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Re: An Overview of Basic Radiation Effects on Polymers and Glasses

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An Overview of Basic Radiation Effects on Polymers

In general, most polymers can tolerate radiation quite well, with very low doses in the range of 10 to 100 Gy (1 krad to 10 krad) causing little to no degradation in mechanical properties and/or performance. The focus of this memo is to highlight some general trends based on a literature review for a number of polymers at varying degrees of radiation exposure. In most cases, the polymers discussed can also tolerate moderate doses between 1 kGy and 10 kGy (0.1 and 1 Mrad) with little to no consequence. In all cases where polymeric materials are exposed to radiation it is recommended to consult a subject matter expert (SME); this is especially important for polymer applications in radiation environments close to their "tolerance level," where a more detailed radiation study may be warranted.

An Overview of Basic Radiation Effects on Glasses

Literature reveals that glasses undergo densification and other attendant physical property changes when exposed to radiation. The threshold for damage in pure silica is close to 100 Mrad, while for multicomponent glasses, it is certainly lower. While several studies show no changes in physical properties such as strength, toughness, and thermal expansion in the 10 – 100 krad range, one study shows a 20% strength loss in soda-lime glass as low as 8.8 krad exposure. There are no reports of radiation studies on multi-component glasses of interest in NW; however, it is safe to assume that their damage threshold would be significantly lower than that for pure silica, ie., <<100 Mrad. Any change to the mechanical and elastic property would lead to some changes in the residual stress states in the glass components. Loss of strength observed in some studies could lead to cracking even if the residual stress state itself does not change. It is likely that any radiation effects in our applications will be negligible (save for some darkening). However, because data on materials to be used in the seals are non-existent, it is recommended that a materials-based study be commissioned to explore these effects. In this study, commonly used glass and glass-ceramics should be tested in the GIF, and relevant properties such as density, thermal expansion, elastic moduli, strength, and toughness should be monitored as a function of dose.

Table of Contents

1. Polymers

1.1. Introduction

The dominant driving forces for active polymer radiation chemistry research include advances in nuclear energy capability/reliability,[1] super-conducting fusion magnet technologies,[2] medical device sterilization,[3] and space applications. As such, extensive efforts are ongoing that aim to investigate neutron and gamma radiation effects on organic material performance in application relevant environments. To this end, numerous articles, books, and presentations exist in the literature that identify and discuss radiation effects on many different classes of polymers and polymer composites.[4-6] The intent of this memo is to concisely discuss some of the current/accepted findings, as well as some recent unpublished data regarding radiation effects on organic materials of interest to the Department of Energy complex. A number of tables and graphs are copied directly from the literature.

Fig. 1 shows a general overview of radiation-induced polymer behavior for the main classes of polymers (for example, thermosets, thermoplastics, and elastomers) and helps set the tone for this memo.[6] Based mostly on ultimate elongation or similar mechanical data, all of the different classes of polymers listed in Fig. 1 exhibit little property degradation from radiation exposure for doses between 1 and 10 kGy (0.1 to 1 Mrad).[6] One notable exception is Teflon when irradiated under air at 1 kGy. For clarity, all radiation units presented in this memo are SI units, where 1 Gy is equivalent to 100 rad. This figure is intended as a general guide for relative comparisons of materials. The data were taken from numerous literature sources and represent approximate radiation tolerances of individual polymeric materials exposure to simulated non-oxidizing and more highly oxidizing conditions. It should be noted also that rough numbers are usually discussed for radiation effects on polymers, as there is a high degree of scatter in the data; thus tables and publications usually mention orders of magnitude exposures. Several factors play a role in measuring radiation resistance, including sample-specific parameters (formulation, thickness), exposure parameters (dose rate, temperature, environment), and measurement parameters (time elapsed post-irradiation). In most cases, the mechanical property considered was tensile elongation at break. Where elongation data were unavailable, some other important mechanical property data, such as bend strength, was considered. Data were taken at a variety of high dose rates, primarily in the range of 10^4 to 10^5 Gy/hr or above, and within or near a lower dose rate range of 5 to 50 Gy/hr, with exposure in air; sample thicknesses were primarily in the range of 0.4 to 1.5 mm; and samples were irradiated at, or slightly above, room temperature. It is not clear how diffusion limited oxidation (DLO) or dose rate effects were taken into consideration. Mechanical properties were usually measured shortly after irradiation was completed.

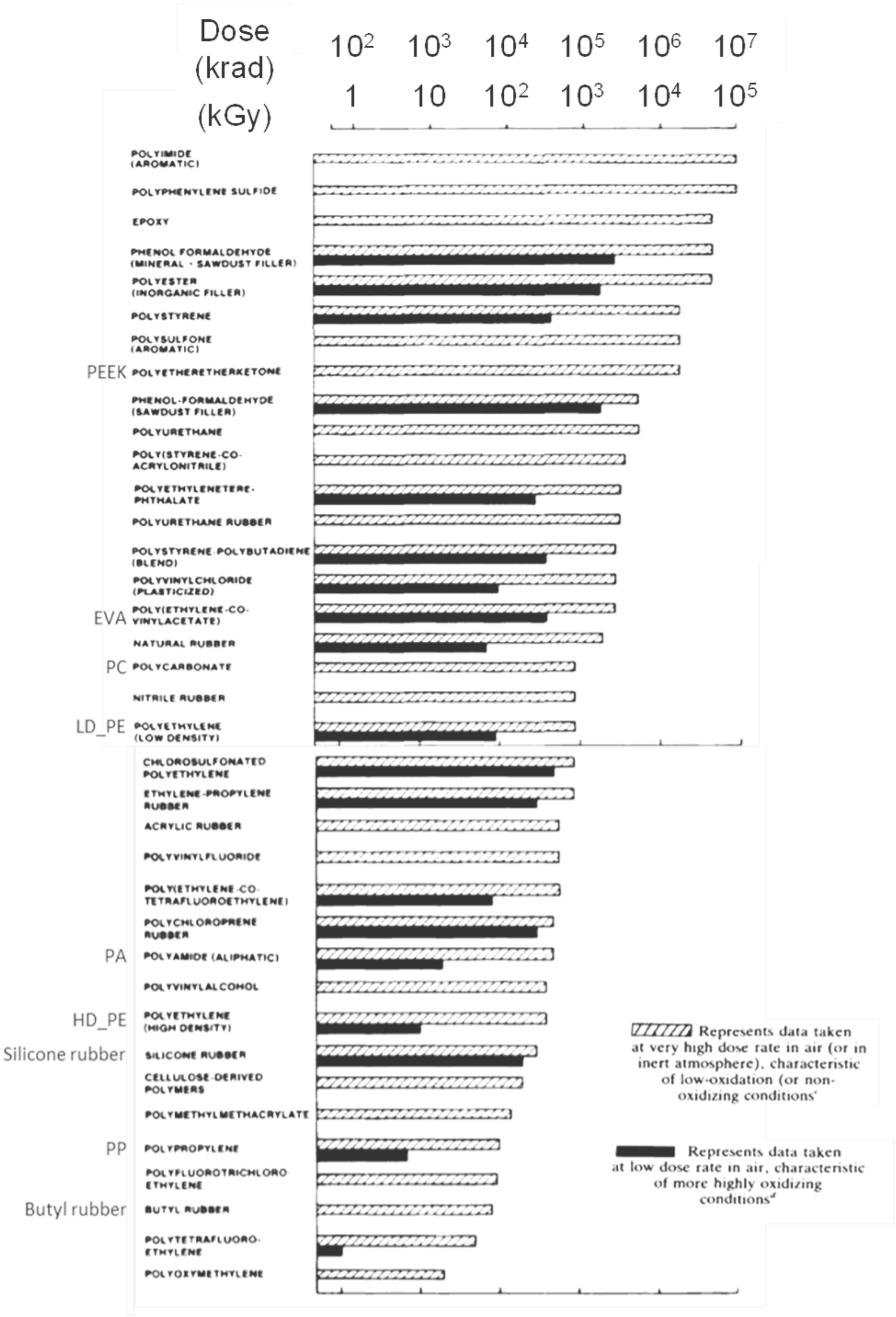


Fig. 1. Data from numerous literature sources showing approximate radiation tolerances of individual polymeric materials for exposures simulating non-oxidizing and more highly oxidizing conditions.[6]

1.2. Aliphatic Polymers

Aliphatic polymers are a class of thermoplastic materials that includes straight carbon chains, branched chains, and non-aromatic rings. Some key aliphatic polymers are polyolefins like polyethylene (PE), polypropylene (PP), ethylene propylene, and others. In general, aliphatic polymers have reasonable radiation tolerance, as evidenced by Fig. 1.[6] However, aliphatic polymers are generally not as radiation-resistant compared to those that incorporate aromatic rings,[6] like polyether ether ketone (PEEK)[7] or the class of epoxy thermosets.[8,9]

1.2.1 Polyethylene (PE)

Polyethylene (PE) (see structure in Fig. 2) is a widely employed polyolefin. PE is commonly used, in varying formulations, as an insulating material for low and medium voltage nuclear power plant cables; here radiation is part of PE's normal environment. As a result, a significant amount of research discusses PE performance, or more often cross-linked PE (XLPE) to varying extents of degradation (for example, to varying doses at different dose rates).[1,10-12] In short, PE can tolerate on the order of 10 kGy (1 Mrad) or more depending upon the thermal environment, its crystal structure (that is, amorphous, semi-crystalline, or highly crystalline), cross-link state, and the application specific performance requirements. At ambient temperatures the mechanical performance (for example, tensile elongation or tensile strength, among others) is quite good, despite radiation exposure.

$$\sim\left[\mathrm{CH_2CH_2} \right]\sim_n$$

Fig. 2. Chemical structure of polyethylene.

As an example, Fig. 3 shows tensile elongation data as a function of dose (^{60}Co) for a cross-linked medium to high-density ethylene-vinyl acetate co-polymer that is used as an insulation material for low-voltage nuclear power plant cables. It is quite evident that little to no degradation occurs for this polyolefin until the total exposure dose nears ~50 kGy (1 to 5 Mrad). Similar behavior has been routinely observed for other polyolefins, such as ethylene propylene rubber (EPR).[1,11,13,14] These data suggest that polyolefins are quite resistant to high (on the order of multiple Mrad) radiation exposure at a broad range of temperatures.

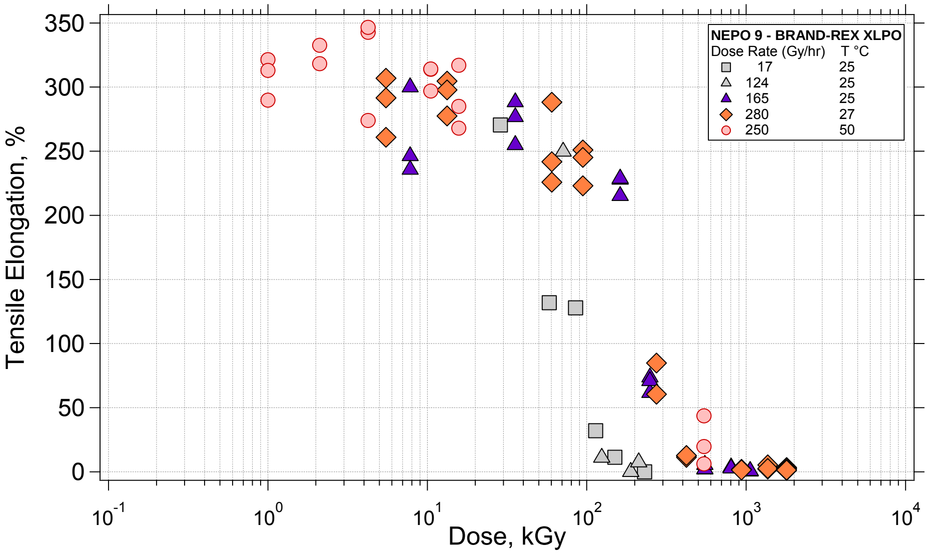
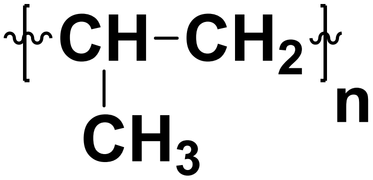


Fig. 3. Tensile elongation data for a cross-linked ethylene-vinyl acetate (EVA) co-polymer cable insulation irradiated at varying dose rates and temperatures.[15]

1.2.2 Polypropylene (PP)

Another type of polyolefin, polypropylene (PP), is also tolerant to high doses of radiation, similar to PE. The chemical structure of PP is shown in Fig. 4. One particular application of PP where radiation tolerance is critical is in the medical industry, where gamma radiation is used during the sterilization process.[3] Recently, researchers (Gonzalez and co-workers from within the reference[3]) subjected injection-molded PP test coupons to continuous irradiation (up to ~2 yr) in air and then performed mechanical testing as a function of accumulated dose.[3] Fig. 5 shows the stability of injection molded PP coupons subjected to manual bending, where unacceptable performance was noted to be a loss of 40% of the original properties (that is, $f/f_o < 0.6$). The data clearly show that PP performed acceptably up to ~40 kGy (4 Mrad) over a 700-day time period. The fact that PP is very resistant to radiation up to ~10 kGy (1 Mrad) is widely accepted, noting similar behaviors in the literature.[16-18]



$$\left[\mathrm{CH}(\mathrm{CH_3}) - \mathrm{CH_2} \right]_n$$

Fig. 4. Chemical structure of polypropylene.

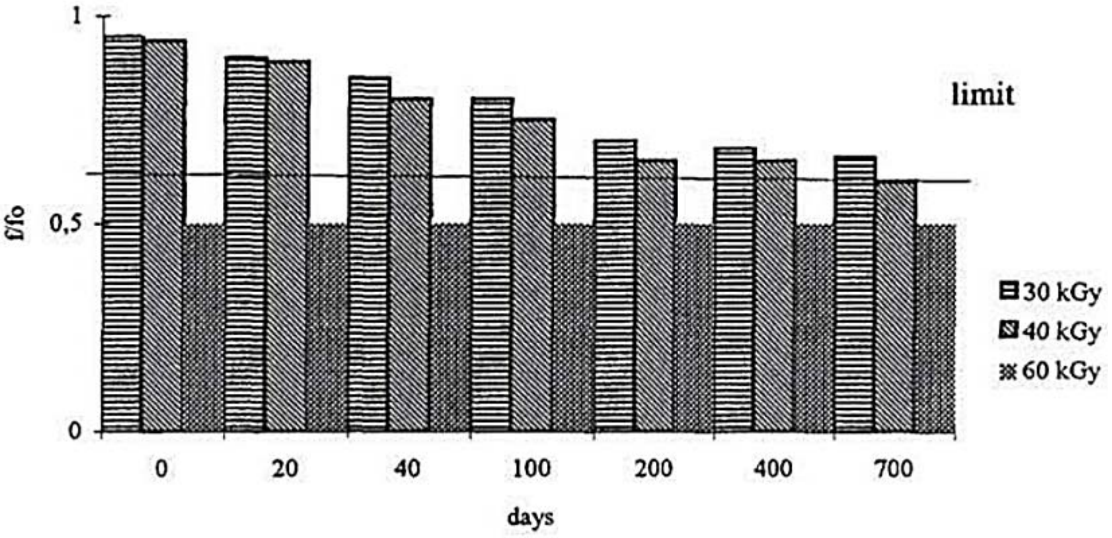


Fig. 5. Stability of injection molded sheets of PP irradiated to 30 and 40 kGy.[3]

1.2.3 Butyl Rubber

Butyl rubber is a very common elastomer that is often used as an environmental seal, O-ring, and even as the insulating material for electrical wires. Because of the widespread use of butyl rubber, an abundance of aging data exist under a multitude of accelerated aging and environmental exposure conditions in varying form factors.[19-21] Recently Scagliusi et al. studied the radiation-induced degradation of butyl rubber with a ^{60}Co source at 5 kGy/hr up to ~200 kGy (20 Mrad).[22] This work likely simulated non-oxidative degradation processes because of DLO and showed that as the radiation dose increased, the tensile strength, tensile elongation (Fig. 6), and durometer (hardness) all decreased.[22] However, it is important to note, in general, that butyl rubbers will maintain basic mechanical properties to doses of ~10 kGy without "significant" damage (seeFig. 1),[6] consistent with the data presented by Chandra et al. and Scagliusi et al.[21,22]

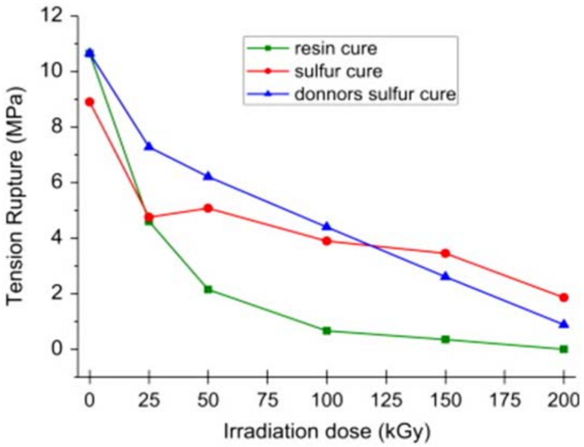


Fig. 6. Tensile elongation at break data for butyl rubber cured with varying agents.[22]

1.2.4 Polyamide

Polyamides (for example, Nylon, Kevlar 29, and others) can be prepared in multiple ways enabling applications to range from textiles (e.g., woven fibers/yarns) to rigid plastics (for example, Nylatron©, Nycast©, etc.). Fig. 7 shows the chemical structure of Nylon 6.6, a very commonly used aliphatic polyamide.

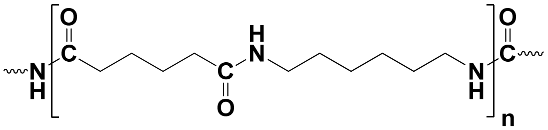


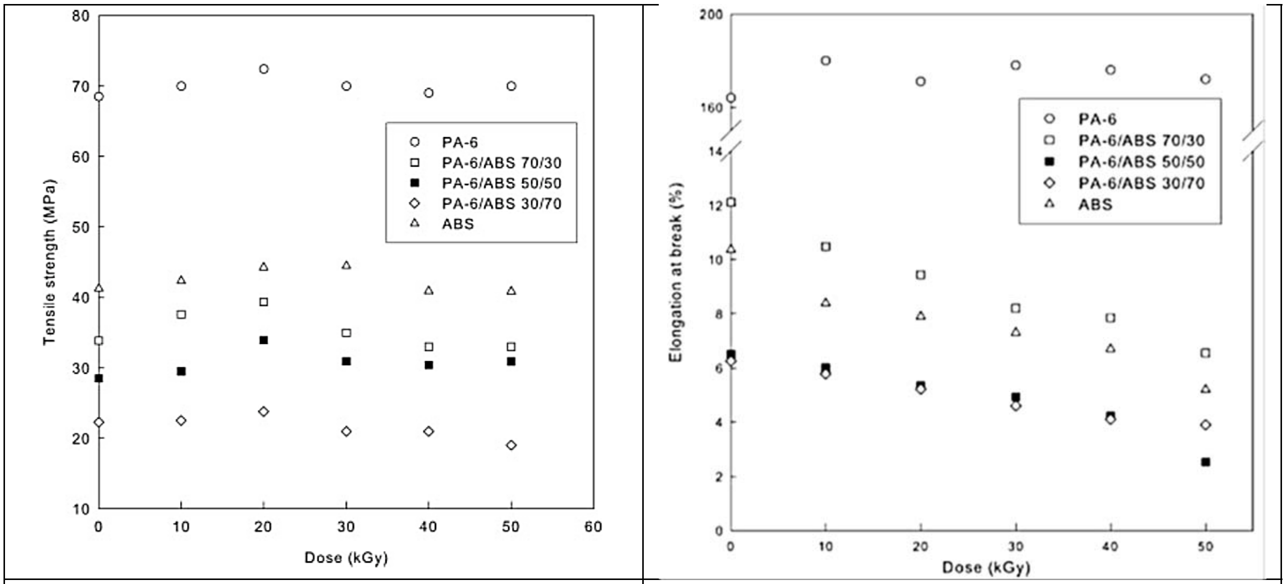
Fig. 7. Chemical structure of Nylon 6.6 (a commonly used aliphatic polyamide).

From a thermal-oxidative perspective, numerous publications exist that discuss nylon aging behaviors. In short, nylon is a relatively robust material.[23-26] Unfortunately, less is known about the mechanical property performance of polyamides in radiation environments. Fadel et al. studied the effect of neutron irradiation on Nylon 6, Dralon© (an acrylic material), and polyester fibers[27] to varying fluence (ϕ). In short, all Nylon 6, Dralon©, and polyester retain their tensile properties (elongation and strength) up to $\phi \sim 10^5$ n/cm^2.

Comparatively, Birkinshaw et al. studied the effect of gamma radiation on Nylon 6.6 for medical implant applications under high dose rate conditions.[28] In short, Birkinshaw showed that doses up to ~25 kGy (2.5 Mrad) resulted in ~5% loss in tensile modulus and yield stress.[28] This mechanical behavior is similar to that discussed for the polyolefins (for example, PE and PP). Further information discussing the radiation effects on polyamides can be found in the literature.[29] Although the discussion of radiation tolerance focuses solely on polyamides such as nylons, it has been widely accepted that as the chemical moieties, or functional groups, that compose the polymer backbone become more aromatic, the general radiation resistance increases;[8] hence, the worst-case degradation sensitivity can be considered for an aliphatic polyamide like nylon.

1.2.5 Acrylonitrile-Butadiene-Styrene

Acrylonitrile-butadiene-styrene (ABS) is a widely used co-polymer because of its ease of processing (e.g., injection molding), light weight, toughness, and chemical resistance. A large portion of the radiation research for ABS plastic has been dedicated to the area of ultraviolet (UV) light and photo-oxidation; for example, ABS is often used in outdoor applications like automotive bumpers. However, Hassan recently published an investigation on the effect of gamma radiation on polyamide 6 (for example, PA-6 or Nylon 6), PA-6/ABS blends, and ABS.[30] As can be seen in the figures, no degradation in tensile strength (Fig. 8(a)) was observed for ABS irradiated up to 50 kGy (5 Mrad, the dose rate was ~5 kGy/hr); however, the tensile elongation decreased by ~40%, indicating that the ABS became more brittle (see Fig. 8 (b)).[30] The relatively high dose rate again suggests that material degradation in this case was explored under non-oxidative and perhaps dominant cross-linking conditions. As shown in this work, ABS plastic performs well at moderate to low doses of radiation; for example, at less than 10 kGy it exhibits no change in mechanical performance. For more details regarding ABS performance, in particular weathering, a comprehensive chapter written by Massey is available in the open literature.[31]



(a) (b)

Fig. 8. Tensile (a) strength and (b) elongation of polyamide 6 (PA-6), PA-6/ABS blends, and ABS as a function of gamma dose.[30]

1.3. Aromatic Polymers

It is generally accepted that as the polymer incorporates more aromatic ring structures (differing from the simpler aliphatic backbones, like PE), the polymer becomes more radiation-resistant.[8] Applications for these materials tend to center around structural needs (composite or composite alternatives) and rigid dielectric materials (for example, rotating knobs, wheels, discs, electronic packaging, etc.), or polymers used as "permanent" adhesives.

1.3.1 Polycarbonate

Polycarbonate (PC) is a thermoplastic that is broadly employed in electronics as resistors, capacitors, data storage/media, and similar applications, because of its high heat-resistant and flame-retardant properties. As such, radiation-induced behaviors are available in the literature. Chen et al. studied the mechanical behaviors of electron beam radiation on PC up to 250 kGy (25 Mrad) at a beam energy of 2 MeV.[32] (For comparison, most of the polymers discussed thus far were irradiated with ^{60}Co, which has two discrete energies of 1.17 and 1.33 MeV.) Fig. 9 shows that no degradation in tensile elongation and strength occurs at doses lower than 50 and 100 kGy, respectively.[32]

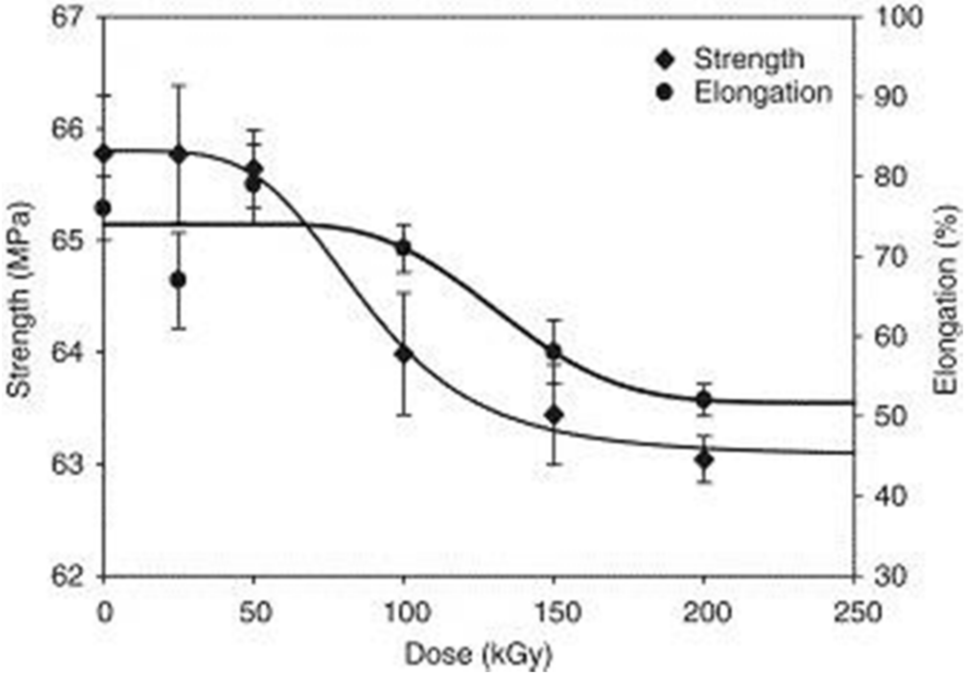


Fig. 9. Tensile strength and elongation of polycarbonate irradiated with a 2-MeV electron beam.[32]

Similarly, Sardinha de Melo et al. investigated the toughness of gamma-irradiated Lexan Margard polycarbonate.[33] The gamma source used was a ^{137}Cs (avg. energy ~1.176 MeV) with an operating dose rate of 2 kGy/hr (±2%). Fig. 10 shows that the most sensitive mechanical property tested was elongation at break, which decreases at low gamma doses. However, tensile strength and apparent fracture toughness are minimally affected at doses less than ~20 kGy (2 Mrad).[33] In any case, the work presented by Chen[32] and Sardinha de Melo[33] supports the fact that PC is sufficiently robust to withstand low to moderate doses of radiation, corroborating the data presented in Fig. 1.[6]

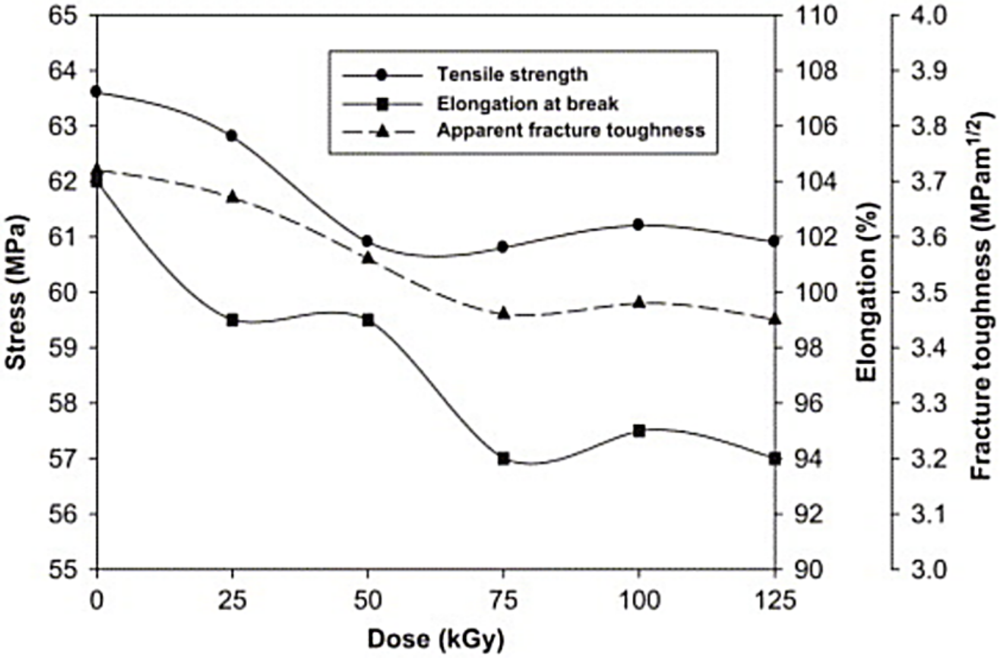


Fig. 10. Mechanical behavior of radiation-aged polycarbonate as a function of dose.[33]

1.3.2 Bisphenol A-Based Epoxies

Bisphenol A-based epoxies are used in numerous applications, including those that are high-consequence. As such, the aging behavior for varying degradation environments is widely published in the literature, including radiation.[9,34-38] White and co-workers recently performed neutron (^{252}Cf point source, avg. energy ~2.3 MeV) and gamma (^{60}Co multi-pin array) radiation-aging experiments in air on an aromatic amine cured epoxy (for example, EPON828-1031/DDS) up to a neutron fluence (ϕ) and gamma dose up to ~2×10^{13} n/cm^2 and ~920 Gy, respectively.[39] As expected,[40] thermal and mechanical analysis demonstrated no reduction in glass transition temperature or adhesion strength (measured by napkin ring torsional tests; see Fig. 11).[39]

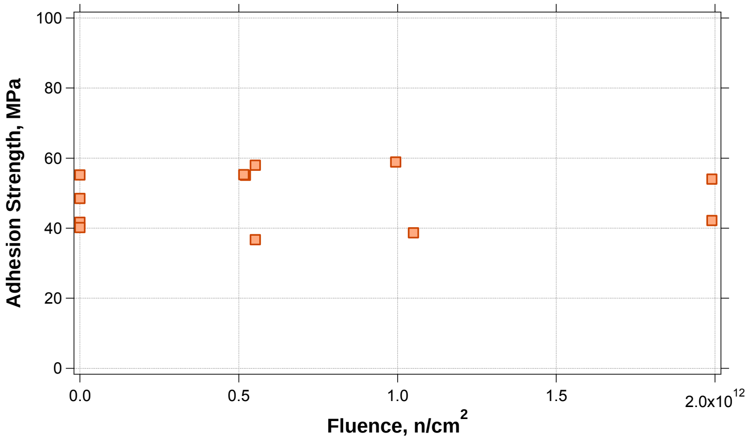


Fig. 11. Adhesion strength data measured for radiation-aged EPON828-1031/DDS as a function of neutron fluence.[40]

Comparatively, Kline investigated the thermal neutron and gamma radiation effects on EPON828 with ϕ and doses varying between 5 × 10^{14} n/cm^2 to 1 × 10^{16} n/cm^2 and 11 kGy (1.1 Mrad) to 220 kGy (22 Mrad) at 60 °C, respectively. Mechanical testing demonstrated no changes in tensile strength as a result of radiation exposure (T = 54 °C, ϕ ~ 1 × 10^{16} n/cm^2, and γ dose ~220 kGy).[38] Further, Rivaton and Arnold studied the effects of neutrons (average energy of ~14 MeV and flux of ~2 × 10^8 n/s, maximum ϕ up to ~4.5 x 10^{13} n/cm^2 over 40 hr) on a wide range of polymers, including epoxy resins and quantified structural changes through physical and chemical analysis, including thermal analysis.[41] No variation in glass transition temperature was observed for the epoxies included in their investigation.[41] In general, a reduction in glass transition temperature can be correlated to a decrease in mechanical properties, such as a reduction in adhesion strength in the case of epoxies.[34]

1.3.3 Polyether Ether Ketone

Polyether ether ketone (PEEK) is a widely employed thermoplastic prepared through step growth polymerization techniques; Fig. 12 is the molecular structure of the PEEK repeat unit. Exceptional thermal (that is, T_g ~144 °C [42] and T_m ~340 °C [7]), mechanical (for example, tensile strength of ~90 MPa), and chemical and fatigue resistance make PEEK an ideal polymer for some high-performance applications.[7, 42-44] Low-rigor processability characteristics afford facile fabrication of injection moldings, wire coatings, fibers, films, and composites.[44] As such, PEEK is frequently used in many high-consequence systems and components with long service lifetimes.

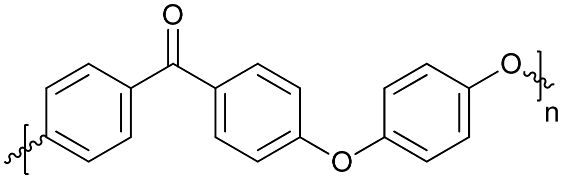


Fig. 12. Chemical structure of polyether ether ketone (PEEK).

Li et al. performed gamma irradiation experiments to develop a better understanding of ionizing radiation effects on PEEK.[46] Experiments were conducted at room temperature and 77 K using a ^{60}Co source; doses achieved were 100, 200, and 600 kGy; unfortunately no dose rates were reported. Standard electron spin resonance (ESR) techniques were employed to characterize free radical formation and decay. There was no evidence of radical formation from irradiation at room temperature (if any free radicals were formed, they were short-lived); experiments performed at 77 K showed changes in ESR spectra (see Fig. 13). They concluded that PEEK was reasonably resistant to gamma radiation in terms of stable radical formation yields, but point out that *significant* changes were observed in the dielectric properties of PEEK at relatively low gamma radiation doses.[46]

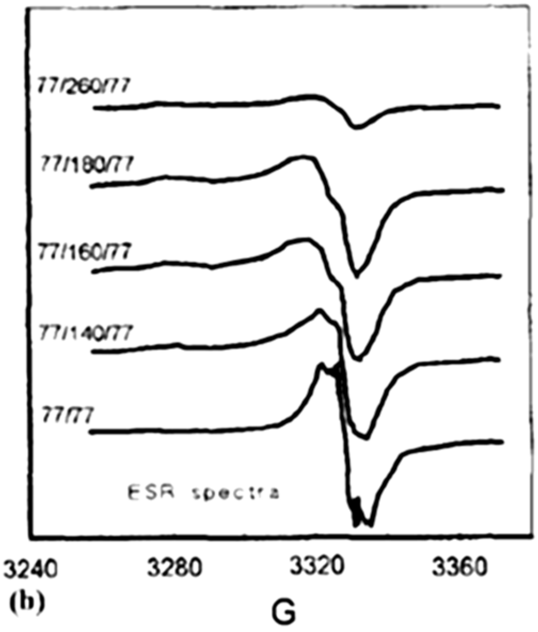


Fig. 13. ESR spectra of irradiated PEEK at 77 K.[46]

1.3.4 Polydiallyl Phthalate

Polydiallyl phthalate (DAP)(see Fig. 14) is another widely used rigid thermoplastic known for its strength and good insulating properties. Unfortunately, technical data are limited in the literature regarding its performance after radiation exposure. However, it has been tested as a surrogate material for muscovite, where scientists track fission products of ^{238}U to date fossils and minerals.[47] In short, Yoshioka et al, demonstrated that DAP was resistant to a thermal and fast neutron fluence (ϕ) of ~4 × 10^{15} n/cm^2 and 9 × 10^{14} n/cm^2, respectively. No mechanical properties were measured, but the DAP resin did perform as required as a radiation detector after neutron exposure.[47] From a performance perspective, the aromatic moieties in the diallyl phthalate (DAP) polymer backbone suggest that low doses (10 kGy or 1 Mrad or less) of radiation should not cause any concerns for degradation of mechanical properties.

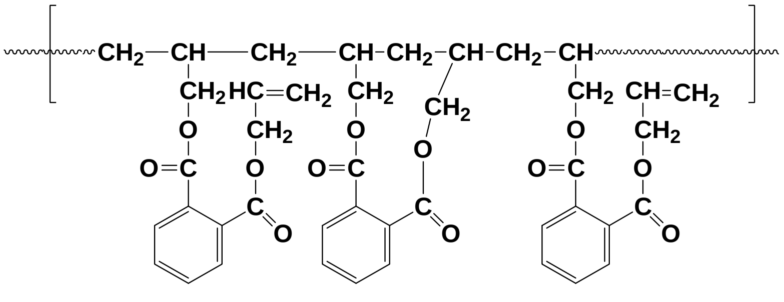


Fig. 14. The chemical structure of diallyl ortho-phthalate (DAP) Note: it is usually crosslinked and does not contain any residual unsaturation.

1.3.5 Vespel SP-1

Vespel SP-1 (polyimide) is a high-strength, temperature-resistant thermoplastic (see Fig. 15). A great deal of research has already been performed on Vespel SP-1 because of its potential in nuclear energy, outer space, fusion reactors, and other high-consequence applications. Takamura and Kato studied the effect of neutrons and gamma rays on varying organic insulators and epoxies, including Vespel SP-1.[49] This work revealed that gamma and fast neutron doses and ϕ at low temperatures and ~100 MGy, 8.7×10^{16} n/cm^2, respectively resulted in no significant degradation of Vespel SP-1.[49,50]

Comparatively, Coltman and co-workers also conducted a study of glass-fiber-filled epoxies, Vespel SP-1, Spaulrad (Spauldite, a high-pressure aromatic polyimide laminate reinforced with E-glass woven fabric ~70% by weight), and Norplex (formerly Kerimid, a resin of bismaleimide and aromatic diamines reinforced with E glass 40 to 60% by weight.[50] Coltman demonstrated that after 100 MGy (10^{10} rads), only 8% of its original flexural strength was lost when irradiated at 77 K (-196 °C); the linear flexural modulus increased at 100 MGy when irradiated at 300 K (~27 °C, see Fig. 16).[50]

More recently, White et al.[51] performed radiation "robustness" experiments on pristine Vespel SP-1 (^{60}Co) up to 100 Gy. As expected, three-point-bend tests demonstrated no reduction in flexural modulus at low doses (Fig. 17). Differential scanning calorimetry of the virgin and irradiated Vespel SP-1 also exhibited no variation in thermal properties.

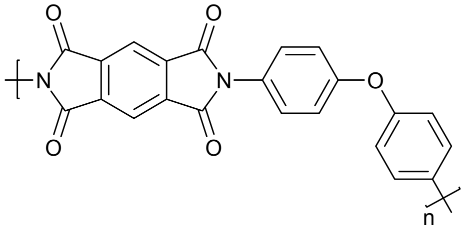


Fig. 15. The chemical structure of a polyimide (that is, Vespel SP-1).[48]

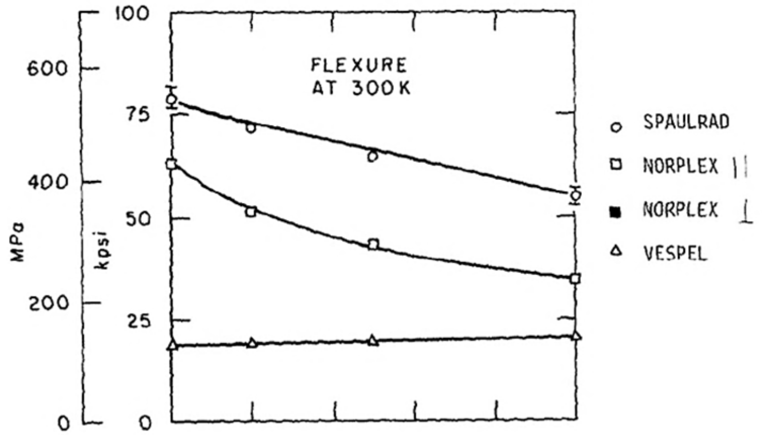


Fig. 16. Linear flexural strength of pure and glass-fabric-filled polyimides after irradiation at 307 K.[50]

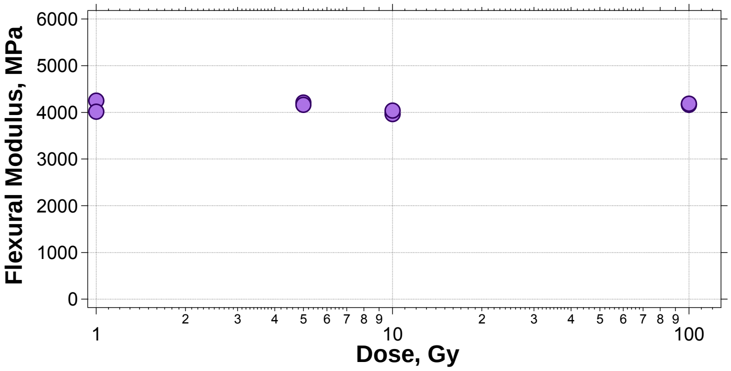


Fig. 17. Three-point-bend data for the flexural modulus of Vespel SP-1 irradiated to 100 Gy.

1.3.6 Torlon® 4203

Torlon® 4203 (polyamide-imide) is another aromatic thermoplastic. Similar to Vespel SP-1, it is different because of the amide functional groups incorporated into the backbone of the polymer (Fig. 18). Unfortunately, because Torlon® 4203 is still relatively new, not much technical data exist for radiation-induced behaviors. The mechanical properties of Torlon® 4203 are often described to be similar to those of Vespel SP-1, that is, very resistant to neutron and gamma radiation damage. White et al. recently performed radiation "robustness" experiments on pristine Torlon® 4203 (^{60}60) up to 100 Gy. As expected, three-point-bend tests demonstrated no reduction in flexural modulus occurred at low doses (Fig. 19). Differential scanning calorimetry of the virgin and irradiated Torlon® 4203 also exhibited no variation in glass transition temperature.

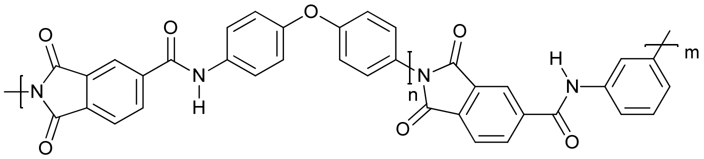


Fig. 18. The chemical structure of a polyamide-imide (e.g., Torlon® 4203).

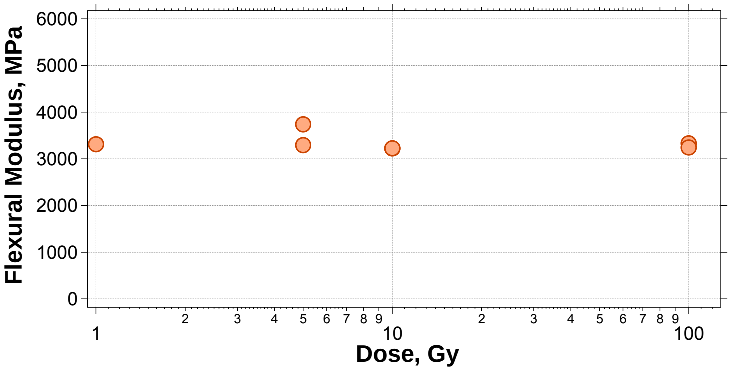


Fig. 19. Three-point-bend data for the flexural modulus of Torlon® 4203 irradiated to varying doses.

1.4. Silicone Rubbers

Silicone rubbers (see Fig. 20), like many other elastomers, have been used in varying radiation environments. In short, they are very radiation-resistant up to ~100 kGy (10 Mrad) of dose; minimal damage can be expected up to ~1 MGy, depending upon the thermal environment. Some material applications include, but are not limited to gaskets, seals, and cable insulations. As such, radiation-aging data exists for varying physical and chemical properties as a function of dose. For example, Maxwell et al. studied the effects gamma radiation (^{60}Co) on a silicone composite (RTV-5370) up to nearly 4 MGy; because of a high dose rate, non-oxidative radiation chemistry is expected to be dominant in this study.[52,53] Fig. 21 shows that little change occurs for RTV-5370 up to ~500 kGy (50 Mrad); however, at higher doses the silicone hardens, clearly evident by the increasing storage modulus (a measure of a materials elasticity).[52]

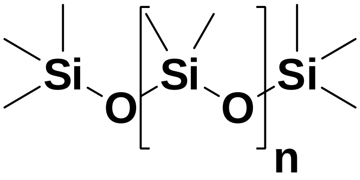


Fig. 20. Chemical structure of generic silicone rubber.

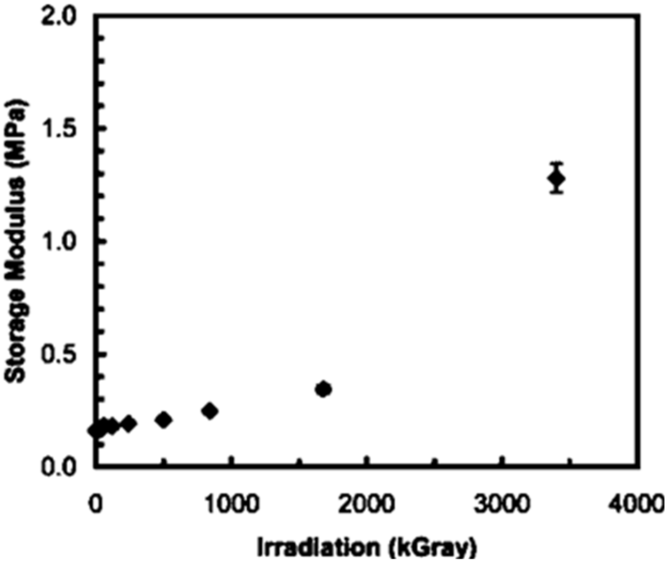


Fig. 21. Equilibrium storage modulus as a function of dose for gamma-irradiated RTV-5370.[52]

For comparison, White and co-workers studied the effects of gamma radiation on silicone elastomers used as nuclear power plant cable insulations.[54] Fig. 22 shows that ~2500 hours at a dose rate of 39 Gy/hr, which equals an exposure dose of ~100 kGy or 10 Mrad at 100 °C, is required to reduce the tensile elongation by nearly half of the original properties.[15] This degradation process correlates with a very small increase in gel content, indicative of some cross-linking, and above 100 kGy yields indications for possibly embrittlement trends (that is, significant loss of tensile elongation properties).

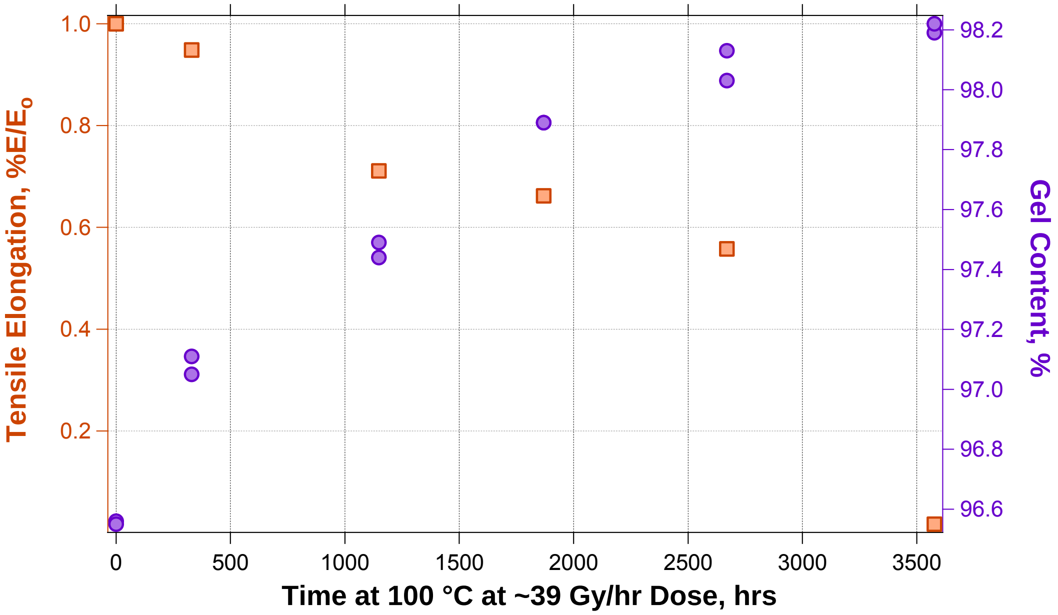


Fig. 22. The effects of gamma irradiation on the tensile elongation and gel content of silicone nuclear power plant cable insulations.[15]

1.5. Fluoropolymers

Among the varying classes of polymers, fluorinated polymers (or fluoropolymers) are generally considered the most radiation sensitive polymers due to their complex morphology, particularly under oxidative conditions, and do not perform as well as non-fluorinated polymers (see Fig. 1) tolerant between ~1 kGy and 1 MGy, depending upon the environment. A general memo was written a number of years ago on the topic of HF generation and irradiation of fluoropolymers.[55] No dramatic issues were found.

1.5.1 Polytetrafluoroethylene

Polytetrafluoroethylene (PTFE), more commonly known as Teflon, is used in numerous applications because it is generally considered to be non-reactive, resistant to chemical degradation, and tolerant to high temperatures. The chemical structure of PTFE is shown in Fig. 23. Because of the widespread use of PTFE, radiation performance data are available in the current polymer literature. Chipara and Chipara studied the effect of dose and dose rate on virgin PTFE up to ~80 kGy at dose rates ranging from 80 Gy/hr up to 240 Gy/hr at room temperature.[56] If should be noted that this work could have been under diffusion limited oxidative conditions.

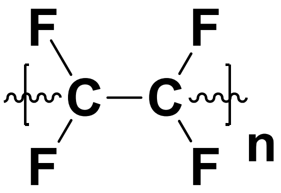
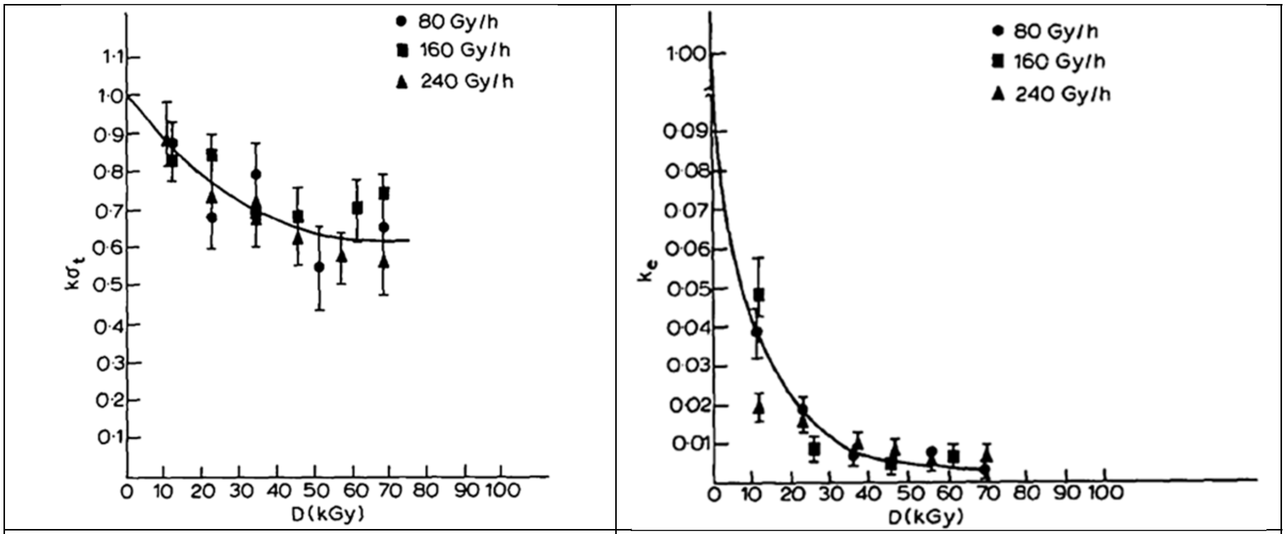


Fig. 23. Chemical structure of a PTFE.

Fig 24a shows that as the dose increases up to ~80 kGy (8 Mrad), only 40% of the initial tensile strength is lost for PTFE. However, a significant degradation in tensile elongation at break was observed for doses less than 10 kGy (see Fig. 24b)[55]; this is consistent with data presented in Fig. 1.[6]



(a) (b)

Fig. 24. Gamma radiation dose and dose rate effects on the tensile strength (a) and tensile elongation at break (b) for PTFE.[56] For (a) and (b) the y-coordinate is normalized tensile strength and tensile elongation at break, respectively.

1.5.2 Fluorinated Ethylene Propylene

Fluorinated ethylene propylene (FEP) is another widely used fluorinated polymer, often as an insulating material for electrical wires. However, based on its copolymer structure it is much more amorphous than PTFE. It is different from PTFE because it can be melt-processed, lending itself more readily to injection molding. The chemical structure of FEP is shown in Fig. 25. Because the chemical moieties that compose FEP and PTFE are similar, they respond to ionizing radiation in the presence of oxygen in a reasonably similar manner.[57] It has been reported that after 10 kGy (1 Mrad) of radiation the tensile strength of FEP is reduced by ~10%.[58]

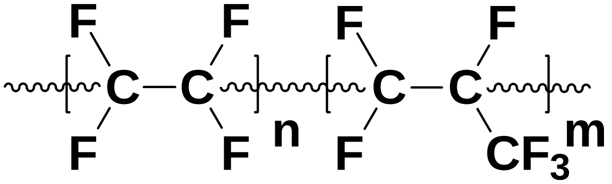


Fig. 25. The chemical structure of fluorinated ethylene propylene (for example, FEP).

1.6. Polymers Conclusions

In any critical application a scoping radiation study could be performed as confirmation of material performance; however, in general, most polymers can tolerate radiation quite well, with doses in the range of 10 to 100 Gy (1 krad to 10 krad), as was of primary concern for this review, causing no degradation in mechanical properties. In many cases, polymers can in fact accommodate much higher doses between 1 kGy and 10 kGy (0.1 and 1 Mrad) with little to no consequence. It is important to note that radiation chemistry can vary considerably between oxidative and inert conditions, with oxidative chemistry being normally more severe and often associated with more dominant polymer scission reactions. Hence, specific combined exposure conditions can convolute otherwise generic trends in polymer radiation degradation.

The focus of this memo was to highlight general trends with regard to organic materials. Further detailed studies are warranted for any critical application, and an SME should be consulted before any decisions are made.

2. Brittle Materials

2.1. Introduction

Glasses commonly used in seals in nuclear weapons (NW) are primarily oxides of silicon (Si) with additions of sodium (Na), potassium (K), aluminum (Al), calcium (Ca), barium (Ba), and others to impart the required properties. When a glass is exposed to radiation, numerous changes in properties can take place. Optical absorption bands may be induced causing color changes, phase separations may occur, or the glass structure itself may be disrupted, leading to changes in density, thermal expansion, hermeticity, strength, and toughness. Early interest in radiation effects on glasses may have originated from the observation by Roentgen that the glass door knobs inside his X-ray room turned blue.[59] Current research is limited, driven mostly by the potential for use of glasses as nuclear waste repositories, as scintillator materials, and in space-radiation environments. This section will briefly review the understanding of the mechanism of radiation damage, and then focus on the few papers that describe changes in physical properties relevant to NW applications of interest. While information on materials used in NW is not available, inferences will be drawn regarding the expected effects of γ radiation on them based on information on other glasses.

2.2. Mechanisms of Radiation Damage in Glasses and Origin of Color Centers

There are two different interactions of radiation with the glass structure: energetic interactions and particle-particle interaction. High-energy radiation (such as high-energy photons from γ radiation) can provide enough energy to electrons in the valence band in the glass to jump over the band gap (ionization). Any extra energy absorbed is converted to kinetic energy. The energetic electrons move through the glass matrix, and may be trapped by flaws to form defect centers, recombine with positively charged holes, or produce secondary electron cascades. Very high energy electrons can also lead to atomistic displacements, creating vacancy-interstitial pairs. Calculations show that energies of the order of 4 to 25 eV are required to displace an atom from its location in the structure. Electrons moving at relativistic speeds can provide this energy. To displace a Si atom, the electron energy needed is ~0.26 MeV, and for an oxygen (O) atom, 0.16 MeV is required. A 10^8 rad ^{60}Co dose can induce 10^{16} displacements of Si atoms/gm of material whereas the same radiation dose produces 10^{25} ionizations/gm. As ionizations predominate during radiation exposure of glass, it is apparent that most ionizations result in recombination, while only a small fraction result in the creation of a defect centers by atomic dislodgement. High-energy particle radiation interacts with the glass matrix through collisions (as discussed above), charge interactions, and transmutation of glass atoms into radioactive species.

There are several types of defects in a glass structure that can trap electrons and holes created by ionization. The first are Schottky (vacancy) and Frenkel (vacancy-interstitial) defects that occur spontaneously in structures so as to minimize the free energy by allowing disorder in the structure. The second category of defects is a substitutional cation, which occurs when a second species cation is doped to form the glass. If the cation has a lower valence, a site where a hole could be trapped is created. For equivalent ions, the electro-negativity difference may still allow the dopant to be a trap. When a network modifier such as an alkaline oxide is added, non-bridging oxygens are created that are potential hole traps. The final type of defects is multivalent cations that can change valence states, and can trap either holes or electrons. An obvious effect of the irradiation of glass is induced optical absorption, which leads the glass to color or even become opaque. Radiation-induced defect centers are the cause: new excited states have been created that can get populated by energy absorption from the incident optical beams. This type of radiation damage is essentially “metastable”: high-temperature anneals can lead to the recovery of the colorless state. While radiation damage leading to color changes is of immense importance for optics, for applications in NW we more concerned with the changes in physical properties of glasses as a result of radiation.

2.3. Changes in Physical Properties of Glasses When exposed to Radiation

2.3.1 Fused Silica

Radiation exposure of vitreous silica causes changes in density, with compaction occurring for pure silica, and a slight expansion followed by compaction for silica with impurities. The compaction is fit to a power law with the radiation dose as:

$$\frac{\Delta\rho}{\rho} = A^{'}D^{c} \qquad (1)$$

where ρ is the density, D is the radiation dose, and $A^{'}$ and c are constants. The dose exponent, c, is dependent on the nature of the radiation source, and the effect of that particular radiation on silica. The value of c is close to 1 for radiation that produces atomic displacement (for neutrons, He^+ and D^+), and ~2/3 for ionizing radiation (γ radiation[60-65], e-beam[60,61], and UV).[66] The basic building block of vitreous silica (and most sealing glasses) is the SiO_4 tetrahedron, with an O-Si-O angle of 109.5°. These tetrahedrals are placed in a network structure with an average Si-O-Si angle of 145°, with the angles distributed in a wide range (120° to 180°). X-ray and neutron diffraction studies of the silica structure after particle bombardment reveal that the tetrahedron unit maintains its shape, while the Si-O-Si angles undergo changes. After a dose of 2.8×10^{20} neutrons/cm^2, the structure of vitreous silica reaches a saturated state of damage, and the mean angle is reduced by ~10°.

Primak and Kampwirth[60,61] observed dilatation of silica using neutrons, 40 to 600 keV electrons, 140 keV H^+, D^+, He^+, and gamma rays. The dilatations are explained as compaction of the silica structure resulting from oxygen moving into some of the "free volume," the structural change being similar to that occurring on pressure, shock, or thermal compaction of vitreous silica. The normal effect of ionizing radiation is to cause a contraction. Vitreous silica materials, which color in the visible (presumably because of their aluminum content), show a very rapid radiation-induced expansion. Thus, depending on the impurity composition, the vitreous silica may show an initial decreased contraction or an initial expansion followed by subsequent contraction. In the coloring vitreous silica, the expansion annealed at a lower temperature than the contraction. The expansion and the contraction appear to be quite independent. The expansion is explained as caused by breaking Al-O links in the structure, the contraction by a compaction of the Si-O structure.

Shelby[64,65] reported the density, refractive index, thermal expansion, and helium permeability of vitreous silica after exposure to a ^{60}Co gamma source (2.3*103 rad/s) for different exposure times such that the dose ranged from 10^8 to 10^{10} rads. A volume compaction, $\Delta V/V$ of ~10^{-5}-10^{-3}, was observed with higher values being obtained at higher doses. As might be expected from the volume compaction, the refractive index, n, increases with dose, and a linear relationship between the quantities $(n-n_0)/n_0$ and $(\rho - \rho_0)/\rho_0$ is obtained. Here the subscript 0 refers to the unirradiated base state. The thermal expansion coefficient of the glass decreases with increasing degree of compaction, and can be as much as 10% lower than the unirradiated material. In NW applications, this level of compaction would lead to large strains in the seals, causing damage and possibly compromising hermeticity.

Higby et al.[67] measured the density change, thermal expansion, and elastic constants of several ultra-low expansion materials including vitreous silica after 2 MeV electron irradiation of 10^9 rad. They found that coefficient of thermal expansion (CTE) data on irradiated Optosil (trade name for a vitreous silica) essentially superimpose on the data taken before irradiation. This result indicates that the CTE is unaffected by the rad dose in the measured temperature range. However, since the zero-CTE range of amorphous SiO_2 is near 190 K, they indicated that it was possible that radiation-induced changes at temperatures lower than those measured by them could exist. For density changes as a function of dose, they reported the values of $A = -4.84 \pm 0.824$, and the exponent $c = 0.32 \pm 0.102$ in Eq. 1. The exponent is different than reported by previous investigators, and was attributed to possible impurity effects. They are also reported increase in the elastic moduli of the material: the values of Poisson's ratio, bulk modulus, and elastic modulus are characterized by slightly higher values after irradiation. The increases in the elastic constants (which range from about 0.25% to 0.5%) are consistent with the increases in measured densities (approximately 0.25%). These general observations suggest that the microstructural compaction processes that govern the density increases also modify the elastic behavior.

Piao et al.[68] describe vitreous silica as a two-phase system with a low-temperature phase A with Si-O-Si bond angle of 145°, while this angle for the high-temperature phase B is 135°. The solid-state phase transition is higher than the glass transition temperature. This model is based on the observed volume change induced by hydrostatic pressure, and fast neutron, ion, electron and photon radiation. On irradiation, the bond length is found to have increased, and the bond angle is reduced. This means that while the material before irradiation is in Phase A, radiation induces a transition to Phase B. They used thermodynamic arguments to show that compaction induced by knock-on radiation in vitreous silica is different from compaction induced by ionizing radiation, which is the purported cause for different values of c in Eq. 1.

2.3.2 Borosilicate Glasses

Shelby[64,65] reported the density, refractive index, thermal expansion, and helium permeability of borosilicate glasses after exposure to a ^{60}Co gamma source ($2.3*10^3$ rad/s) for different exposure times such that the dose ranged from 10^8 to 10^{10} rads. The composition ranges studied are shown in Table 1.

Table 1. Composition (in mole %) of Glasses Studied

Designation	SiO_2	B_2O_3	Al_2O_3	Na_2O	Other Oxides
CGW-3320	75	12.7	2.4	4.6	5.2
CGW-7052	71	17	4.8	5.7	1.5
CGW-7740	81	12	2	4.2	1
CGW-7913	97	2.5		0.5	
GSC-4	85	13.3	0.5	1.2	

The results of volume compaction as a function of dose are shown in Fig. 26. The glass composition has a major effect on compaction, with glasses with higher boron content showing higher compaction. The slopes of the curves are roughly similar, indicating that composition does not influence the rate of compaction. Vitreous silica has the highest radiation tolerance. From these results, we can surmise that multi-component glasses used in NW will have a lower threshold for measurable compaction than the materials shown here.

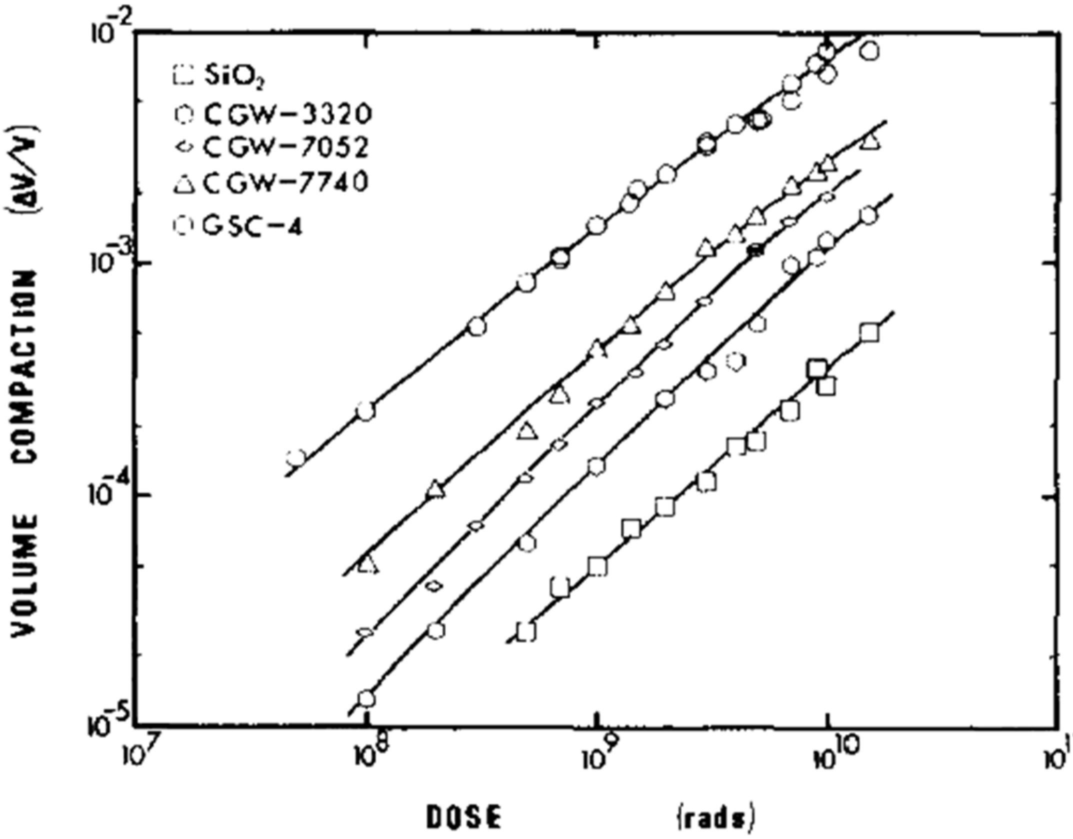


Fig. 26. Effect of dose on the volume compaction of borosilicate glasses.

The thermal expansion of the glasses decreased with increasing dose (dose is proportional to change in volume) as shown in Fig. 27, while the helium permeability did not change for any of the glasses studied. The observed effect of boron content suggests that these atoms are directly involved in the densification. It is likely that the effects are related to the two-phase nature of these glasses: the glass consists of a continuous phase with a composition near that of pure vitreous silica, with droplets or co-continuous alkali borate phase. The properties of this phase are different than that of silica, with a glass transition temperature more than 500 °C below that of silica. As a result, relaxation in this phase occurs much faster. So if incident radiation breaks the B-O bond, the glass network can relax before the bond can reform, leading to a much higher compaction in glasses containing boron.

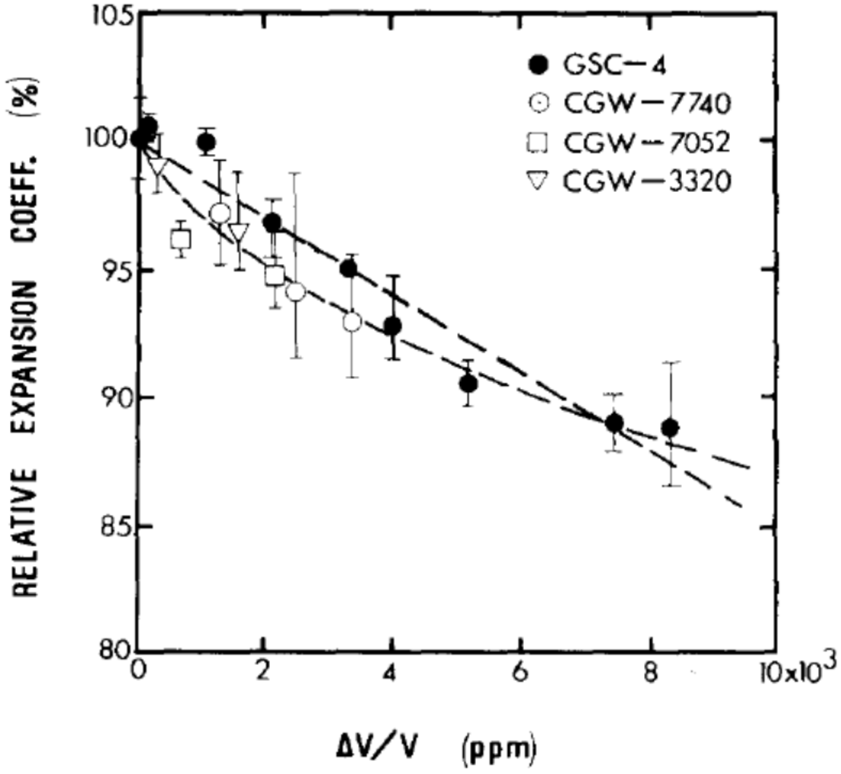


Fig. 27. The relative thermal expansion of the borosilicate glasses, as a function of the compaction.

Zdaniewski et al.[69] studied the effect of ^{60}Co gamma radiation on the strength-related mechanical properties of a borosilicate glass (CGW 7740). Although the glass darkened considerably, only a very slight densification was observed after irradiation to levels of 10^8 rads. Exposure of the glass to 10^2 rads resulted in only a slightly noticeable darkening; however, the dose of 10^4 rads resulted in a distinct brown color that became more intense at higher doses. After a dose of 10^8 rads, the glass was an opaque dark brown. It was surmised that the electronic defect structure of the glass was modified by the irradiation. The strength distributions were not appreciably changed by the irradiation. Their strength results (as Weibull strength distributions) in water and inert nitrogen environments for different radiation doses are shown in Fig. 28. While the strength for the water environment is lower because of sub-critical crack growth, there is no significant difference in strength for different rad doses. The calculated slow crack growth parameter, or N value, was also unchanged. The radiation also did not affect the elastic modulus or the fracture toughness of the glass. Gamma radiation does not affect the strength below 10^8 rads.

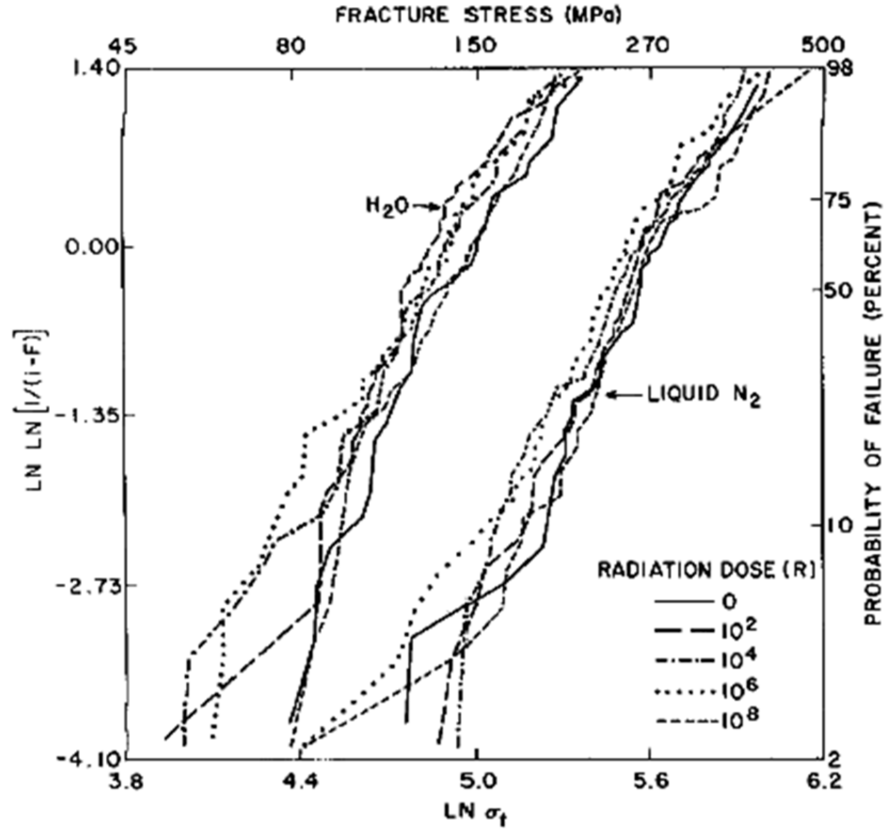


Fig. 28. Strength distributions for different levels of gamma ray exposure for borosilicate glass.

Weber and Matzke[70] reported the hardness, fracture toughness, and elastic modulus of several complex borosilicate glasses that were being studied as host materials for nuclear waste. They found that the hardness of the glass decreased with increasing dose of alpha particles, while the critical load to induce cracking increased. They also measured an increase in fracture toughness with dose for these glasses.

Moody et al.[71] reported the fracture toughness of silica and borosilicate glasses after exposure to 10^{10} rad of γ-ray exposure, and found that the fracture toughness was unaltered. However, glasses that had been exposed to hydrogen and then radiated showed an increase in fracture toughness. The authors attributed this to the formation of hydroxyl and hydride groups, which strengthens the bridging of the Si-O and B-O bonds.

2.3.3 Other Glasses

Swift[72] reported interesting results on the compositional effects on strength after radiation effects on soda-lime-silica glass. For samples that contained a small amount of arsenic oxide, radiation with γ-ray led to slight increase in strength (~20%). However, samples without arsenic lost ~20% strength even after exposure to mild (88 Gy = 8800 rad) radiation. However, further exposure to 880 Gy and 8800 Gy (= $8.8*10^5$ rad) did not degrade the strength further. The reason for this strength loss was not discussed.

Wiedlocher et al.[73] studied the effect of low-earth orbit on the strength of several commercial glasses: BK7, fused silica, Pyrex, Vycor, and soda-lime silica. Strength samples were exposed at a 460-km orbit for 5.8 years, and were tested after recovery. The strengths after radiation were found to be within the scatter of the measurements for the untreated materials. The authors did not provide an estimate of the radiation dose experienced by the samples.

Ram and Ram[74] studied the infrared and Raman spectra of glass ceramics based on the $PbO-Cr_2O_3-B_2O_3$ glass composition system. The bands characteristic of BO_3 and BO_4 functional groups are present in all the samples. An incorporation of Al_2O_3 (up to 5 mol %) in the initial glass composition considerably changes the glass network structure and relative concentrations of BO_3 and BO4 groups. The composition 50 PbO-20 Cr_2O_3-25 B_2O_3-5 Al_2O_3 (in mol %) reveals a maximum fraction of boron in the BO_4 group. A sample of this composition heat treated at 850 ~ C for 25 hours shows a maximum crystallization fraction with $Pb_2O.CrO_4$ as a prominent crystalline phase. The glasses irradiated with γ rays inhibit the crystallization into the $Pb_2O.CrO_4$. They also show relatively smaller thermal conductivity.

Sharma et al.[75] studied the optical and structural properties of $xPbO.2xBi_2O_3.(1-3x)B_2O_3$ glasses of different composition using UV-VIS and Fourier transform infrared spectroscopic techniques. Effects of gamma radiations on glass network and structural units were studied by irradiating glass samples with a ^{60}Co radioisotope to the overall dose of 2.5 kGy. Irradiation causes compaction of the borate network by breaking the bonds between trigonal elements, which leads to a decrease in the optical band gap energy.

Akhtar et al.[76] reported the transmission loss for several commercial glasses when exposed to different doses of γ-radiation. Their results reveal the compositional dependence of darkening, with pure silica glass (Suprasil) showing negligible effects, while for other glasses (such as BK7), the effect of radiation on transmission appears to saturate. Their results are reproduced in Table 2.

Table 2. Transmission Effects of Radiation on Commercial Glasses

Glass Type	Wavelength (nm)	10^6 rad, Transmission Difference (%)	$4X10^6$ rad Transmission Difference (%)	10^7 rad Transmission Difference (%)
BK7	670	-45	-65	-70
	1064	-1.9	-2.9	-3
JGS1	670	-0.05	-0.01	-0.05
	1064	-0.01	0.02	-0.05
K5	670	-50	-55	-57
	1064	-1.7	-2.1	-2.5
SF11	670	-4	-4.43	-6.7
	1064	-0.5	-0.5	-0.62
QK1	670	-15	-26	-28
	1064	-2.7	-4	-4.6
ZF7	670	-5.6	-12	-16
	1064	-2.6	-3	-3
Suprasil	670	-0.08	-0.4	-0.06

Glass Type	Wavelength (nm)	10^6 rad, Transmission Difference (%)	4X10^6 rad Transmission Difference (%)	10^7 rad Transmission Difference (%)
	1064	-0.05	-0.1	-0.07
Quartz	670	-0.18	0	0.01
	1064	-0.05	-0.05	-0.01

Tandon and co-workers[77] have recently measured the strength of optical fibers exposed to 10^5 rad, 5 × 10^6 rad, and 24 × 10^6 rad at the Gamma Irradiation Facility (GIF) at Sandia National Laboratories. These fibers are primarily composed of fused silica, with a surface glass layer of germanium (Ge) doped silica. The strength results are shown in Fig. 29, where testing was conducted at 50% relative humidity, one day after the radiation exposure. A strength loss of 4 to 5% can be observed. As optical fibers have a polymer coating, and the coating properties can also change because of radiation, the entire loss of strength cannot be attributed to changes in the glass alone.

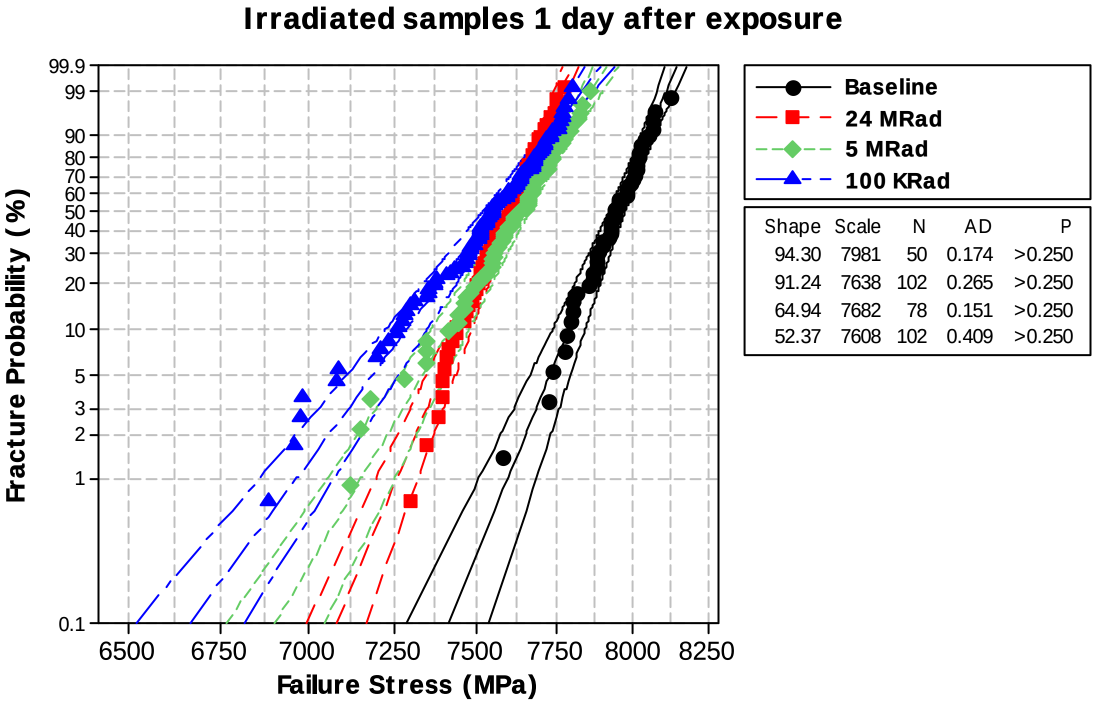


Fig. 29. Change in the strength of optical glass fibers (predominantly SiO_2) after radiation exposure at the GIF at Sandia National Laboratories.

2.4. Implications for NW Components

Although literature on radiation effects on glasses is extensive, there are very few studies that focus on the mechanical and elastic properties after radiation. Shelby's work cited here shows that at 10^8 rad exposure there is a decrease in thermal expansion and increase in density of silica and borosilicate glass. Higby's work on silica shows a slight increase in elastic constants.

Zdaniewski et al. showed no change in toughness or elastic modulus while Moody's work shows no change in toughness up to 10^{10} rad. Swift's work shows a 20% strength loss in soda-lime with 8.8×10^3 rad exposure. Fused silica is known to be the most tolerant to radiation damage. We expect, therefore, that the multi-component glasses of interest in NW would have significantly lower damage thresholds ($<<10^8$ rad), but how much lower has not been determined. Any change to the mechanical and elastic property would lead to some changes in the residual stress states in the glass components. Loss of strength observed in some studies could lead to cracking even if the residual stress state itself does not change. It is likely that any radiation effects in our applications will be negligible (save for some darkening). However, because data on materials to be used in the seals are non-existent, it is recommended that a materials-based study be commissioned to explore these effects. In this study, commonly used glass and glass-ceramics should be tested in the GIF, and relevant properties such as density, thermal expansion, elastic moduli, strength, and toughness should be monitored as a function of dose.

3. Acknowledgment

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