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Brookhaven National Laboratory

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Upton, New York

HANFORD ENGINEERING DEVELOPMENT LABORATORY
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ABSTRACT

CR-39 Polymer, a new solid state track recorder with unprecedented sensitivity to lightly ionizing particles (such as protons) is being developed for eventual neutron dosimetry applications in the Fusion Materials Irradiation Test Facility and elsewhere. The diameters of proton tracks have been found to vary smoothly and reproducibly as a function of energy from 0.20 to 18.0 MeV. Preliminary results on the response of CR-39 polymer to proton tracks as a function of angle show a rapid decrease of the registration efficiency from 100% to 0 for angles of incidence less than 75°. Proton recoil track size distributions in CR-39 polymer irradiated with monoenergetic neutrons of varying energy are presented. Some proposed high energy neutron dosimetry and radiography systems using CR-39 polymer are discussed.

INTRODUCTION

Since the recent discovery of the track recording properties of CR-39 polymer [1, 2], this solid state track recorder (SSTR) has been shown to have a number of unique and useful properties not found previously. Among these properties, those most advantageous for neutron applications are:

(1) High sensitivity to lightly ionizing particles. A measure of this sensitivity is the broad energy range of proton track registration [3].

(2) Homogeneous bulk etch rate. Samples of CR-39 polymer SSTR may be etched for periods of time resulting in tens of microns of surface removal, and the surface of the SSTR retains its excellent optical quality.

(3) High resistance to β and γ radiation. Samples of CR-39 polymer have been exposed to a total β-γ dose of 10^7 Rads and were still found to record discernible tracks from a lithium foil [4].

(4) The response to lightly ionizing particles can be changed by altering the etching conditions [5]. In neutron fields, for example, tracks from alpha particles and heavier ions can be revealed while at the same time discriminating against neutron induced proton recoil tracks.

These properties make CR-39 polymer an excellent SSTR candidate for neutron applications. The heretofore unavailable capability for proton track registration over a wide energy range makes possible many dosimetry applications which rely on the H(n,p) reaction. This is a particular advantage, since the H(n,p) cross section and angular distribution are quite well known over a broad energy range (including the entire neutron energy spectrum range of the Fusion Materials Irradiation Test [FMIT] Facility).

The response of CR-39 polymer is being calibrated for eventual neutron dosimetry in FMIT and also for other applications in U.S. nuclear reactor energy programs.

EXPERIMENTAL

CR-39 polymer SSTR have been exposed to proton beams using 90° scattering through a thin ($100 \mu g/cm^2$) gold foil. For protons of energy greater than 10 MeV, protons scattered forward at 45° have been analyzed using a magnetic spectrometer to avoid contamination due to inelastically scattered protons and reaction products.

For protons, the CR-39 polymer is etched in 6.25N NaOH solution to which 0.5 mole % Dowfax Surfactant has been added. The temperature of the etchant is maintained at 70.0±0.1°C and the SSTR are typically etched for 16 hours.

Track densities and track size distributions are obtained with the aid of a computerized Quantimet 720 system coupled to an optical microscope.

RESULTS AND DISCUSSION

Results on the response of CR-39 polymer to protons in the energy range from 0.2 to 6 MeV and to alpha particles in the energy range from 3.2 to 6.1 MeV have been reported previously [3]. The diameters of normally incident tracks were measured as a function of energy, resulting in an integral response for alpha particles and a rapidly varying response for protons. The proton results have since been extended to an energy of 18 MeV. Figure 1 shows microphotographs of normally incident proton tracks with energies of 9.0, 12.0, 15.0, and 18.0 MeV. The mean diameters of these tracks, measured with the aid of the Quantimet, are plotted as a function of energy in Figure 2. The line in Figure 2 is a result of a computer code which simulates etching in CR-39. CR-39 polymer continues to show a differential energy response as a function of proton energy up to 18 MeV. Experiments are in progress to extend this calibration to higher proton energies.

Experiments have been initiated to calibrate the proton response of CR-39 polymer as a function of angle of proton incidence. CR-39 polymer SSTR have been exposed to scattered proton beams at incidence angles of 90°, 85°, 80°, 75°, 70°, 65°, and 60°. Microphotographs of 5 MeV protons incident at 90°, 85°, 80°, 75°, and 70° are shown in Figure 3. At 80° and 75° the track profiles become more elipsoidal and at 70° the tracks become very faint. At 65° and 60°, the proton tracks were not visible. Preliminary results on the etching efficiency as a function of angle are shown in Figure 4. The track fading at 70° is accompanied by a 50% reduction in registration. For angles greater than or equal to 75°, the response is essentially unity, whereas for angles less than 65°, the response is zero. The response at all angles has been normalized to unity at 90°. Normally incident protons register with 100% efficiency up to at least 5 MeV [3]. The angular response of CR-39 polymer SSTR to 8 MeV protons has been found to be quite similar to the response at 5 MeV. This similarity is to be expected, since for a 16 hour etch, the cone angles for 5 and 8 MeV protons are nearly the same. This rather simple, "step function" angular response should simplify the use of CR-39 polymer under conditions of isotropic track incidence. On the other hand, it may be possible to use this response to provide some angular information in non-isotropic neutron fields.

Further angular response measurements are in progress.

APPLICATIONS OF CR-39 POLYMER SSTR IN NEUTRON DOSIMETRY

Several applications of CR-39 polymer SSTR for neutron dosimetry in FMIT have been proposed. In the simplest of these, CR-39 polymer could be used to record proton recoils from a hydrogenous radiator exposed to neutrons as shown in Figure 5. The response of CR-39 polymer above 18 MeV is still under investigation, but by using a large scattering angle from the radiator, the range of expected recoil proton energies can be compressed from the 40 MeV range of neutron energies down to less than 18 MeV. The resulting proton recoil diameter spectrum can be converted into an energy spectrum using the calibration curve in Figure 1, and the proton energy spectrum can be unfolded to reveal the incident neutron energy spectrum.

Other reactions may offer unique advantages for FMIT neutron spectrometry. Among these are the $^{6}Li(n,\alpha)$ and $^{10}B(n,\alpha)$ reactions [6,7]. CR-39 polymer SSTR have been shown to provide an integral response (constant diameters) to alpha particles in the range from 3-6 MeV [3]. For the $^{6}Li(n,\alpha)$ reaction, an integral response would be expected for the alpha particles whereas the product tritons would result in smaller tracks with differential energy response. When etched under less sensitive chemical conditions, proton tracks are not revealed in CR-39 and the alpha particle energy response becomes differential [5]. Thus, with proper calibration alpha spectrometry measurements can be made in high energy neutron fields that would otherwise result in a background of proton recoils from neutron interactions with the hydrogen atoms in the CR-39 polymer.

The concept of neutron pinhole radiography has been advanced for FMIT [8] and calculations have shown that adequate spatial resolution can be obtained with such a device [9]. Passive radiometric foils can be placed in the image plane of the collimator and the spatial distribution of the reaction products can be determined to map the image of the neutron source. Alternatively, SSTR may be placed in the image plane, and the resultant track densities can be related to neutron fluence at a given point. Figure 6 shows a sample of CR-39 polymer that was exposed using a D-T 14 MeV neutron source. It is encouraging to note that most of the proton recoil tracks have circular profiles indicating that they are incident nearly normal to the surface (as is expected from angular calibration data) and that most of the tracks have small diameters corresponding to energies near 14 MeV (compare with Figure 1). The unexpected simplicity of this 14 MeV neutron induced proton recoil response augers well for the prospects of unfolding the proton recoil spectrum to determine the incident neutron energy spectrum. Additionally, the spatial distribution of proton recoil tracks should be proportional to the intensity of incident neutrons so that a two dimensional inverse image of the neutron source is obtained.

An alternative to in-beam pinhole neutron imaging as described above is to use the arrangement shown in Figure 7. A proton radiator of some suitable material such as polyethylene ($[CH_2]_n$) is placed in the image plane of the pinhole collimator and the resultant proton recoils are viewed through a proton pinhole collimator using CR-39 polymer. In order to obtain normal incidence for the recoil protons, the CR-39 is placed at an angle to the proton image plane, so that corrections for projection angle and small geometric efficiency differences must be made to obtain an inverted image of the proton recoil distribution which is, in turn, an inverted image of the source neutron distribution.

In the event that the background caused by neutron induced proton recoils in the CR-39 polymer becomes a problem, ^{6}Li or ^{10}B could be used as an alpha radiator and an alpha particle image can be obtained with the SSTR. Alternatively, the recoil protons can be degraded to lower energies (which are not present in great abundance from direct interactions of neutrons with the CR-39 polymer) and the larger diameter tracks from the low energy protons can be easily distinguished from the smaller neutron induced proton recoil background tracks in the polymer.

This latter method has resulted in the concept of a radiographic neutron camera.[10] A prototype model of this radiographic neutron camera is shown in Figures 8 and 9. The prototype camera will be used to explore neutron source imaging using benchmark fields and low intensity mockups. The camera is shown in position next to a sealed tube D-T 14 MeV neutron generator. Protons produced in a proton radiator placed next to the neutron source will be degraded by passing through pressurized gas and a pinhole image of the degraded proton recoils will be projected on a CR-39 polymer track recorder.

A Fresnel zone plate[11,12] could also be used in place of the pinhole collimator to provide higher efficiency. The three dimensional Fresnel shadowgraph image would, in this case, only provide an image of the two dimensional image from the proton radiator. Through benchmark testing and optimization, the prototype radiographic camera will evolve into a neutron imaging device suitable for use at FMIT and other fusion environments.

The encouraging response for 14 MeV proton recoils in CR-39 polymer shown in Figure 6 has led to attempts to quantify the response to incident neutrons. The proton track diameter distributions as obtained by the Quantimet are shown in Figure 10 for incident neutron energies of 0.57, 2.1, 5.3, and 15.1 MeV. A 1 mm thick high density polyethylene radiator was used in surface contact with the CR-39 for these exposures. For the three higher energy exposures, peaks are found at diameters corresponding to slightly less than the diameter expected for a direct knock-on proton. This apparent shift to higher energy is caused by etching of proton recoils formed within the CR-39 polymer. These tracks are not exposed to the etchant for the full 16 hours resulting in smaller diameters. The increase in intensity of these peaks with increasing neutron energy corresponds to an increase in effective thickness of the radiator due to the larger range of the recoil protons. The absence of a peak near the maximum proton energy in the 0.57 MeV neutron irradiation is probably due to a combination of the decreased effective thickness of the radiator foil and lack of contrast for the shallow proton recoil tracks resulting in a loss of optical efficiency for detection of the tracks. This latter effect is enhanced by the fact that the recoil protons are emitted isotropically (in the center of mass system) resulting in shallower tracks within the angular range of registration. Also, the dominant source of proton recoils at this energy is the hydrogen atoms in the CR-39 polymer so that all tracks will not be etched for the full 16 hours. This loss of efficiency at low energy is also apparent in the higher energy exposures where low energy incident tracks are expected due to proton recoil energy degradation in the radiator.

A major peak at approximately 3μm is present in all of the diameter spectra. This peak is due to either incompletely etched low energy protons or to carbon or oxygen recoils from neutron inelastic scattering within the CR-39 polymer.

On the basis of these results, further exposures are being conducted with 14 MeV neutrons to attempt to simplify the characteristics of the diameter spectra by optimizing the radiator thickness and etch time. It is likely that the optimum radiator may be no radiator at all in the case of high intensity, high energy neutron exposures.

CONCLUSIONS

CR-39 polymer is an extremely promising material for use in high energy neutron dosimetry applications. Its unprecedented wide energy response for protons, variable response characteristics, radiation resistance, and high optical quality make it ideal for this purpose. Because of these unique characteristics, applications of CR-39 polymer in the FMIT, as well as other Magnetic Fusion Energy, Light Water Reactor, and Fast Breeder Reactor environments are currently being developed.

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Figure Captions

Figure 1. Tracks from normally incident protons in CR-39 polymer solid state track recorders.

Figure 2. Proton track diameter as a function of energy for CR-39 polymer solid state track recorders etched in 6.2N NaOH for 16 hours at 70°C.

Figure 3. Microphotographs of tracks in CR-39 polymer from 5.00 MeV protons with the indicated incidence angles. The arrows indicate faint tracks at 75° and 70° incidence.

Figure 4. Relative track response of CR-39 polymer as a function of incidence angle for 5.00 MeV protons.

Figure 5. CR-39 polymer neutron-induced proton recoil spectrometry using large angle scattering.

Figure 6. Proton recoil tracks resulting from 14 MeV neutrons. The extremely large track near the center of the field is probably an α particle track produced by the decay of ^{222}Rn or one of its daughters.

Figure 7. Double pinhole radiography using neutron and proton pinhole collimators.

Figure 8. Radiographic neutron camera showing holders for mounting of radiators, collimators, and CR-39 polymer track recorders.

Figure 9. Radiographic Neutron Camera. Camera (foreground) is placed adjacent to a 14 MeV (D-T) neutron generator.

Figure 10a. Diameter distribution for tracks produced in CR-39 polymer irradiated with 0.57 MeV neutrons. The curve represents a smooth fit to a histogram with a bin size of 0.25μm. The CR-39 polymer was etched for 16 hours at 70.0°C in 6.25 NaOH. The diameter corresponding to the maximum proton recoil energy is indicated with an arrow.

Figure 10b. Track diameter distribution for CR-39 polymer irradiated with 2.1 MeV neutrons.

Figure 10c. Track diameter distribution for CR-39 polymer irradiated with 5.3 MeV neutrons.

Figure 10d. Track diameter distribution for CR-39 polymer irradiated with 15.1 MeV neutrons.

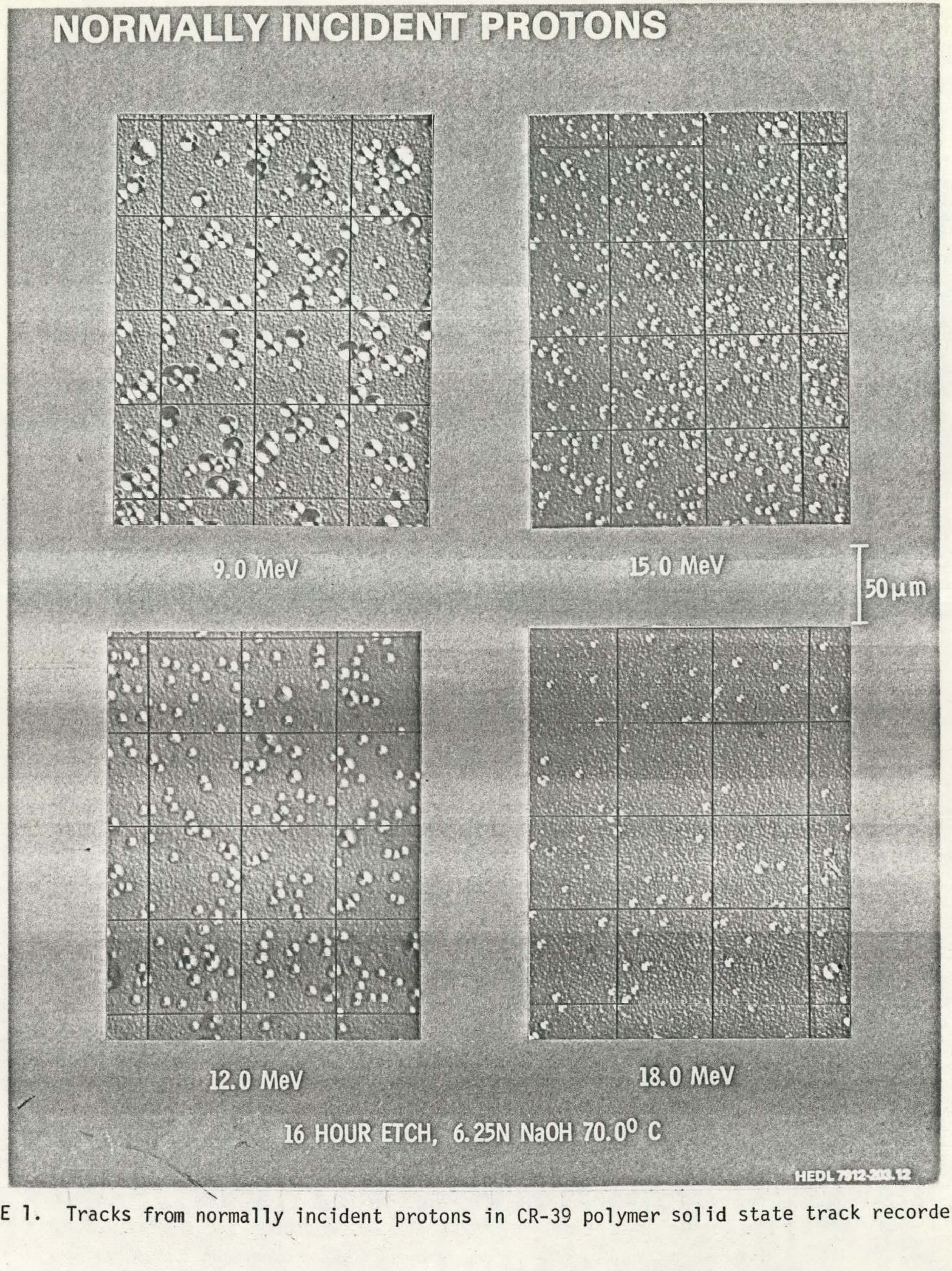
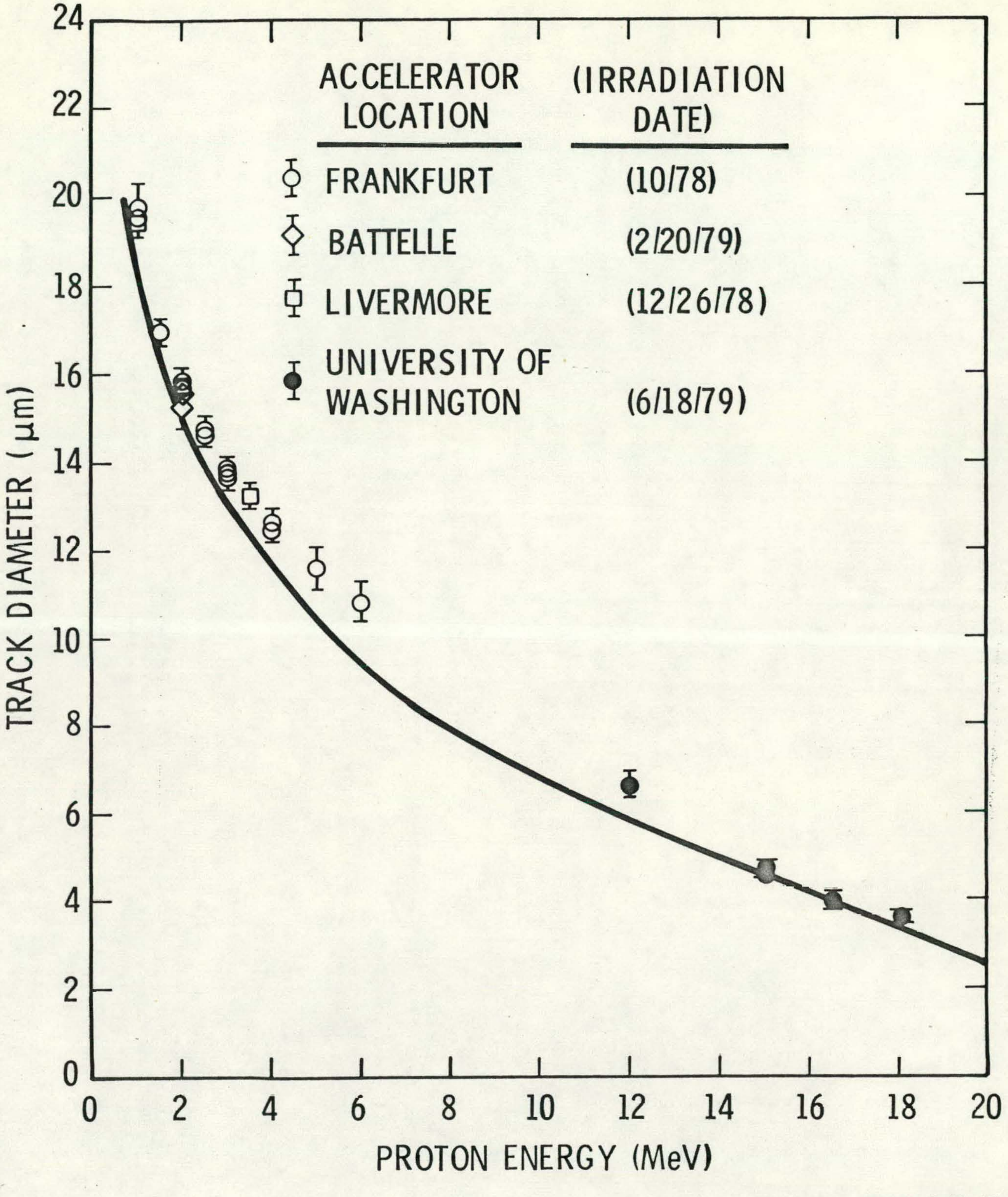
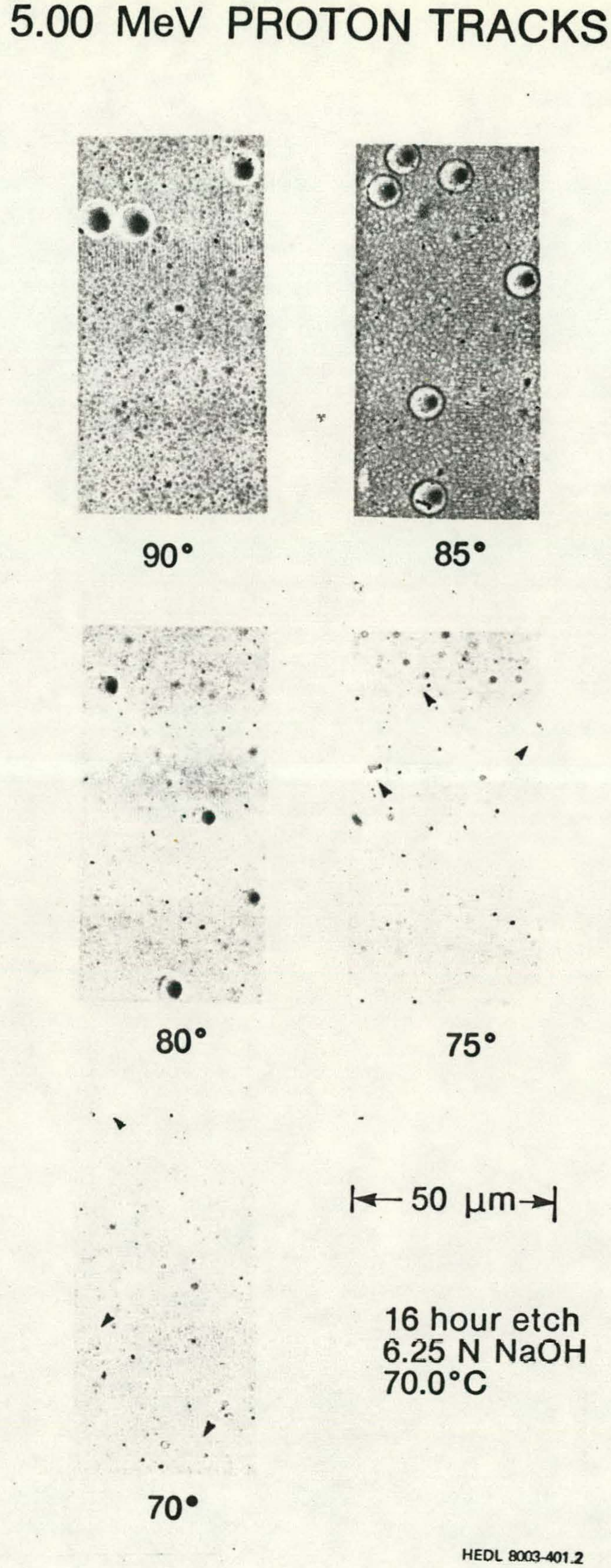


FIGURE 1. Tracks from normally incident protons in CR-39 polymer solid state track recorders.



HEDL 7908-146.7a

Figure 2. Proton track diameter as a function of energy for CR-39 polymer solid state track recorders etched in 6.2M NaOH for 16 hours at 70°C.



HEDL 8003-401.2

Figure 3. Microphotographs of tracks in CR-39 polymer from 5.00 MeV protons with the indicated incidence angles. The arrows indicate faint tracks at 75° and 70° incidence.

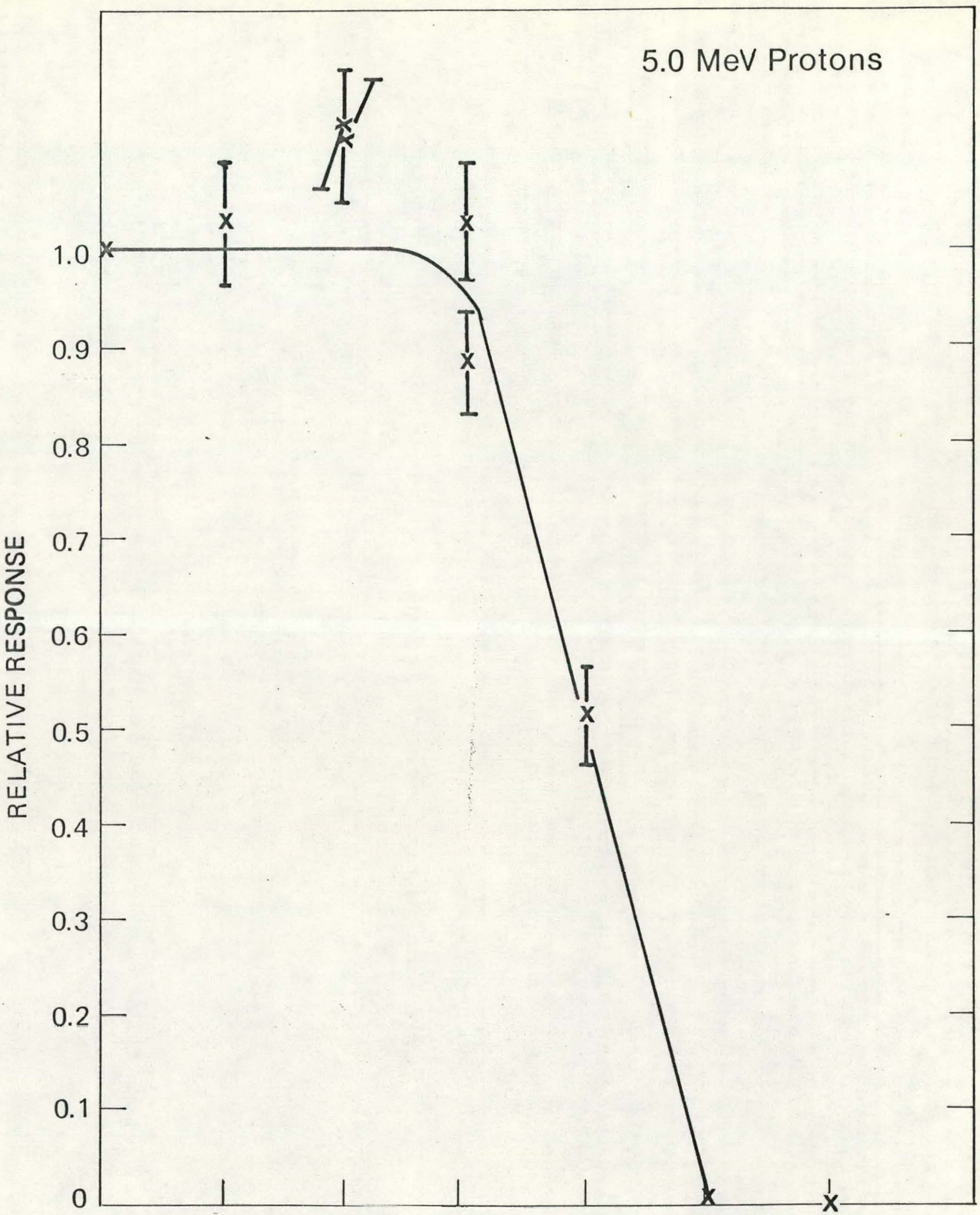


Figure 4. Relative track response of CR-39 polymer as a function of incidence angle for 5.00 MeV protons.

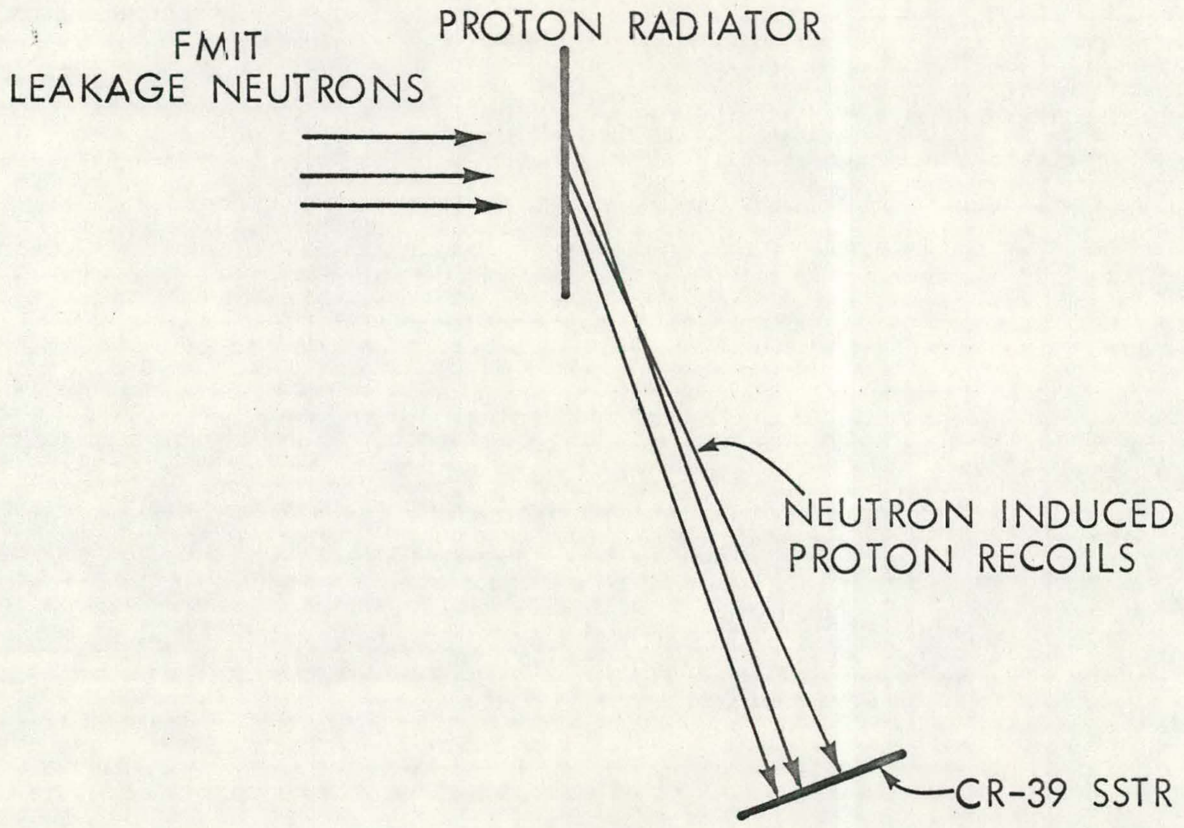


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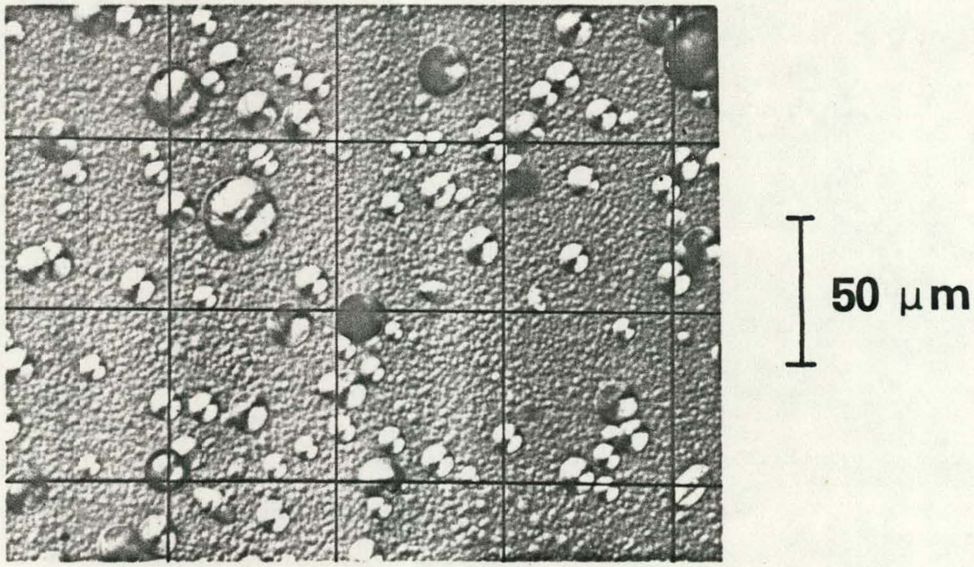
FIGURE 5. CR-39 Polymer neutron-induced proton recoil spectrometry using large angle scattering.

PROTON RECOIL TRACKS RESULTING FROM 14 MeV NEUTRONS



CR-39 POLYMER

16 HOUR ETCH, 6.25 NaOH 70.0°

HEDL 7912-203.11

Figure 6. Proton recoil tracks resulting from 14 MeV neutrons. The extremely large track near the center of the field is probably an α particle track produced by the decay of ^{222}Rn or one of its daughters.

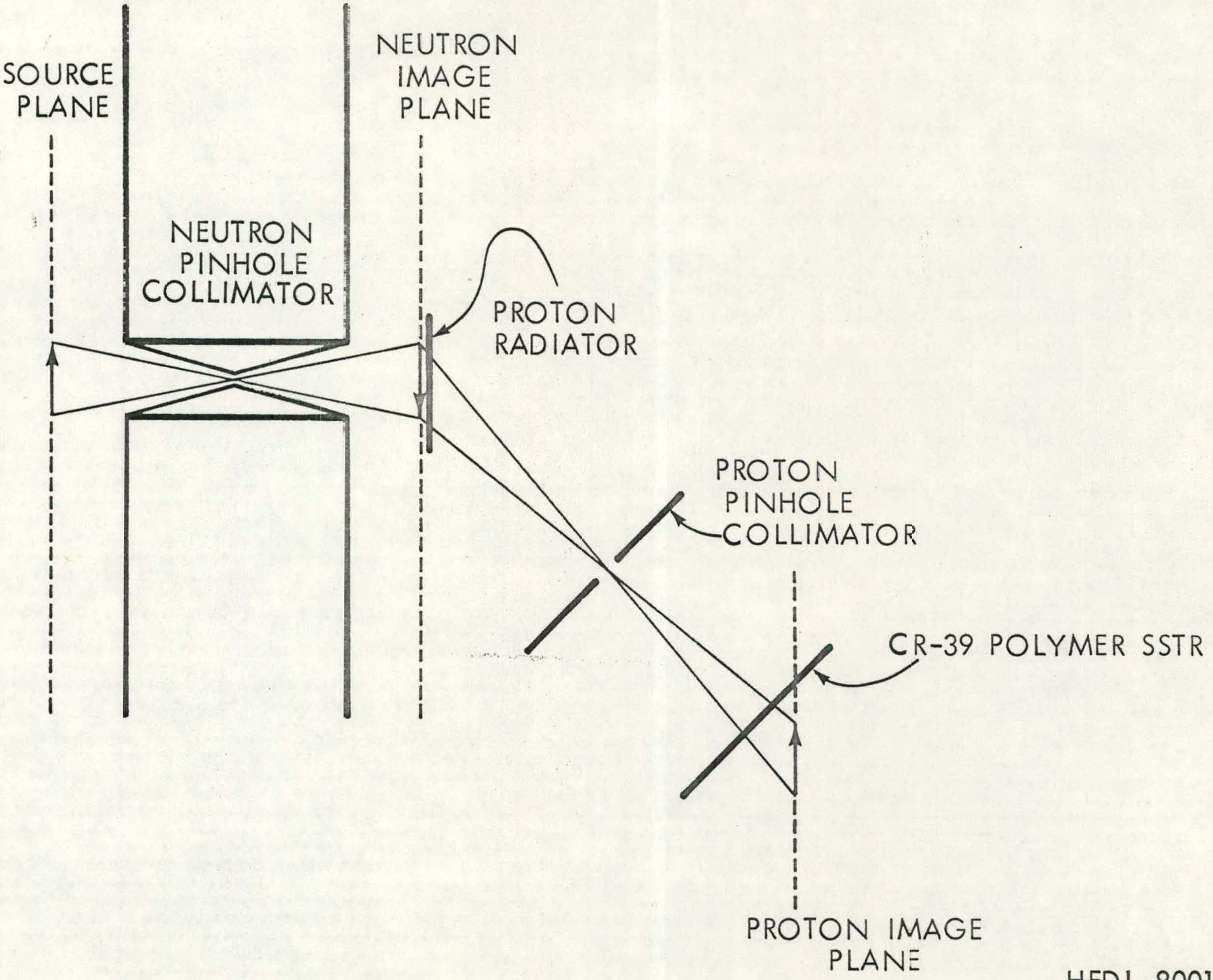


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FIGURE 7. Double pinhole radiography using neutron and proton pinhole collimators.

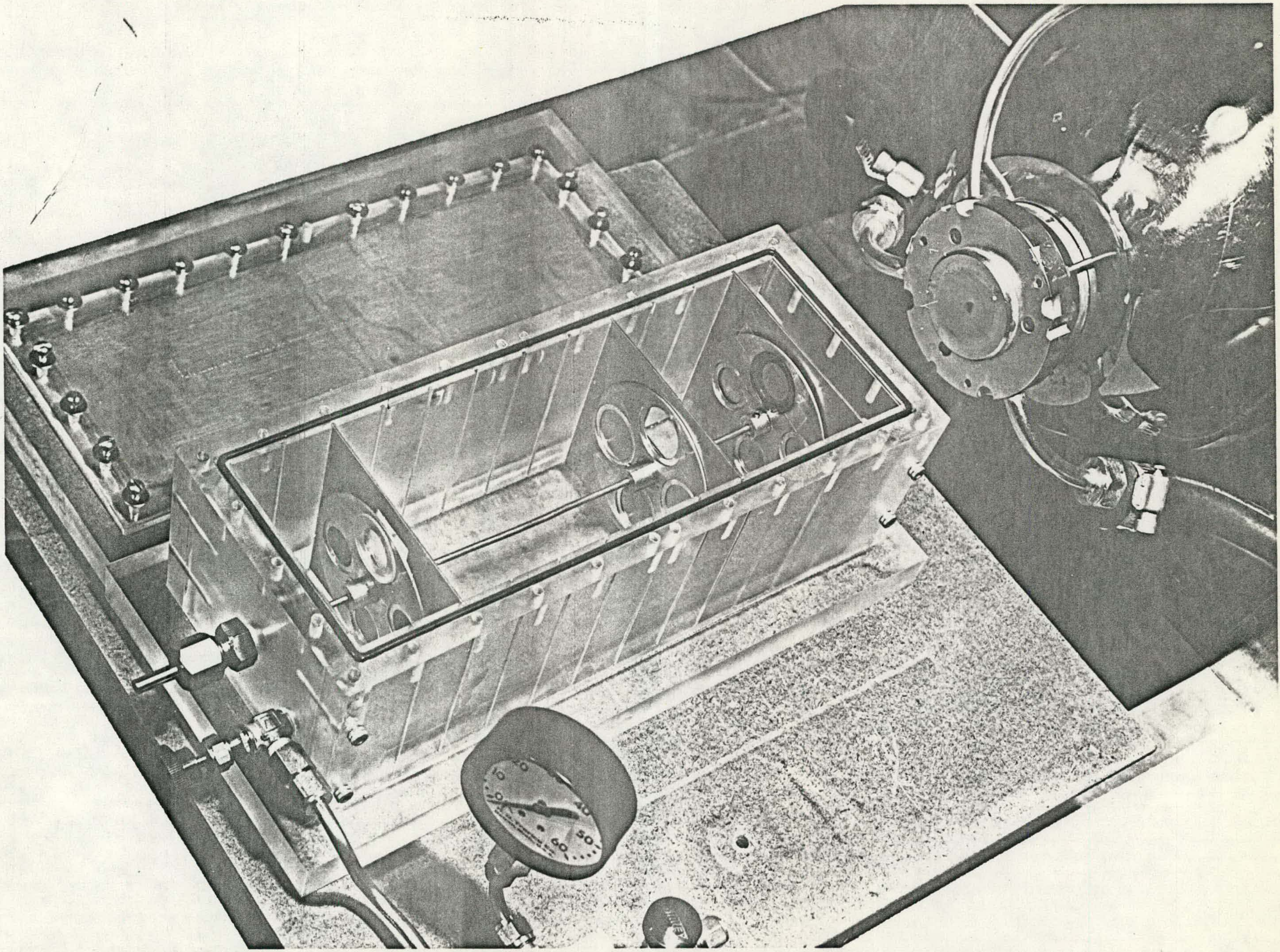


FIGURE 8. Radiographic neutron camera showing holders for mounting of radiators, collimators, and CR-39 polymer track recorders.

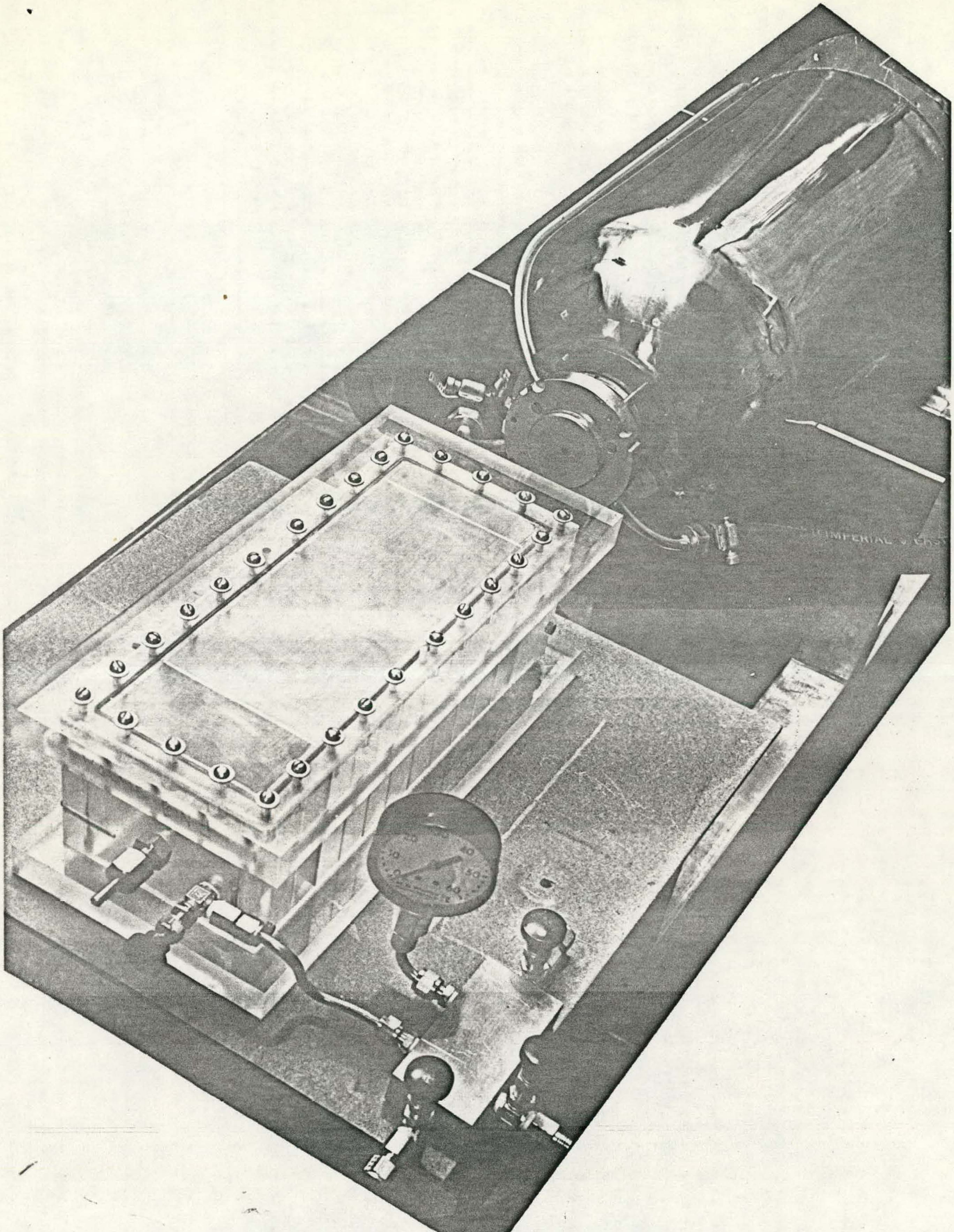
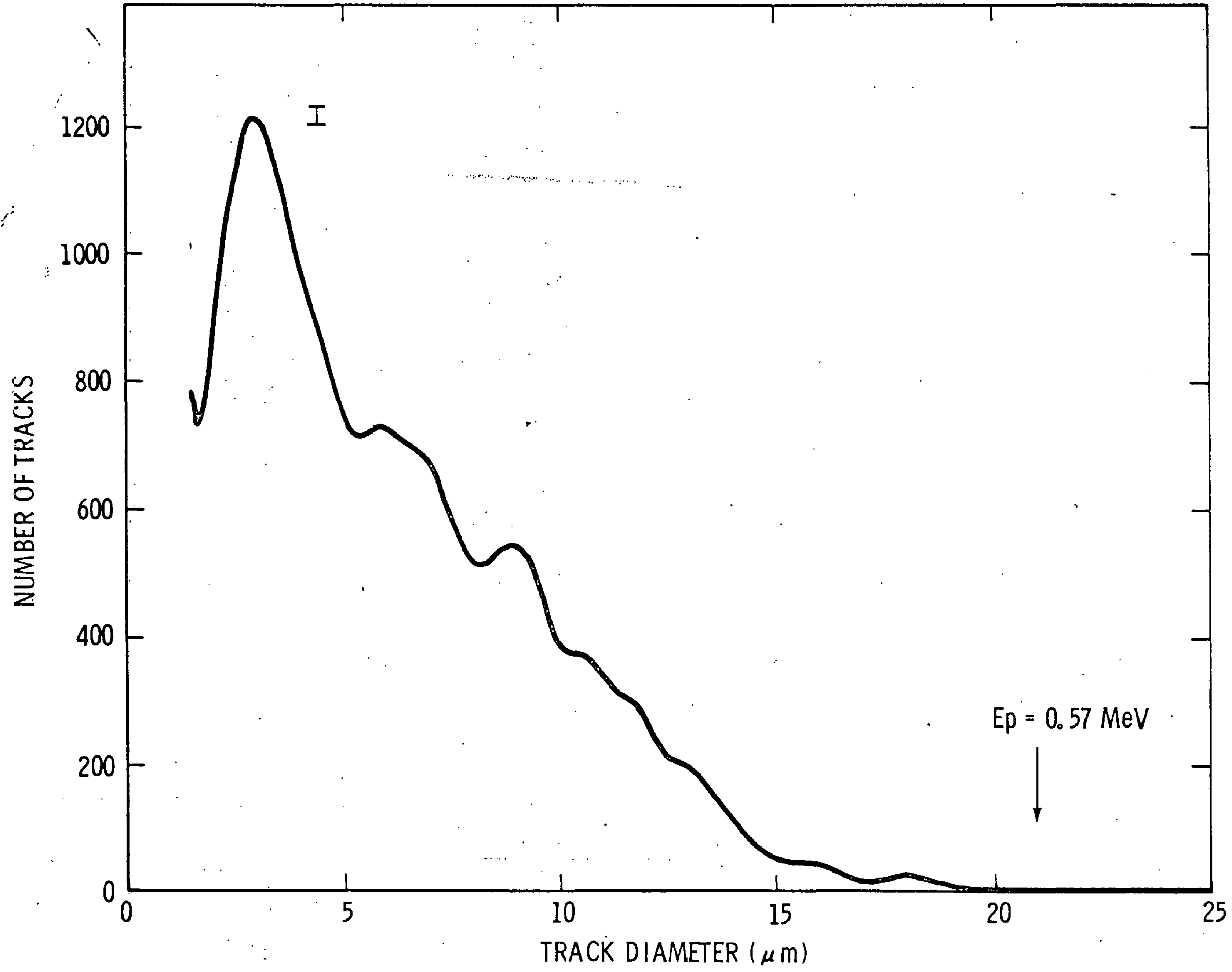
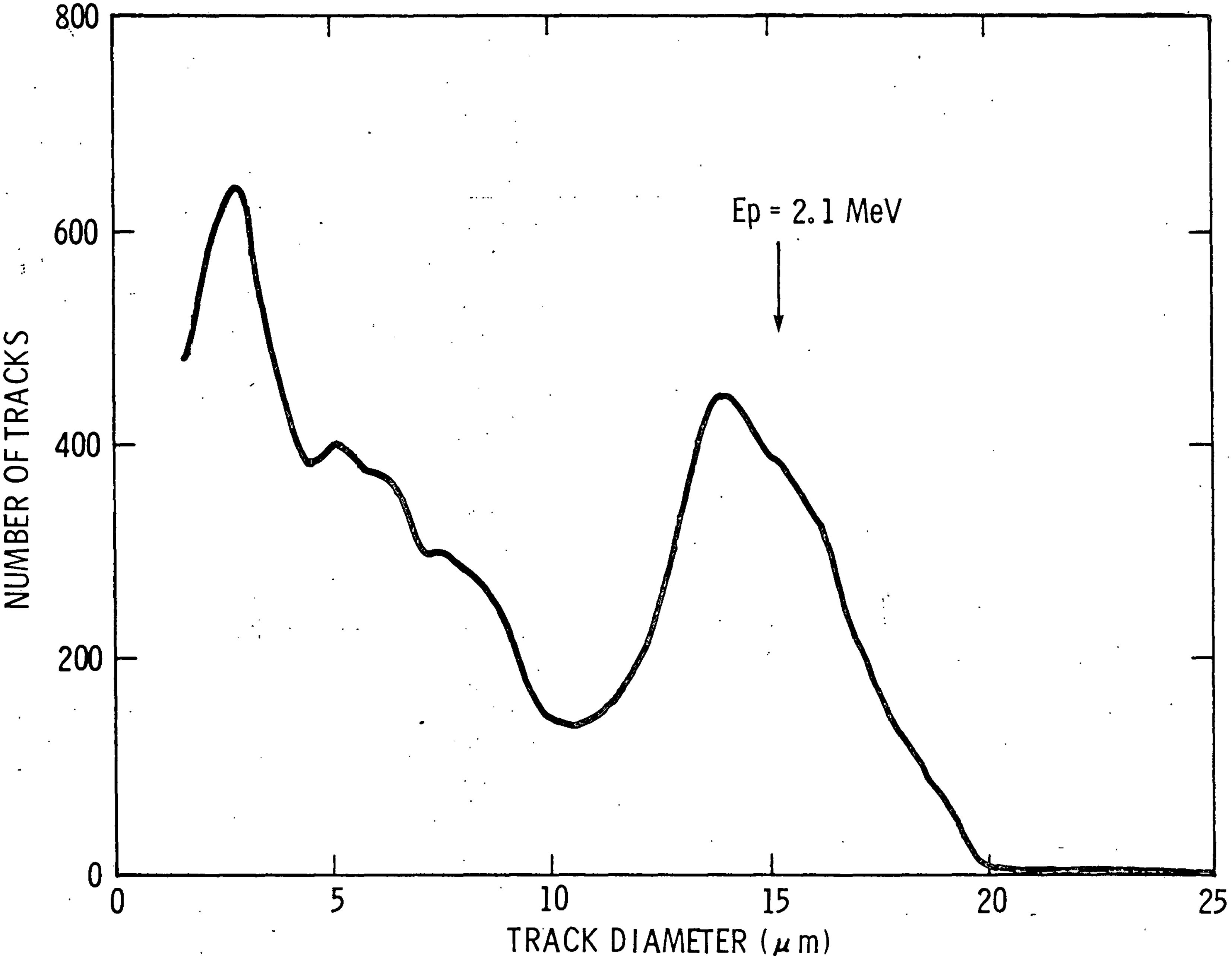


FIGURE 9. Radiographic Neutron Camera. Camera (foreground) is placed adjacent to a 14 MeV (D-T) neutron generator



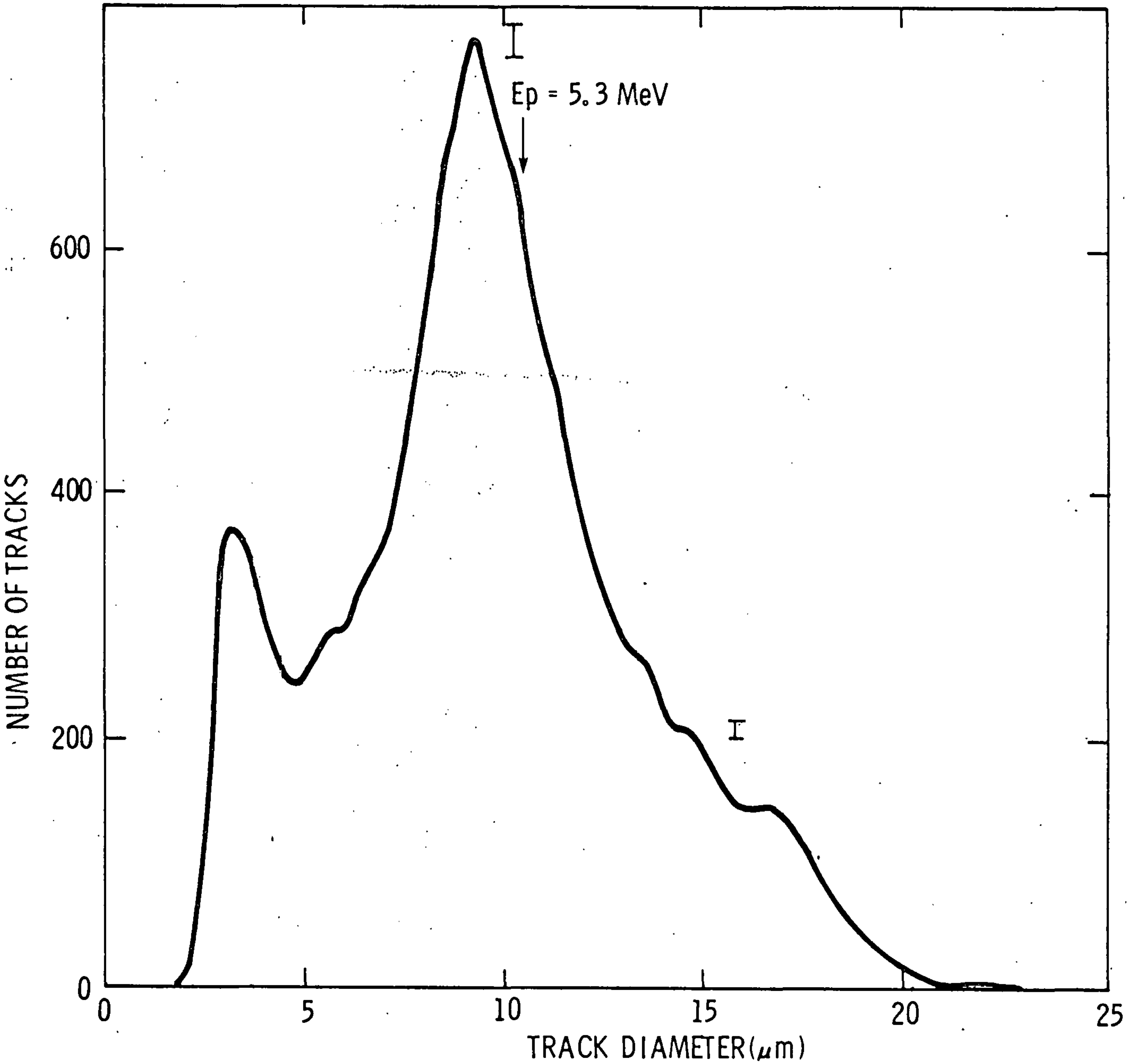
HEDL 8004-157.3

Figure 10a. Diameter distribution for tracks produced in CR-39 Polymer irradiated with 0.57 MeV neutrons. The curve represents a smooth fit to a histogram with a bin size of 0.25μm. The CR-39 polymer was etched for 16 hours at 70.0°C in 6.25 NaOH. The diameter corresponding to the maximum proton recoil energy is indicated with an arrow.



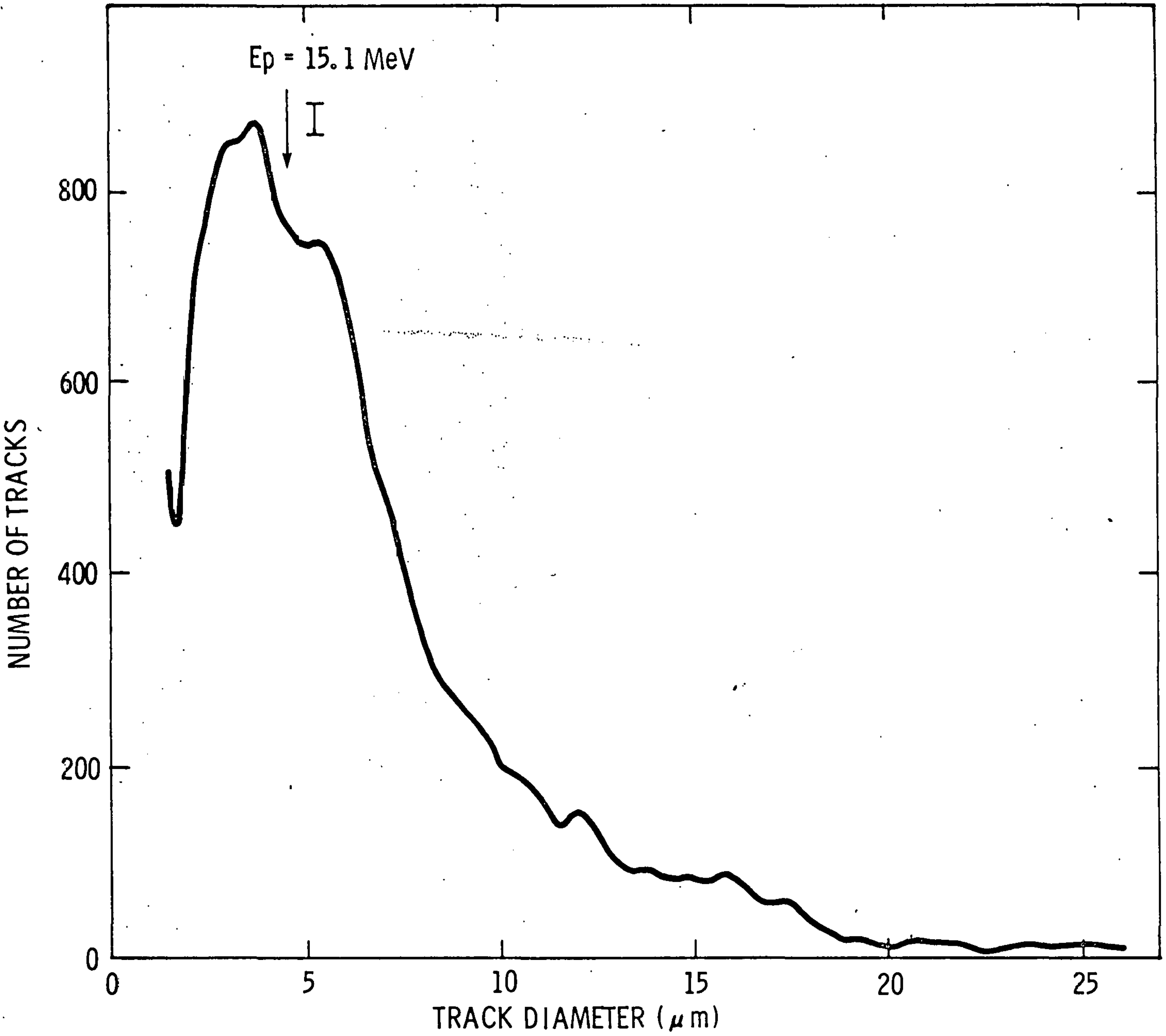
HEDL 8004-157.2

Figure 10b. Track diameter distribution for CR-39 Polymer irradiated with 2.1 MeV neutrons.



HEDL 8004-157.1

Figure 10c. Track Diameter Distribution for CR-39 Polymer irradiated with 5.3 MeV neutrons.



HEDL 8004-157.4

Figure 10d. Track Diameter Distribution for CR-39 Polymer irradiated with 15.1 MeV neutrons.