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**ADVANCES IN SSTR TECHNIQUES FOR DOSIMETRY
AND RADIATION DAMAGE MEASUREMENTS**

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and Frank H. Ruddy**

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ABSTRACT

Solid state track recorders (SSTR) have been applied in diverse nuclear reactor research by many workers throughout the world. Two recent advances are described which possess outstanding relevance for reactor research, namely the evolution of SSTR radiation damage monitors and the development of CR-39, a new plastic SSTR of extremely high sensitivity. Results from high fluence irradiations of natural quartz crystal SSTR are used to illustrate the concept of the SSTR radiation damage monitor. Response characteristics of CR-39 are presented with emphasis on the remarkable proton sensitivity of this new SSTR.

I. INTRODUCTION

Application of Solid State Track Recorders (SSTR) in reactor dosimetry extends from high temperature environments found in high power reactor cores to very low flux environments in out-of-core locations, critical assemblies or away from reactor (AFR) experiments. A neutron energy region covering more than eight decades, from thermal to fusion energies, arises in these applications. The intensity range of neutron flux/fluence is even greater, extending over more than thirteen decades. The ability of SSTR techniques to meet the needs of reactor experiments in such a diversity of environments rests upon many unique attributes. Many of these advantageous characteristics were emphasized in the earliest reactor applications⁽¹⁾ and have subsequently been stressed at more recent symposia.^(2,3)

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An important illustration of these unique SSTR characteristics lies in the rather unexpected development of a nondestructive actinide assay technique for spent reactor fuel assemblies, which is presented in a companion paper at this symposium.⁽⁴⁾ Here the combination of high sensitivity and virtually unmatched discrimination against background radiation field components, particularly gamma-rays, has opened up an entirely new area of AFR experiments.^(5,6)

Although less spectacular, some progress has also been effected for in-core SSTR applications in high power (HP) reactor environments. While the HP needs of U.S. nuclear reactor energy programs are just as urgent, the multiplicity of effects arising in high fluence (HF) irradiations introduces considerable complexity. Three major effects which can limit SSTR-HF applicability are: annealing, radiation damage, and background track production. It must also be noted that the existence of track pile-up produces accuracy limitations at higher track densities.

Additional work bearing upon HF applicability is in progress, namely: 1) annealing studies in natural quartz crystals, 2) extension of track pile-up limitations, and 3) development of advanced automated track scanning systems. Results from all of these current efforts will be reported in sequel.

This exposition focuses upon two recent SSTR advances that appear destined for significant exploitation in reactor environments. One is applicable for HF experiments, while the other is restricted to very low power/fluence irradiations. The recent HF development, presented in the next section, is not actually a dosimetry technique per se, but deals with the newly evolved concept of the SSTR radiation damage monitor.⁽³⁾ The remarkable properties of CR-39, a new plastic SSTR material useful for very low power/fluence dosimetry experiments, are described in the last section.

II. SSTR RADIATION DAMAGE MONITORS

Mica and natural quartz crystals have been identified as outstanding SSTR candidates for accurate HF absolute fission rate measurements.⁽⁸⁾ Sources of naturally occurring mica and quartz crystals exist that are sufficiently free of natural uranium and thorium to render background track production negligible.⁽⁸⁾ Techniques have been developed to prepare and quantify very low-level SSTR fission deposits, down to the sub-nanogram/cm² regime, for many actinide isotopes of interest.⁽⁹⁾ Annealing studies in mica reveal that extended applications to above 500°C are possible without entailing any quantitative correction.⁽¹⁰⁾

Even more promising are preliminary annealing investigations of natural Brazilian quartz crystals, which reveal perhaps the best fission track retention characteristics of all available SSTR-HF candidates. This outstanding property is shown in Figure 1, which

presents preliminary data on the annealing correction for the 100 plane. The basic parameter in this empirical annealing relation is the maximum etched track length ℓ in the surface plane resulting from normally incident fission fragments. Tracks from isotropic and normally incident fission on the 100 plane and 001 plane are shown in Figures 2 and 3, respectively. Quartz crystal SSTR are etched in boiling 65% NaOH for 25 minutes. The actual parameter used in this empirical annealing correction, as shown in Figure 2, is the maximum track length in the surface plane of the annealed SSTR relative to that of an unannealed SSTR, i.e., (ℓ/ℓ_0) . On the basis of this and other experiments in progress, the onset of annealing effects occurs only for temperatures above roughly 800°C.

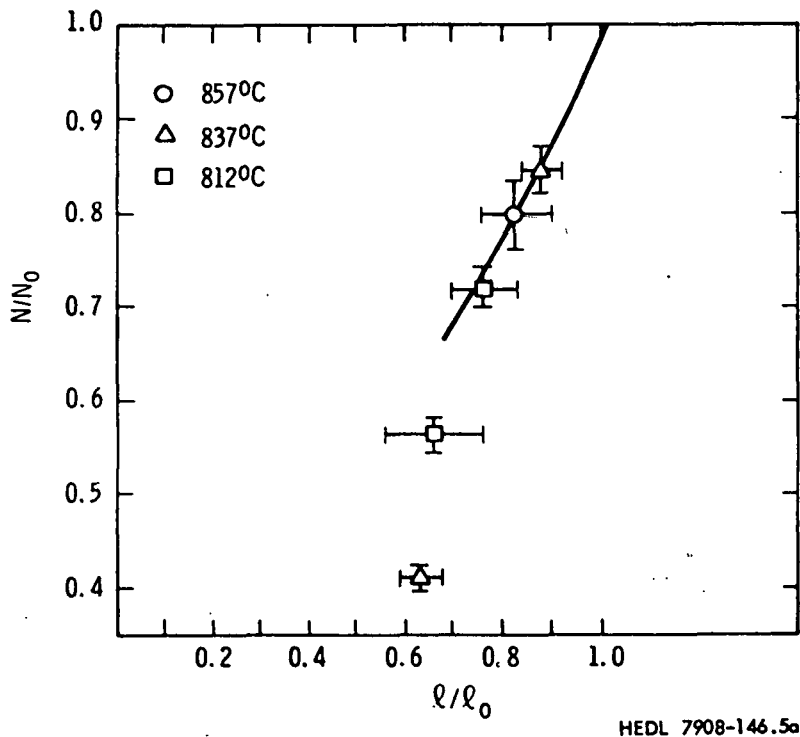
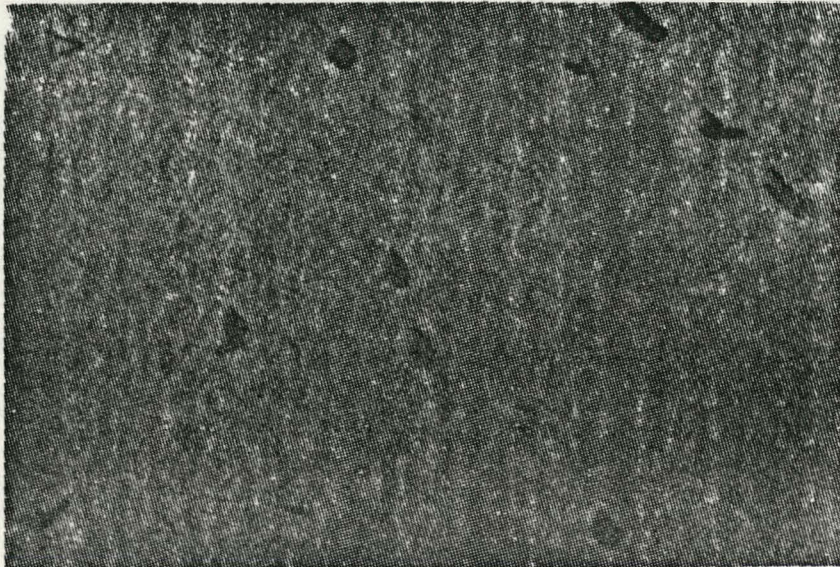
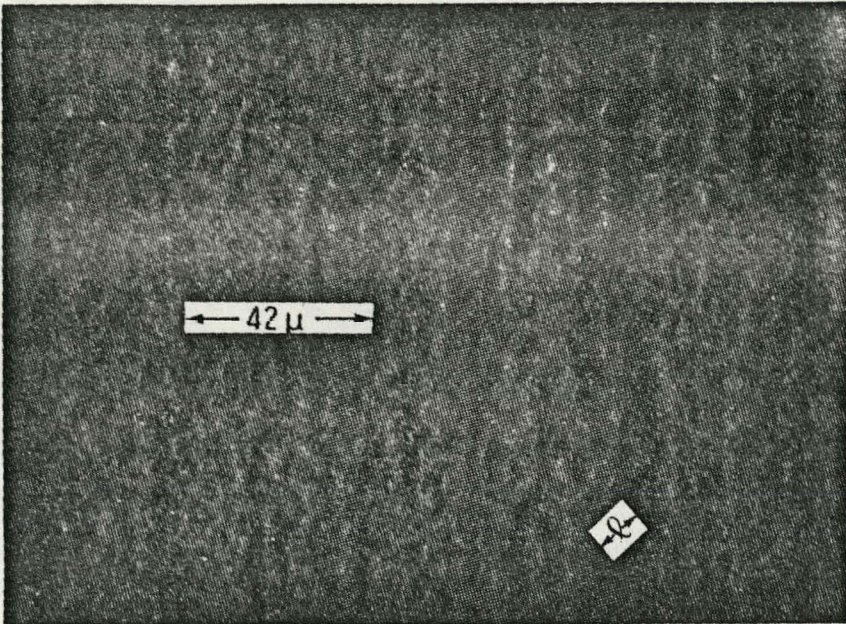


FIGURE 1. The Fraction of Observed Fission Tracks, N/N_0 in 100 Plane Quartz Crystal After a Given Annealing History as a Function of ℓ/ℓ_0 . (Here ℓ/ℓ_0 is the fractional length retained for normally incident fission fragments as shown in Figure 2b.)



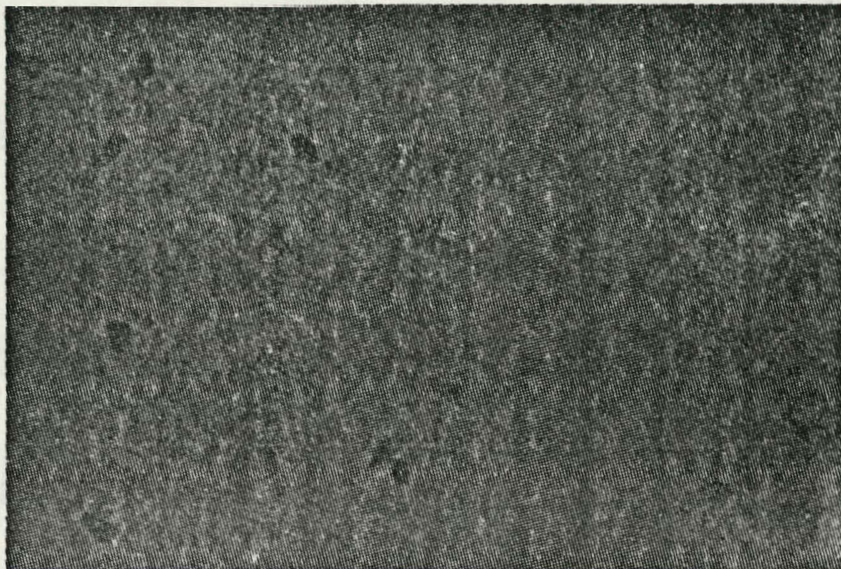
(a) ISOTROPIC INCIDENCE



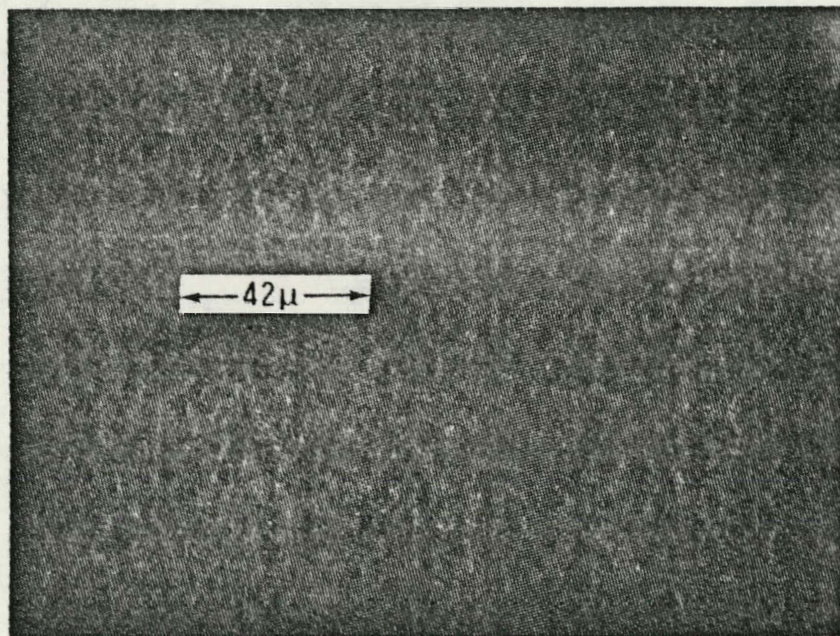
(b) NORMAL INCIDENCE

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FIGURE 2. Tracks in 100 Plane Quartz Crystal SSTR Due to Isotropic and Normally Incident Fission Fragments.



(a) ISOTROPIC INCIDENCE



(b) NORMAL INCIDENCE

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FIGURE 3. Tracks in 001 Plane Quartz Crystal SSTR Due to Isotropic and Normally Incident Fission Fragments.

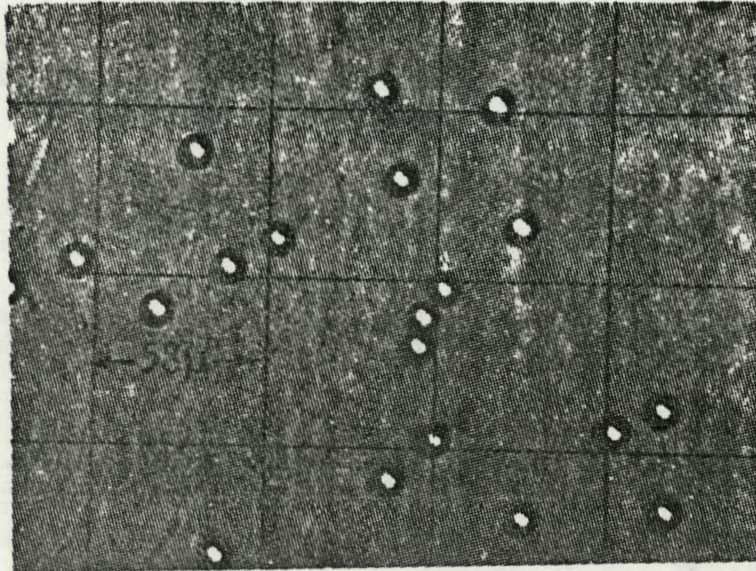
Based on this remarkable combination of attributes, HF fission rate measurements with natural quartz crystal SSTR were forecasted.⁽²⁾ To this end, HF experiments were initiated specifically to examine any effects induced directly in natural quartz crystal SSTR exposed without fission deposits. Irradiations were carried out in the EBR-II core, providing a fast neutron fluence of roughly 4×10^{21} n/cm². To eliminate annealing effects, in-core exposure locations in EBR-II were chosen so that temperatures never exceeded 430°C.

These quartz crystal SSTR were subsequently retrieved, etched, and then examined for background fission tracks. At this fluence level a detectable background track density is expected, even for high purity quartz crystals. However, no tracks were observed. Modifications of the standard etching procedure were introduced, but without success. Even a short five minute etch in 48% HF produced no tracks. There were apparently no fission tracks in these irradiated quartz crystal SSTR!

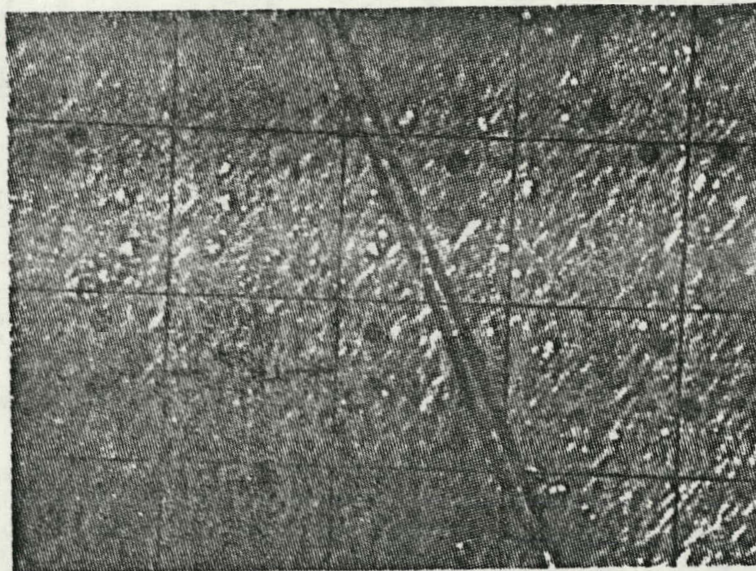
While these investigations did not succeed in producing any fission tracks, a significant change was detected in these irradiated quartz crystals, namely substantially increased bulk etch rate. This observation together with earlier work with mica SSTR led to the conjecture that fast neutron radiation damage of the quartz lattice might be responsible for the disappearance of all fission tracks. In these earlier studies with mica, it was found that mica specimens from diverse geographic locations could possess wide variations in bulk etch rate. These variations appeared to be correlated with both the age and uranium content of the mica specimens. Hence, it was speculated that the observed bulk etch rate variations in these mica specimens could be due to stored radiation damage from the decay of uranium impurities over geological times.

The fast neutron radiation damage speculation for the irradiated quartz crystals was confirmed by exposing these SSTR to normally incident fission fragments. A number of different etching procedures were attempted and fission tracks were finally observed with the etching procedure used for quartz glass. (Fission tracks of good optical contrast are obtained in quartz glass by etching in 48% HF acid for five minutes at room temperature.) Figure 4 compares tracks obtained in this manner for the 001 plane irradiated quartz crystals with tracks produced by normally incident fission fragments in quartz glass. The similarity of these tracks is striking, as is the dissimilarity with customarily observed fission tracks in 001 plane quartz crystals, as previously shown in Figure 3. Crystalline structure no longer dominates the features of these tracks. In fact, the lack of structure is indicative of a more amorphous medium.

One can, therefore, conclude that the irradiated quartz crystal is no longer crystalline, but rather has become amorphous with SSTR properties more akin to quartz glass than quartz crystal. On this



(a) QUARTZ GLASS



(b) IRRADIATED QUARTZ CRYSTAL

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FIGURE 4. Tracks of Normally Incident Fission Fragments in Quartz Glass and in Fast Neutron Irradiated Quartz Crystal ($\sim 4 \times 10^{21}$ n/cm²).

basis, the reason for the disappearance of all fission tracks in the HF irradiated specimens becomes apparent. Quartz glass does not possess anywhere near the retention capabilities of quartz crystal against annealing. In fact, fission tracks are rapidly annealed in quartz glass above 350°C.^(8,10) The nonexistence of tracks in these irradiated quartz specimens is therefore attributable to fast neutron lattice damage acting together with annealing.

Hence, the complexity introduced in HF-SSTR applications is not merely due to the fact that many different effects arise. To be sure, each of these effects can play a significant role in defining the HF limit of applicability of a given SSTR. However, it must be stressed that these different effects can act in consort rather than independently to produce an HF limit. In the example cited above, radiation damage of the quartz crystal lattice produced by a high fluence of fast neutrons altered SSTR annealing characteristics dramatically and as a consequence track fading due to annealing was considerably enhanced.

Although these HF effects are not actually separable, our work to date suggests that radiation damage of the crystal lattice can be the most serious HF limitation of certain SSTR, such as quartz crystals. However, the existence of this specific limitation for HF-SSTR fission rate observations is not a complete deficit. Indeed, on this very basis the concept of SSTR radiation damage monitor can be advanced. For minerals, SSTR characteristics depend intimately upon crystal lattice structure. This lattice structure can, in turn, be dramatically altered at sufficiently high neutron fluence. Hence in HF applications, a number of SSTR attributes have exhibited significant fluence dependence. In particular, track registration as well as bulk and track etch rates undergo substantial change. Consequently, for HF irradiations, any of these SSTR observables afford a unique measure of radiation damage.

These conclusions are obviously qualitative in the sense that considerably more work is required to understand SSTR radiation damage monitors, to define their domain of validity and to learn how to apply them in HF reactor irradiations. In addition, it is also important to correlate quartz crystal SSTR results with changes observed in volume or density due to irradiation-induced swelling. In fact, quartz crystals have been used in this manner for fluence measurements for some time.⁽¹¹⁾

These efforts overlap the need to rigorously define HF limitations of SSTR for fission rate measurements. With respect to such fission rate measurements, it must be emphasized that no final assessment is possible on the basis of the very limited work performed to date. Although our fondest hopes are now somewhat tempered, in-core HF fission rate measurements with SSTR would appear to be feasible for fast neutron fluences up through at least 10^{20} n/cm², which therefore still provides a rather broad domain of applicability for HP reactor environments.

As an example of current HF efforts, Figure 5 displays an advanced dosimetry capsule prepared for irradiation in the Oak Ridge Research Reactor-Pool Side Facility (ORR-PSF). The ORR-PSF is a controlled benchmark field used to simulate a Light Water Reactor-Pressure Vessel (LWR-PV) environment. It can be seen that the relevant dosimetry and damage monitors have all been incorporated in this advanced capsule. A total of ten such HF dosimetry capsules will be exposed at different locations throughout this LWR-PV mockup.

III. RESPONSE CHARACTERISTICS OF CR-39

Proton and alpha-particle irradiations have been carried out⁽⁷⁾ to determine the track registration properties of CR-39 polymer, a newly discovered solid-state track recorder (SSTR) material.^(12,13) The polymer is thermally set plastic produced from the CR-39 monomer, the oxydi-2, 1-ethanedyl di-2-propenyl ester of carbonic acid by catalysis with di-isopropyl peroxydicarbonate. The resulting polymer has a number of properties that are advantageous for reactor dosimetry. Notable characteristics include uniformity of bulk etch rate and extremely high sensitivity for track registration. In fact, CR-39 polymer will record proton tracks and can be subjected to extensive etching and still retain a smooth surface of excellent optical quality.

The response of CR-39 to alpha-particles was investigated in the energy region 3.2 to 6.1 MeV using isotopic sources of ^{252}Cf , ^{244}Cm , ^{241}Am and ^{148}Gd . Track diameter distributions were measured using a computerized Quantimet 720 system. The track diameter distributions obtained from normally incident alphas showed no energy dependence using an etching procedure of ten hours in 6.25N NaOH at 70°C. Consequently, this characteristic makes CR-39 an outstanding candidate for integral (n, α) reaction rate measurements. However, recent work with substantially different etching techniques has revealed that CR-39 can possess alpha-energy dependence.⁽¹⁴⁾ Hence, for observation of (n, α) reactions, it appears that CR-39 can be used for differential as well as integral measurements.

The response of CR-39 to normally incident protons in the range between 1 and 7 MeV is shown in Figure 6. Note the obvious energy dependence of the track diameter. These tracks were etched in 6.25N NaOH for 16 hours at 70°C. Typical proton track diameter distributions obtained by observing several hundred tracks with the Quantimet are displayed in Figure 7. The fact that these distributions are quite narrow, ~5% relative full width at half maximum (FWHM), demonstrates that CR-39 can afford excellent proton energy resolution.

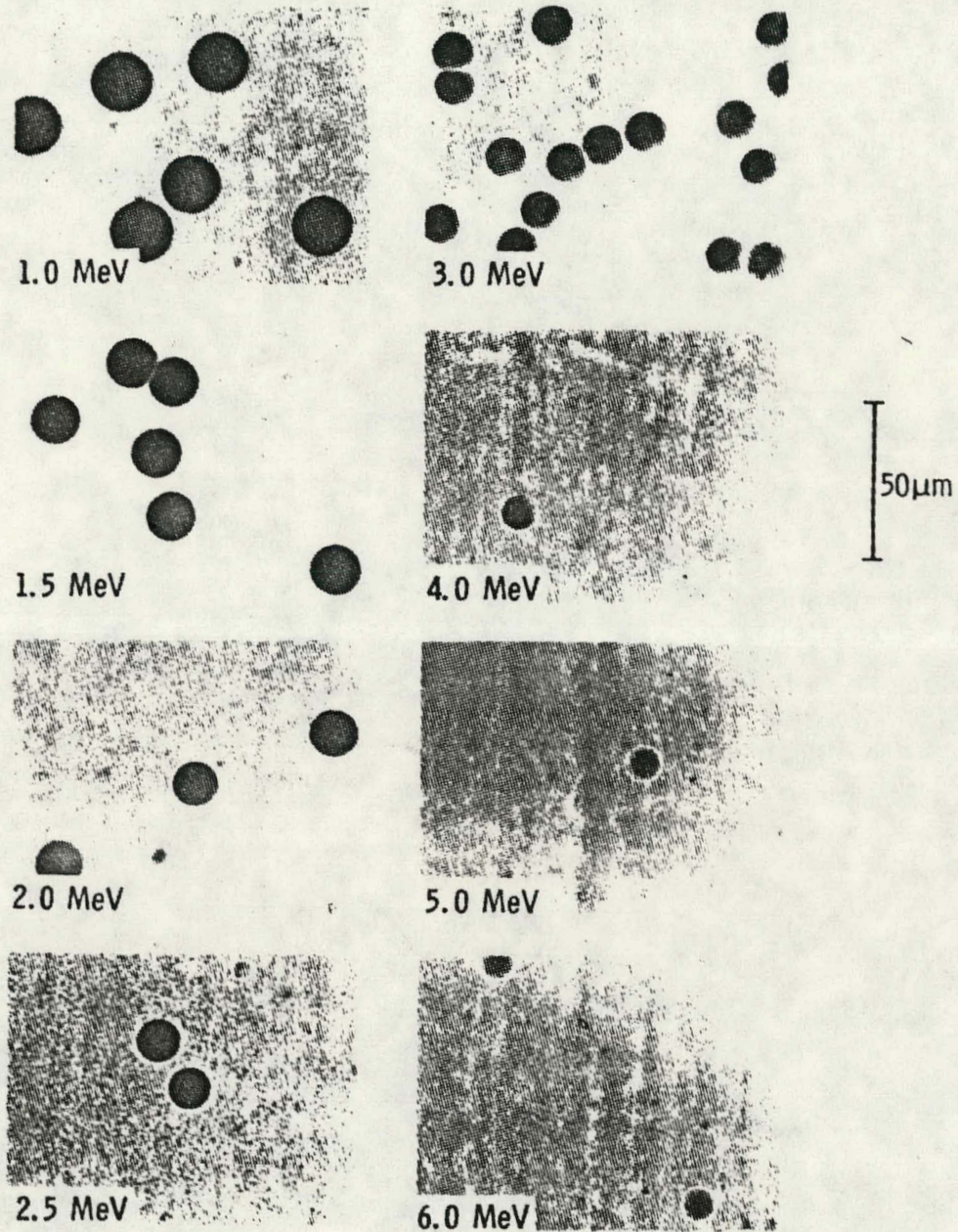
Figure 8 summarizes our proton response investigations of CR-39 to date. Note that the proton energy sensitivity extends to at least 18 MeV. The excellent energy resolution of CR-39 coupled

with this unexpectedly large energy range of proton sensitivity opens up new realms of SSTR applicability in reactor dosimetry. In-situ proton-recoil fast neutron spectrometry for reactor dosimetry as well as energy sensitive techniques for passive fast neutron personnel dosimetry are two such examples. Applications of CR-39 for spectrometry and radiography in magnetic fusion energy (MFE) environments are described in a companion paper presented at this symposium. (15)

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PROTONS



16 HR AT 70°C

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FIGURE 6. The Remarkable Energy Dependent Behavior of the Track Diameter in CR-39 SSTR Due to Normally Incident Protons.

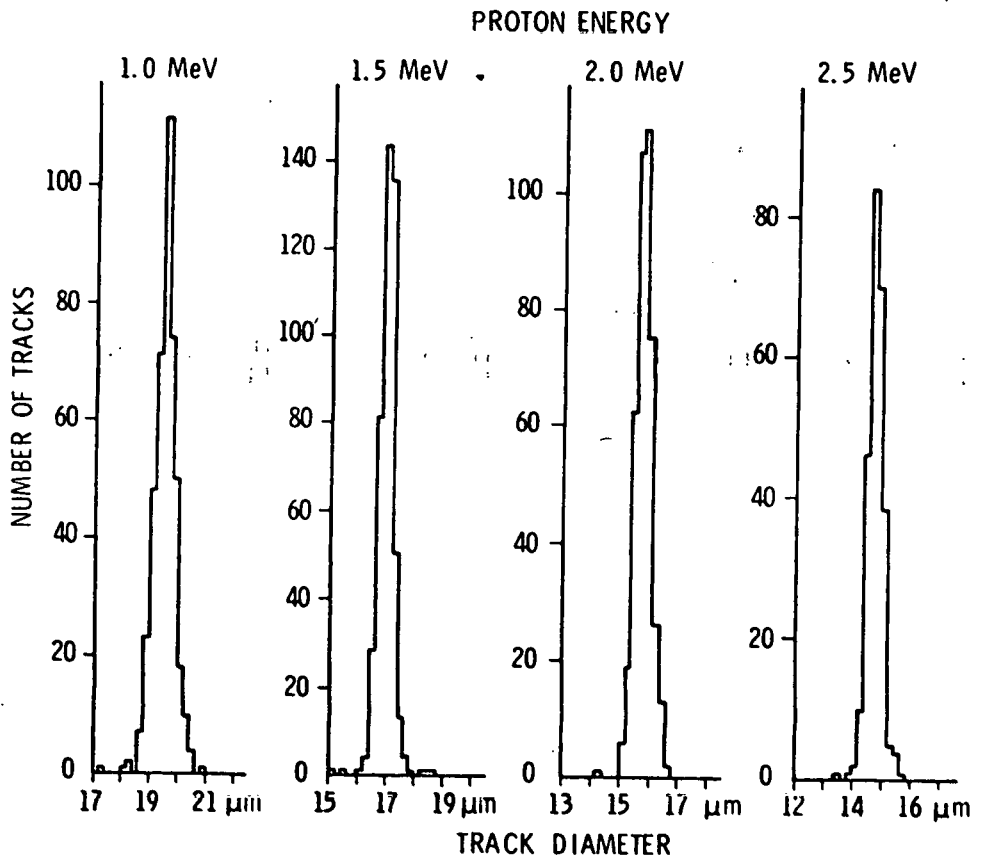
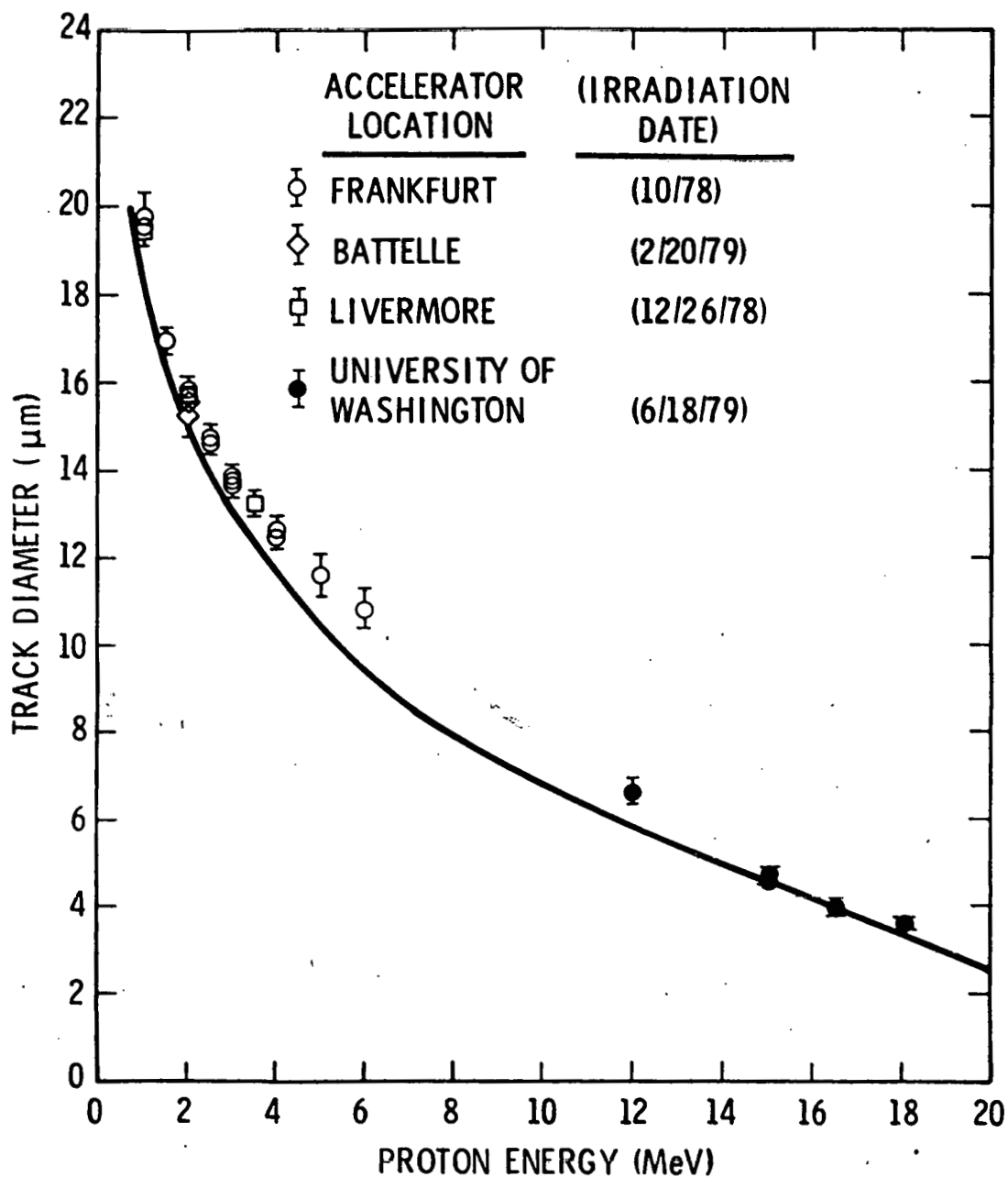


FIGURE 7. Track Diameter Distributions Observed in CR-39 SSTR for Normally Incident Monoenergetic Protons.



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FIGURE 8. Summary of Experimental Calibration Results to Date for the Variation of Proton Track Registration Diameter with Proton Energy in CR-39 for Normally Incident Protons.

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