LET Effects on the Chemical Resistance of Irradiated Polymers

It has been known for many years that radiation-induced changes in the properties of organic polymers depend not only on dose, but also on the linear energy transfer (*LET*) of the radiation, if competing reactions of different kinetic order occur. Usually, this *LET* dependence amounts to a relatively small increase in the effectiveness of high *LET* radiation by less than a factor of 2 or 3 with regard to different mechanical, optical, or chemical radiation effects.[1-4] It is also known that particles exceeding a certain threshold *LET* (>1 MeV/cm²/g, corresponding to the *LET* of an alpha particle less than several MeV in energy, in cellulose nitrate; more in less sensitive polymers) may create enough damage in a cylindrical region along their tracks to increase the chemical etchability in this region. This preferential etching has been used widely as a method for nuclear track visualization in polymers.[5-8] It has, however, not been realized that this effect may lead to an exceedingly high *LET* dependence of the bulk chemical resistance of irradiated polymers.

The bulk etch rate of cellulose nitrate and polycarbonate foils in KOH is increased after gamma radiation exposures in the 10^7- to 10^9-rad range.[9,10] In order to study the *LET* dependence of this effect, foils of four different polymers have been irradiated under secondary particle equilibrium conditions with different doses of gamma radiation from a ^{60}Co source, and with fast fission neutrons from the unshielded Health Physics Research Reactor. After irradiation, they were etched with 28% KOH at elevated temperatures: (1) cellulose nitrate (15 μ, Hercules Pyro Smokeless) at 40°C; (2) cellulose triacetate (500 μ, Triafol T, Bayer) at 60°C; (3) polyethylene terephthalate (13 μ, Mylar, Kimberley Clark) at 60°C; and (4) dioxidiphenylpropane polycarbonate (10 μ, Makrofol or Kimfol, Bayer and Kimberley Clark) at 60°C.

The increase in etching speed is expressed in per cent weight loss per hour of etching in excess of that of the unirradiated control films and plotted as a function of dose (Fig. 1). Obviously, the neutron dose required for a given foil weight reduction is by a large factor less than the gamma dose. The factor depends on dose because the neutron radiation damage is a supralinear function of dose.

The considerable *LET* dependence may be explained by preferential etching along recoil particle tracks which are produced by elastic neutron interactions throughout the polymer. Recoil protons (with the possible exception of those close to the Bragg peak in cellulose nitrate) are below the preferential etching threshold, but recoil carbon, oxygen, and nitrogen nuclei are above it in all the polymers investigated. Based on previous experiments[11,12] concerning the production of etchable recoil particle tracks in polymers by fission neutrons, yielding $\sim 0.3 \times 10^{-5}$ track/n in Makrofol and $\sim 1.1 \times 10^{-5}$ tracks/n in cellulose nitrate, we estimate the density of recoil tracks to be about 10^7/cm² on each plane perpendicular to the direction of neutron incidence throughout the foils after exposure to 5000 rad of fission neutrons. Assuming a diameter of several μ for each of the preferential etch pits after completion of the initial etching phase (with new tracks becoming accessible to the etchant continuously as the etching proceeds), a considerable increase in the bulk etching rate is understandable.

The rapid breakdown of polymers under attack by aggressive chemicals after exposure to particles which either have a high *LET* or produce high-*LET* particles by interaction with the polymer may be of some interest with regard to the resistance of polymers in space radiation environments, nuclear weapons radiation effects, and in nuclear reactors or accelerators. Perhaps there are also applications to charged particle dosimetry. Because of this effect, fast neutron-induced autoradiography[13] using polymer track detectors and the spark-counting technique are limited to a rather low fast-neutron flux.

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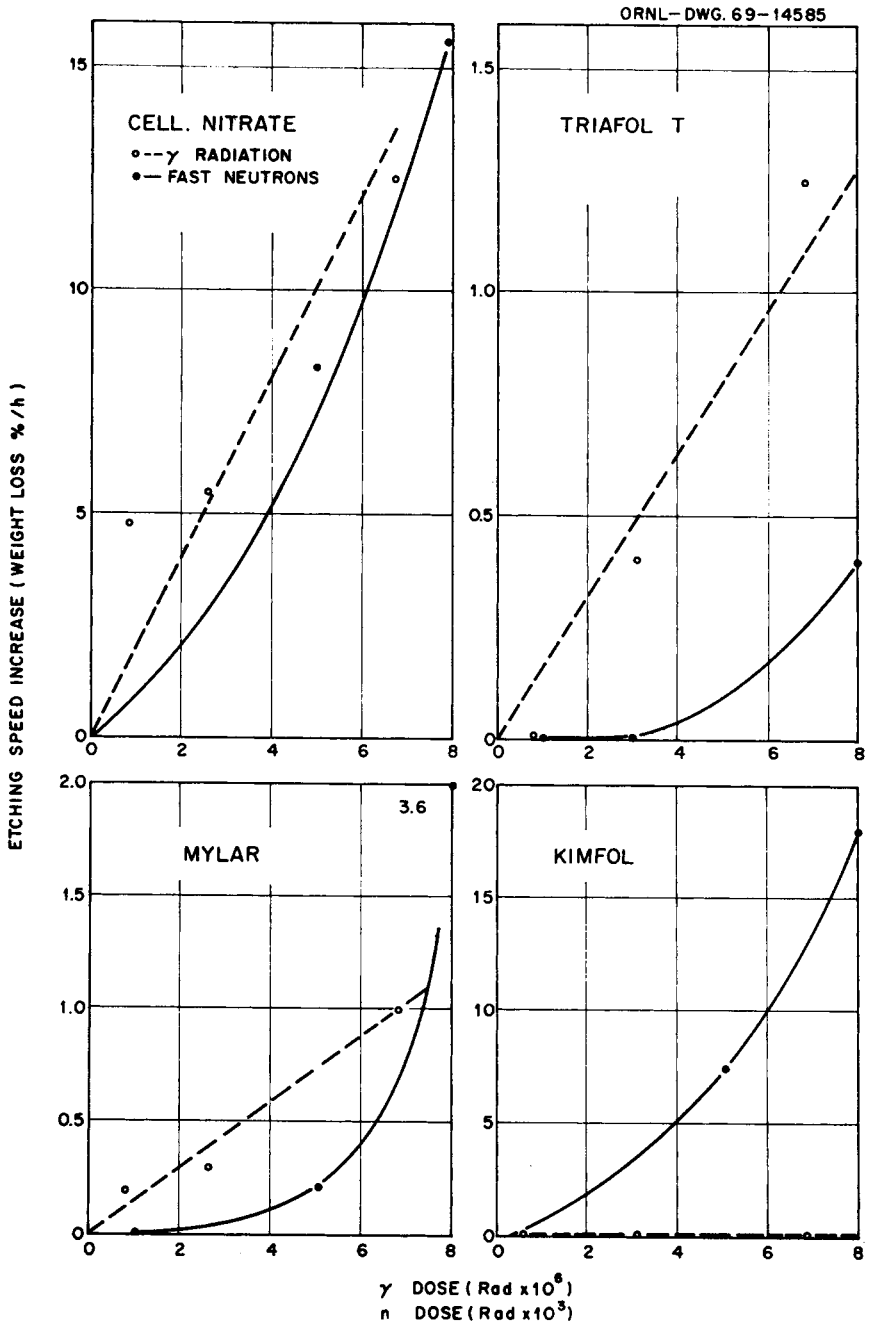

Fig. 1. Increase in etching speed (weight loss in per cent per hour of etching in warm 28% KOH) as a function of fast neutron (krad) and gamma radiation dose (Mrad) in four different polymer foils.

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