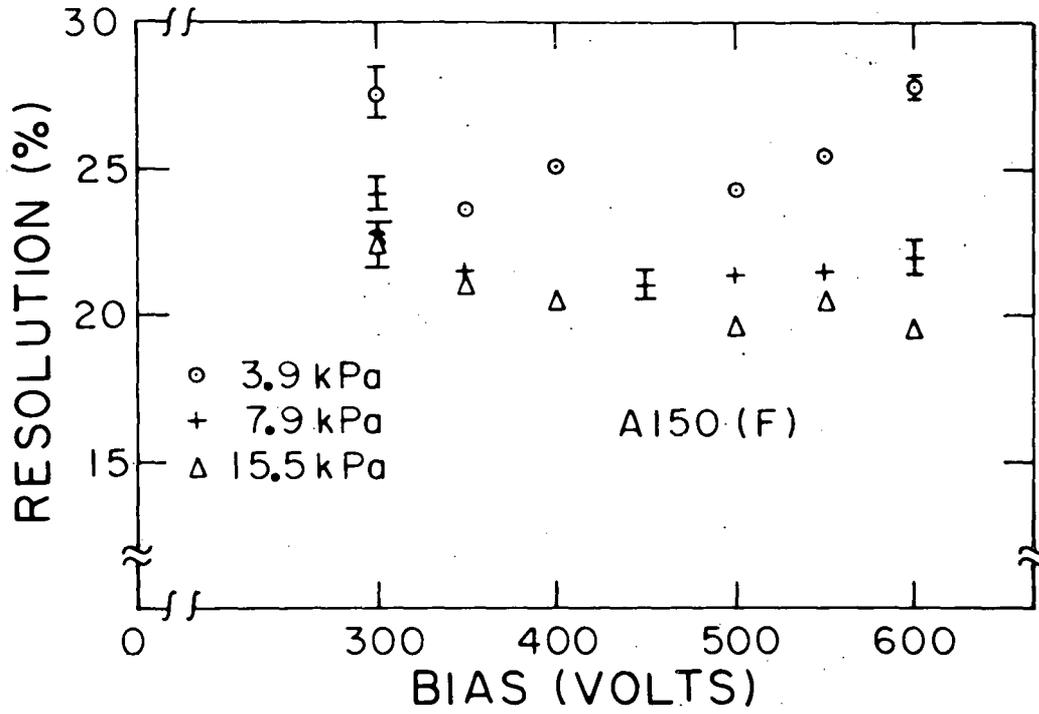


Fig. 5 Resolution for A150 (Mix 5) gas at several pressures plotted vs. applied bias. Resolution values are determined at full-width-at-half-maximum (FWHM) for the calibration α -particle.

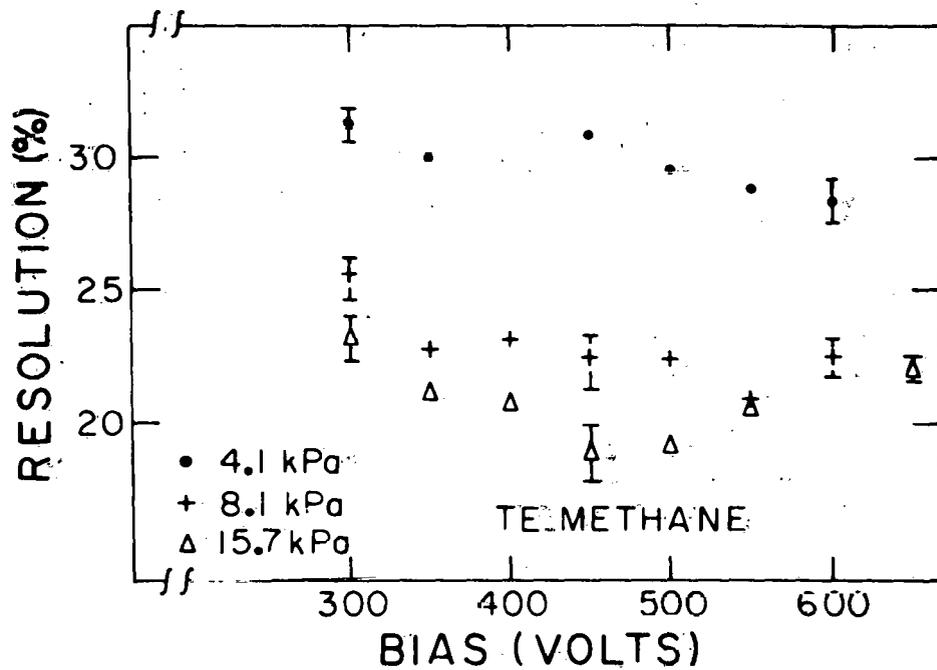


Fig. 6 Resolution for TE-Methane gas at several pressures plotted vs. applied bias. Resolution values are determined at full-width-at-half-maximum (FWHM) for the calibration α -particle.

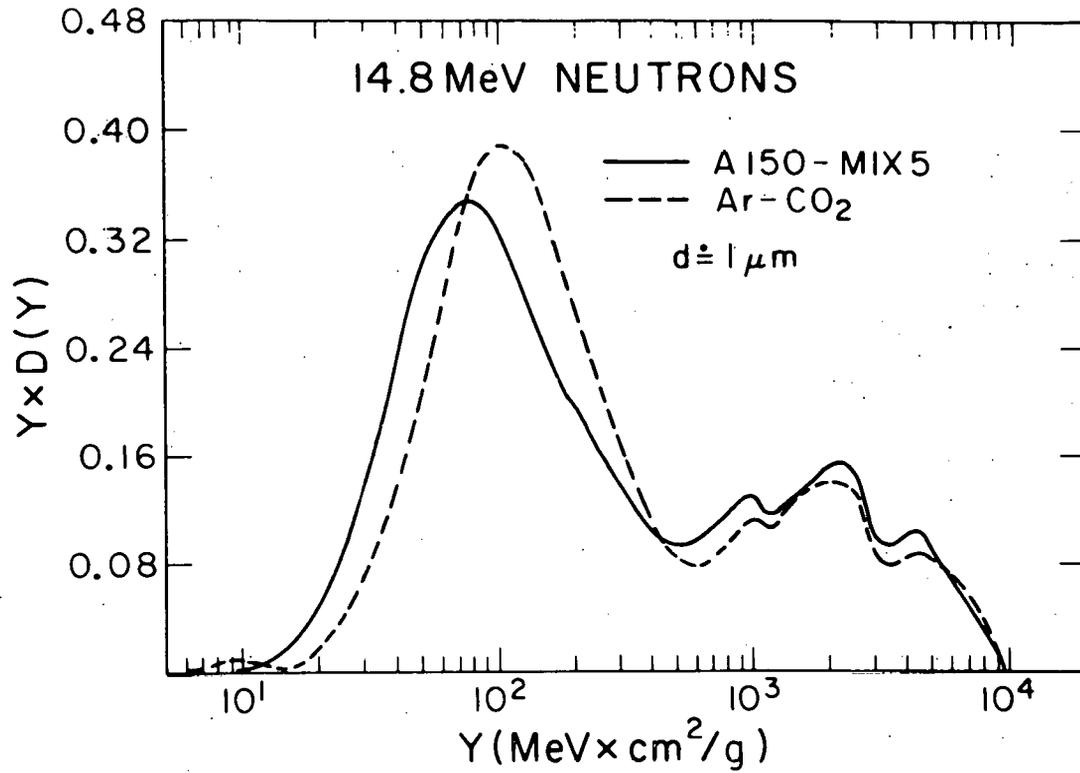


Fig. 7 Plots of the event-size-weighted fractional dose as a function of event-size for exposure to 14.8 MeV neutrons. Curves are shown for A150 (Mix 5) gas and Ar-CO₂ gas. Data are normalized to the stopping power of A150-plastic.