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THE EFFECT OF AIR ON THE RADIATION-INDUCED DEGRADATION OF POLYTETRAFLUOROETHYLENE (TEFLON)

W.W. Parkinson
W.K. Kirkland

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ABSTRACT

The tensile properties of polytetrafluoroethylene (Teflon) specimens of 1/32- and 1/16-inch thickness have been measured before and after gamma irradiation in air and in vacuum. The effect of air was to reduce by about an order of magnitude the dose required for a given fractional decrease in original properties. There was little effect of thickness over the range studied. In both air and vacuum, the tensile strength decreased rapidly to 40-45% of its original value and then remained relatively constant with increasing dose, up to 10^7 rads in the case of air. The elongation-at-break showed a maximum at 10^4 rads followed by a sharp decline to 0.1 inch/inch at 10^6 rads for air irradiations.

Polytetrafluoroethylene (Teflon) is one of the most resistant plastics known to corrosive liquids and gases and to high temperature. In contrast, when subjected to ionizing radiation, it is one of the most rapidly degraded plastics.[1] It has been found also that the rate of degradation is reduced appreciably if the material is in an inert atmosphere rather than in air.[2,3] The effect of air or oxygen would be expected to depend on the local concentration of oxygen within the material, hence to depend on both thickness and dose rate. Since the earlier work[2] had involved thin films which would be extremely sensitive to atmosphere and since polytetrafluoroethylene is used so extensively, this investigation was undertaken on stock of a thickness common in ordinary usage.

Polytetrafluoroethylene is not soluble and melts only with difficulty so molecular weight measurements were not attempted as a measure of the effect of radiation. Instead, tensile strength and elongation-

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[1] C. D. Bopp and O. Sisman, Nucleonics, 13 (7), pp. 28 (1955).
[2] L. A. Wall and R. E. Florin, J. Appl. Polymer Sci., 2, 251 (1959).
[3] R. W. King, N. J. Broadway, and S. Palinchak, REIC Rept. 21, Battelle Memorial Institute, Columbus, Ohio (1961).

at-break were used as indications of degradation. These properties are not only dependent on molecular weight but are also meaningful for the serviceability of the material. Others have used the zero-strength-time as a criterion of radiation damage.[4-7] This property, the time to rupture for a standard stressed specimen after insertion into a preheated furnace, is more simply related to molecular structure than the tensile strength at ordinary temperatures but is less significant for engineering purposes.

In this investigation, commercial sheet Teflon of 1/32- and 1/16-inch thickness was exposed to ^{60}Co gamma radiation in air and in vacuum at a dose rate of 1.2×10^6 rads/hr. The specimens were a reduced version of standard dumbbell tensile specimens, and had a gauge section 1/4 x 3/4 inch with the gauge marks 1/2 inch apart. The specimens irradiated in vacuum were evacuated at 140°C to 5×10^{-6} torr and sealed in borosilicate glass, 4 or 5 to a capsule. The temperature during irradiation was about 25°C. The dose rate of the ^{60}Co gamma source was measured by $Ce(SO_4)$ solution (0.4 M H_2SO_4) which had been standardized versus $FeSO_4$ assuming $G(Fe^{+3}) = 15.6$ ions per 100 ev.

The tensile strength and elongation-at-break of the Teflon specimens are plotted as functions of radiation dose in Figs. 1 and 2, in vacuum and in air respectively. The results of Wall and Florin[2] on film are shown for comparison.

It is evident from the figures that air accelerated the radiation degradation significantly, decreasing by nearly an order of magnitude the dose required to reduce the strength and elongation to half their original values. This accelerating effect of air does not seem to depend on thickness at ~10^6 rads/hr. However, the thin films of Wall and Florin (Fig. 2) lost their tensile strength at a very low dose (4×10^6) in air, compared with our 1/32-inch specimens (>8×10^7 rads).

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[4]A. Nishioka, M. Tajima, and M. Owaki, J. Polymer Sci., 28, 617 (1958).
[5]K. Matsumae et al., J. Polymer Sci., 28, 653 (1958).
[6]A. Nishioka et al., J. Appl. Polymer Sci., 2, 114 (1959).
[7]L. A. Wall and R. E. Florin, "Irradiation of Fluorocarbons," ARL-62-350, Office of Technical Services, Department of Commerce, Washington, D. C. (1962).

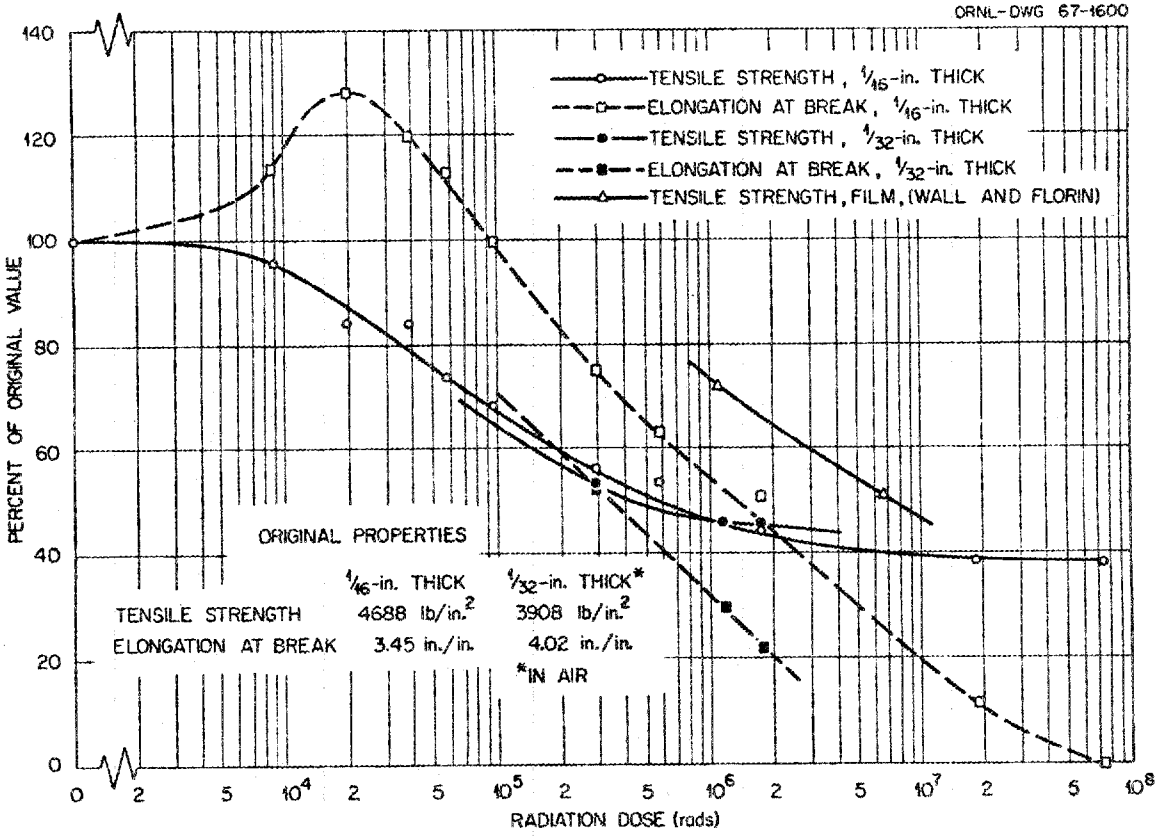

Fig. 1. Teflon, Irradiated in Vacuum.

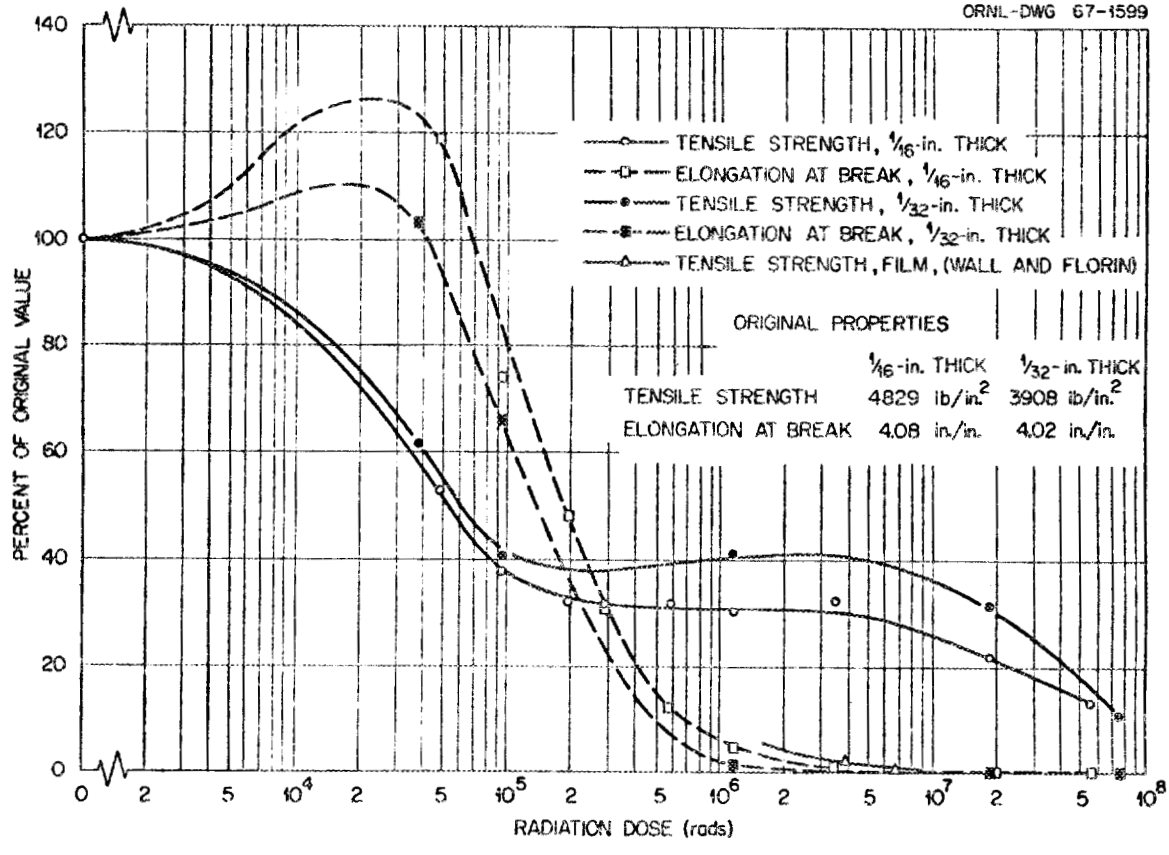

Fig. 2. Teflon, Irradiated in Air.

For engineering purposes, it is significant that the tensile strength of Teflon irradiated in a vacuum decreases to 40-45% of its original value at about 10^6 rads, but remains almost constant from this dose to perhaps 10^8 rads or higher. Although the ductility decreases more rapidly than the tensile strength, useful mechanical properties (elongation > 10%) are retained to doses greater than 5×10^6 rads. At the higher rate of degradation in air, the tensile strength drops to 40% of original at about 10^5 rads and the elongation-at-break drops below 0.1 inch/inch at about 10^6 rads.

The increase in the elongation-at-break in the low dose region is interesting although its technical usefulness is questionable since this property of Teflon is adequate without irradiation for most applications. The molecular or structural changes resulting in the maximum in the elongation are not obvious since the zero-strength-time measurements of Nishioka and coworkers[4,6] indicate that scission is the dominant process even at doses as low as 10^3 rads. These workers did observe, however, a minimum in the density at about 10^3 rads followed by a monotonic increase. In the region of increasing density, an increase of crystallinity was also observed. It was concluded that the increase in density resulted from the increasing crystallinity which was an outgrowth of greater molecular mobility accompanying chain scission.

It seems credible that the initial chain breaks constitute crystal defects giving the decrease in density before the molecular mobility reaches the point which permits crystal growth. Then it may be speculated that at low doses the crystal defects increase intracrystalline slip under stress to give the observed increasing elongation-at-break in this dose region.

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