ADVANCES IN THE DEVELOPMENT OF CR-39 BASED NEUTRON DOSIMETERS

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A combination thermoluminescent dosimeter (TLD) and track etch dosimeter (TED), which can be used for detecting neutrons over a wide energy range, has been developed through recent research in passive neutron dosimetry. This dosimeter uses Li-600 TLDs to detect thermal and low energy neutrons reflected from the body, and the TED polymer of CR-39[a], to detect fast neutrons from proton recoil interactions with the polyethylene radiator or with CR-39 itself. Some form of the combination dosimeter is currently in use at several U.S. Department of Energy (DOE) facilities, and its use is expected to expand over the next year to include all DOE facilities where significant neutron exposures may occur. The extensive research conducted on the TED component over the past six years has continually focused on material improvements, reduction in processing time and dosimeter handling, and ease of sample readout with the goal of automating the process as much as possible.

Material Improvements

Since the beginning of DOE's involvement in track detector development, many improvements have been made in the materials and sample processing. Of the track detector materials available, CR-39, the highly cross-linked polymer of allyl diglycol carbonate, is the best material found to date for neutron detection over a wide range of energies. In recent years a higher purity of CR-39 monomer has been available and has led to a polymer with greatly improved detection consistency and reduced background tracks. With a nominal purity of 93%, this monomer has been dubbed "dosimetry grade." Anticipating that higher purity material may prove even better, further purification on a small scale led to almost nonexistent background on the resulting samples. A larger scale demonstration is underway to determine whether an ultra-high-purity material is worthwhile.

It was believed that the neutron damage of CR-39 was occurring at the polymer cross linkages. Researchers at the University of Connecticut, in conjunction with the Pacific Northwest Laboratory (PNL), provided experimental evidence that proved this was the case. They encased CR-39 into sealed aluminum containers and exposed the samples to several hundred thousand rad. Chemical analysis of the exposed samples indicated the presence of diethylene glycol and led to the conclusion that the carbonate bonds in the side-chain cross linkages were the sites of the majority of neutron-induced damage. With this in mind, they set out to add a second component to CR-39 with more proton-sensitive functional groups

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to make a CR-39 copolymer that is more neutron-sensitive. Chemical bonding and mechanical problems have prevented real success to date, but the efforts continue to develop a more sensitive material.

In addition to material improvements, current research goals are to expand the neutron energy range of CR-39 with the use of specialized radiators and process changes.

Processing Improvements

Processing of CR-39 was initially done by chemical etching and then by a chemical/electrochemical etch. A process using only electrochemical etching at elevated temperatures has been developed that shows a superior efficiency in track detection and has a nearly flat energy response curve over the energy range from about 150 keV to about 3 MeV. Considerable optimization of the electrochemical process has led to selecting parameters that include a two-cycle etch.

For this process, sample chambers are preheated in an oven at 60°C and a 6.5 N potassium hydroxide solution is added to the chambers to begin the etch. The first cycle is operated at about 47,000 V/cm of material thickness and a frequency of 60 Hz for 5 hours. The second cycle uses the same voltage but raises the frequency to 2000 Hz. This cycle runs for 23 min. The samples are then ready for track counting. This process produces 7 to 9 tracks/cm^2-mrem (700 to 900 tracks/cm^2-mSv) over the most efficient portion of its energy range from about 150 keV to 3 MeV.

The sample chamber design has also been greatly improved over the past few years from single cell chambers to 8- and 24-cell chambers. These multi-cell chambers have facilitated processing large numbers of badges and reduce the variation in the microenvironment surrounding the samples. A programmable power supply has been developed to keep the voltage steady and automatically change the frequency at the appointed time.

New image analysis systems provide many more options in sample readout. Optical systems capable of providing a direct count of the sample numbers and digitizing imaging systems capable of also analyzing the size and shape of the tracks are currently in use. The extent of automation varies from track counting only to automated stage movement, sample hoppers, autofocusing of the microscope, and sophisticated software to interface the components and manage the data files.

Track Size Distributions

With the use of automated track counters with size discriminators, it has been possible to evaluate the track size (area) distributions as a function of energy. A difference in track size with energy has been theorized by many researchers, and it is evident from observing tracks from monoenergetic sources versus continuous energy sources that differences in track size exist. Evaluation of samples from a series of

monoenergetic exposures shows such differences exist. Histograms of the shape of the size distribution curve changes in an orderly pattern with energy (Fig.1). A pattern developed going from a single peak to a bimodal formation and then back to the decreasing size distributions. The exposures represented in the figure were about 200 mrem (2 mSv) but fluences were not identical and the graphs were not normalized. Therefore, their shapes, not their magnitudes, are important. Not all of these are specifically distinguishable from each other, and it is too early to tell if they truly have identifying signatures. The 144 keV and 2.8 MeV results, for instance, look very similar.

With too little dose, a pattern may not be evident. With high doses, the tracks become deformed, smaller, and too uniform for analysis. It is not yet known how samples exposed to several spectra may be interpreted. Work is continuing to clarify the track-size distribution relationship and to determine whether it may have any benefit in evaluating dosimeters for spectra to which the personnel were exposed.

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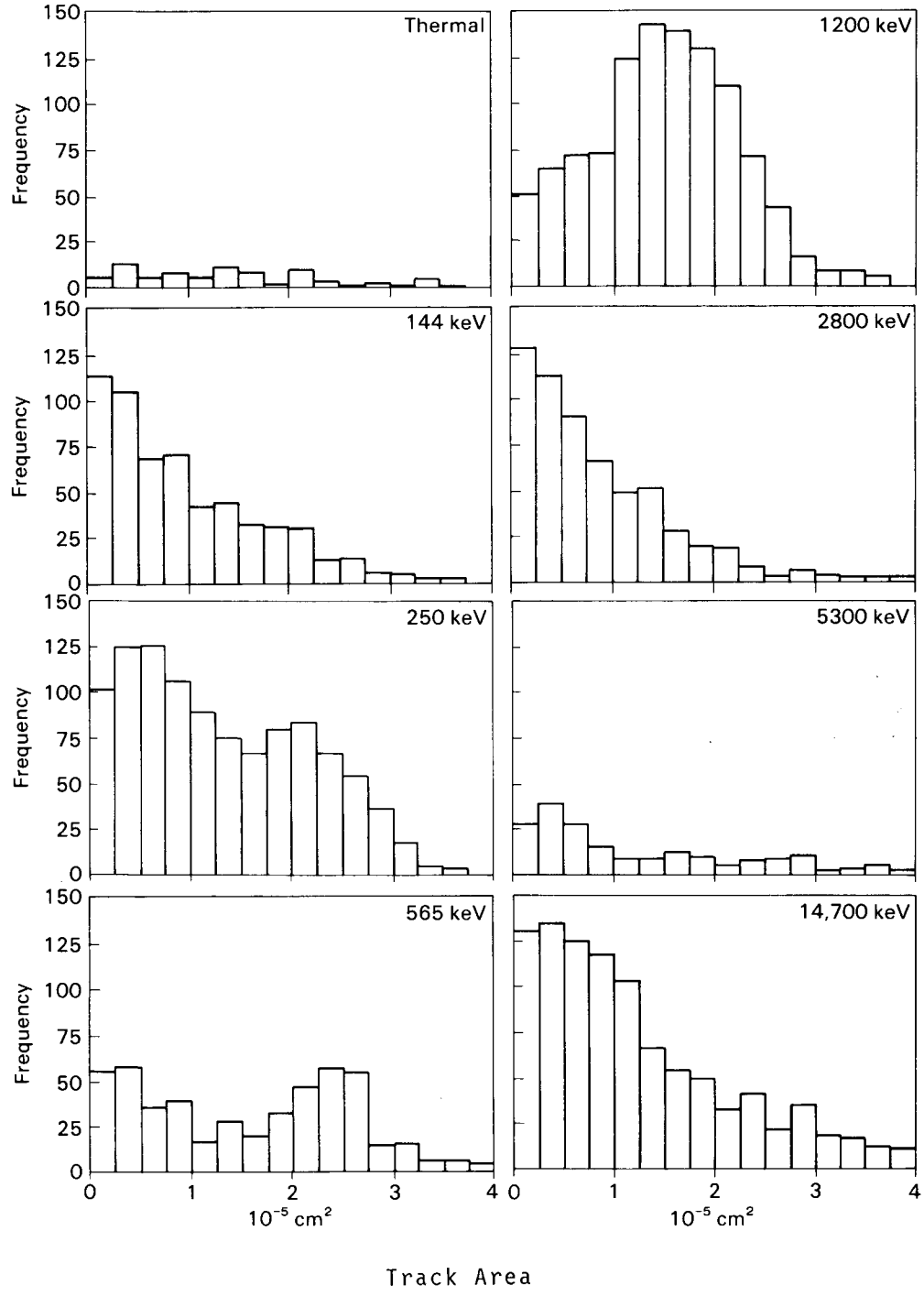

FIGURE 1. Track Size Distribution Histograms for Indicated Neutron Energies (not normalized for fluence)