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Gamma Radiation Aging Study of a Dow Corning SE 1700 Porous Structure Made by Direct Ink Writing

W. Small, C. T. Alviso, T. R. Metz

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Work done by: Tom Metz (3D printing), Cindy Alviso (aging), Ward Small (data acquisition/analysis)
Report written by: Ward Small
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SUMMARY

Dow Corning SE 1700 (reinforced polydimethylsiloxane) porous structures were made by direct ink writing (DIW). The specimens (~50% porosity) were subjected to a compressive strain of ~25% while exposed to a gamma radiation dose of 1, 5, or 10 Mrad under vacuum. Compression set and load retention of the aged specimens were measured after a ~24 h relaxation period. Compression set (relative to deflection) increased with radiation dose: 11, 35, and 51% after 1, 5, and 10 Mrad, respectively. Load retention was 96-97% for the doses tested. The SE 1700 compared favorably to M9763 cellular silicone tested under the same conditions.

MATERIALS AND METHODS

Sample Preparation

Dow Corning SE 1700 clear adhesive is a two-part heat cure reinforced polydimethylsiloxane rubber with a 10:1 by weight mix ratio. After mixing using a Thinky planetary mixer, the resin was vacuum degassed, loaded into a 30 cc syringe, vacuum degassed again, and centrifuged. The trapped air was bled from the syringe and a micronozzle (250 µm inner diameter) was attached to the syringe. The syringe was mounted to the z-stage of a three-axis linear positioning system (Aerotech).

The samples were made using a direct ink write (DIW) 3D printing process [1]. A silicon substrate coated with a teflon mold release agent was mounted on the xy-stage of the positioning system. A positive displacement fluid dispenser (Ultimus IV Model 2800-30, Nordson) was connected to the syringe and programmed to dispense the resin at a constant rate that matched the print speed (15 mm/s). Printing was initiated by executing the tool path program in the A3200 CNC Operator Interface Control software (Aerotech). An eight-layer structure resembling a face-centered tetragonal (FCT) configuration was printed (Fig. 1). The filament diameter was 250 µm and the filament center-to-center spacing was 500 µm. The finished sample was ~75 mm square by ~1.6 mm thick. The silicon substrate with the completed sample was removed from the positioning system and placed in an oven at 150°C for 1 h under nitrogen purge to cure the resin. The cured sample was detached from the silicon substrate and a 28.68 mm diameter die was used to cut out circular disk specimens from the sample. The specimens were purged with nitrogen for 24 h at room temperature immediately before use.

The same 28.68 mm diameter die was used to cut out circular disk specimens from a sheet of M9763 cellular silicone (~2 mm thick). The specimens were purged with nitrogen for 24 h at room temperature immediately before use.

Accelerated Aging and Data Acquisition

Four specimens of the SE 1700 printed material were compressed in a rig comprised of two 3.18 mm thick parallel aluminum plates bolted together and separated by 1.19 mm thick spacers to achieve a compressive aging strain of ~25% (Fig. 2a,b). The actual aging strain varied slightly since the specimen thicknesses varied slightly while the spacer thickness was constant. The rig containing the compressed specimens was sealed in a stainless steel vacuum vessel (Fig. 2c) and evacuated for up to 2.5 h down to

60-65 mTorr. The evacuated vessel containing the compressed specimens was exposed to gamma radiation from a Co-60 source (dose rate = 0.066 Mrad/h) at room temperature at one of three doses: 1, 5, or 10 Mrad. After radiation exposure, the specimens were removed from the compression rig and allowed to relax for 24 h at ambient conditions prior to measurement. The specimens adhered to the aluminum plates, but were not damaged during removal. Before and after aging, the uncompressed specimen thickness and load at the aging strain were measured using an Instron 5967 dual-column load frame with a 50 mm diameter fixed lower platen and a 50 mm diameter spherical seat upper platen (specimen centered). No lubricant was used on the polished steel platens. Three load-unload cycles were performed (test speed = 0.2 mm/min), and the load and crosshead displacement were recorded at 10 Hz. Instrument compliance (~6×10^{-5} mm/N) was measured to correct the crosshead displacement.

The M9763 specimens were tested similarly. The spacers in the compression rig were 1.52 mm thick to achieve ~25% compressive strain.

Data Analysis: Compression Set and Load Retention

The recovered thickness of the uncompressed specimen (*i.e.*, final thickness t_f) was determined based on the compliance-corrected displacement of the crosshead when the load reached ~2 kPa during the third loading phase. Compression set S (relative to compressive deflection) was calculated using the measured original specimen thickness t_o, compressed thickness t_c, and final thickness t_f: $S=(t_o-t_f)/(t_o-t_c)=(t_o-t_f)/\varepsilon t_o$. Note that 100% relative compression set = complete loss of function. The force F_t measured at the aging strain during the third loading phase was used to calculate the load retention. Load retention R was calculated by dividing the force measured while the specimen was under compression by that at time zero (before aging): $R=F_t/F_0$.

RESULTS AND DISCUSSION

Relative compression set and load retention are shown in Table 1 and Fig. 3. Compression set increased with radiation dose. The DIW SE 1700 and M9763 showed similar compression set at 1 Mrad; at higher doses, the SE 1700 showed less compression set than M9763. Load retention for the DIW SE 1700 was nearly constant at 96-97% for the doses tested, despite the increasing compression set. The M9763 showed lower load retention at all doses tested. Interestingly, permanent tensile set (relative to deflection) previously measured for solid (non-porous) SE 1700 was approximately 16, 34, and 50% following gamma radiation doses of 1, 5, and 10 Mrad under 14-32% tensile strain [2]; tension and compression studies yielded similar values for radiation-induced permanent deformation. In both cases the permanent deformation is likely due to radiation-induced crosslinking, a phenomenon known to occur in silicone polymers [3].

Representative stress vs. crosshead position curves are shown in Figs. 4 and 5 for the DIW SE 1700 and M9763, respectively. Both materials showed increasing stiffness (steeper slope) and permanent deformation (compression set) with radiation dose. The load at maximum compression (load retention) was fairly constant for the DIW SE 1700, and decreased with dose for the M9763.

Load retention depends on both the amount of compression set as well as the amount of stiffening, where the compression set and stiffening depend on radiation dose. In addition to radiation-induced crosslinking, some amount of compression set may arise from relaxation of the polymer chains. Another factor that may contribute to the amount of compression set is the distribution of stress in the material during aging, which depends on the structure of the material. Ordered structures such as the DIW SE 1700 and stochastic structures like the M9763 cellular foam experience different stress distributions when subjected to identical compressive strains, possibly resulting in different chain relaxation kinetics.

Plotting load retention as a function of compression set provides another means of comparing materials, and may shed additional light on the aging mechanisms at play. Such a plot is shown in Fig. 6 for DIW SE 1700 and M9763 aged under ~25-30% compressive strain with and without exposure to radiation. The non-irradiated data was taken from previous long-term aging studies in which the materials were aged under various compressive strains at various temperatures [4]; data out to 1 year for the specimens aged at room temperature is shown. Without irradiation, the DIW SE 1700 and M9763 curves are similar. However, the M9763 exhibited higher compression set over time (~7% vs. ~4% for DIW SE 1700 after 1 year) [4], suggesting that it is more susceptible to chain relaxation processes responsible for compression set; the different stress distribution in the foam could play a role. For a given amount of compression set, the irradiated materials exhibit higher load retention than the non-irradiated materials, suggesting that radiation-induced crosslinking imparts stiffness to the materials which boosts the load at maximum strain. For a given amount of compression set, the DIW SE 1700 exhibits higher load retention than M9763.

Recent thermal aging studies performed in accordance with ASTM D395 have shown that an extended cure time can reduce the amount of compression set in SE 1700. Further work is required to determine if the same holds true for radiation aging.

CONCLUSIONS

Measured compression set and load retention suggest that SE 1700 DIW porous structures (FCT, 250 μm filament diameter, 500 μm center-to-center spacing, 8 layers) can retain mechanical functionality following a gamma radiation dose of 1, 5, or 10 Mrad under ~25% compression. The SE 1700 material compares favorably to M9763 cellular silicone.

Compression set and load retention are both valuable measurements in aging studies. The compression set measurement provides insight regarding the potential formation of gaps between the material and adjacent components as they thermally expand or contract in service. Load retention also provides an important functional assessment since the material may be required to maintain a load on adjacent components; however, it is best used in conjunction with compression set measurement since it offers no indication of dimensional changes. For example, the measured load retention may be acceptable even when the material undergoes near 100% compression set, which may not be acceptable; it is not until it reaches exactly 100% compression set that the load retention drops to zero. In addition, it is important to consider the effects of aging on the mechanical properties of the material (e.g., stiffness), since a material may exhibit acceptable compression set and load retention, but may develop higher (or lower) stiffness which may not be acceptable.

ACKNOWLEDGMENTS

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Table 1: Average Relative Compression Set and Load Retention of Porous Structures (n=4)

	1 Mrad	5 Mrad	10 Mrad
DIW SE 1700 (FCT)			
Rel. Compression Set (%)	10.5±0.4	35.1±1.2	50.5±1.0
Load Retention	97.4±0.1	96.2±0.6	95.9±1.4
M9763			
Rel. Compression Set (%)	9.9±0.4	54.2±2.2	73.0±1.6
Load Retention	91.7±0.6	78.9±1.9	68.6±2.9

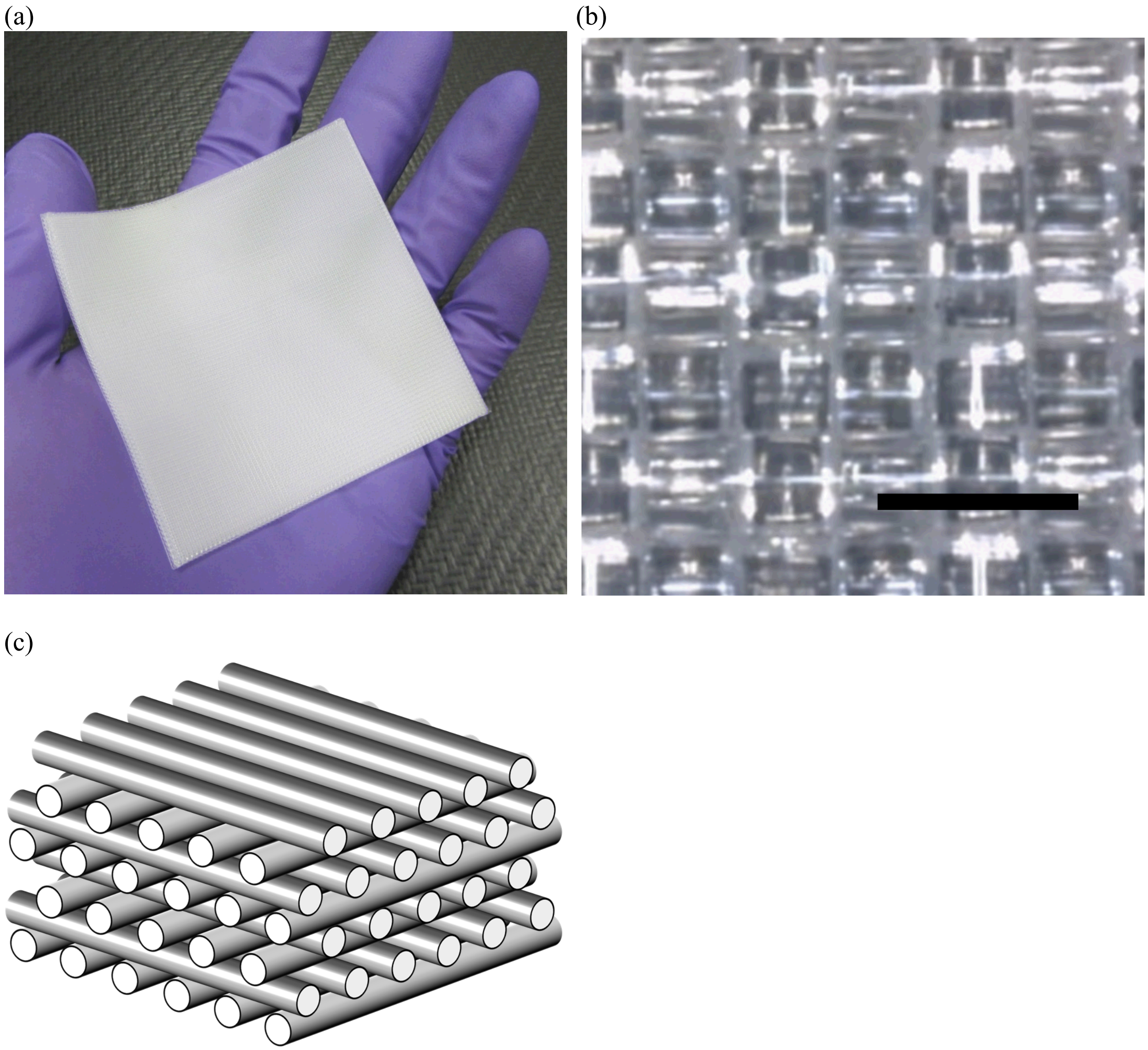


Fig. 1. (a) Photograph and (b) top-view micrograph of a DIW printed SE 1700 sample (bar = 0.5 mm). (c) Illustration of the eight-layer FCT structure.

(a) (b) (c)

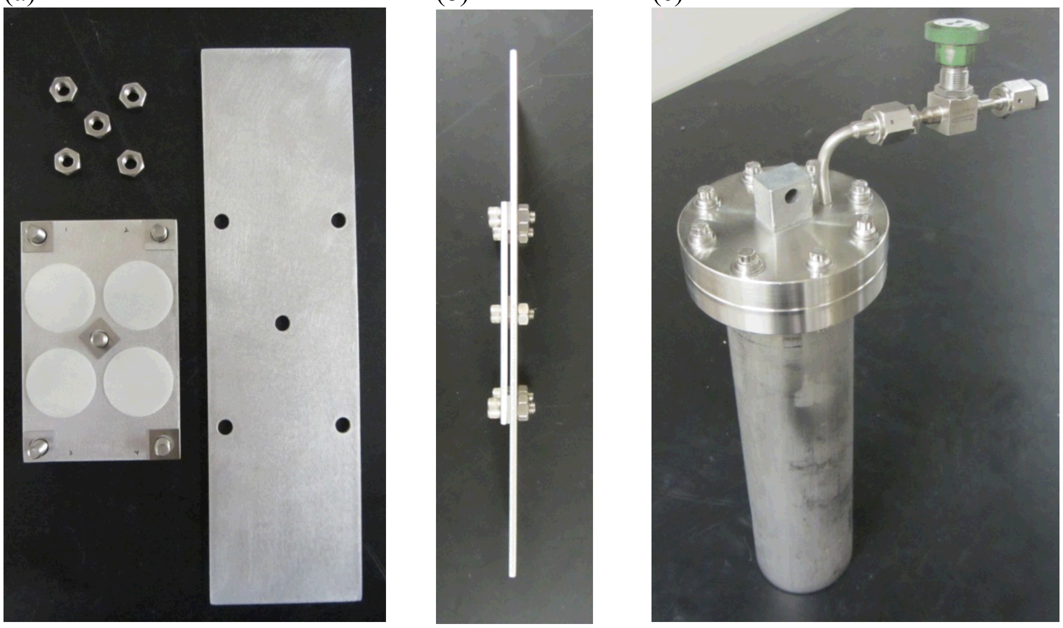
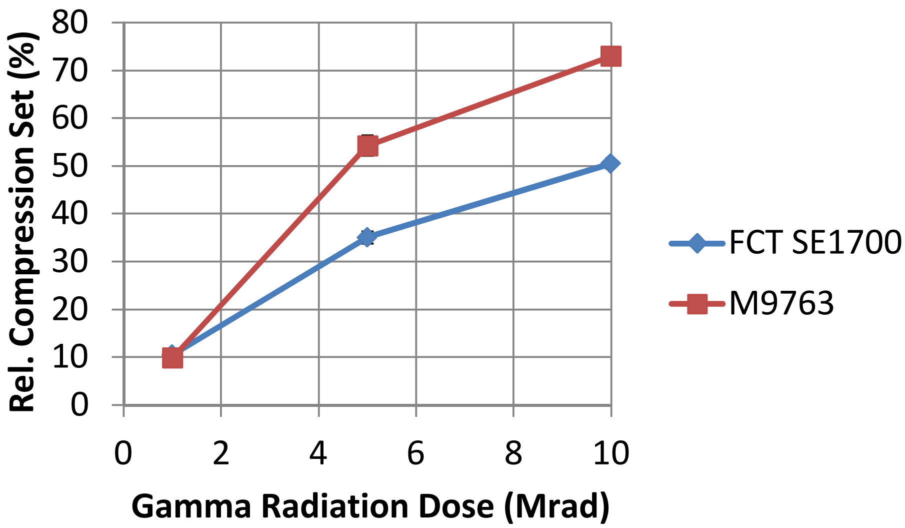


Fig. 2. Compression rig shown (a) disassembled and (b) assembled. The longer plate served to vertically position the specimens at the maximum dose rate zone in the Co-60 pool. The sealed vacuum vessel containing the assembled rig is shown in (c).

(a)



(b)

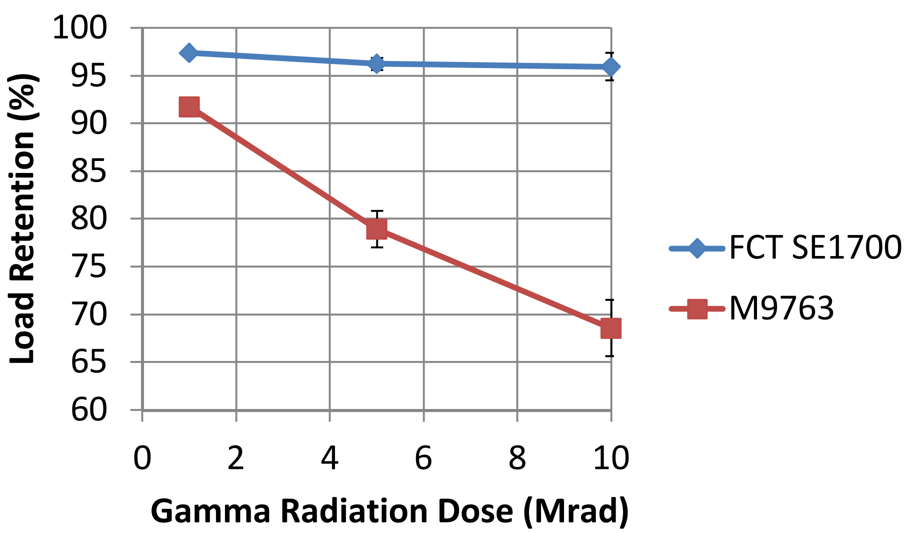
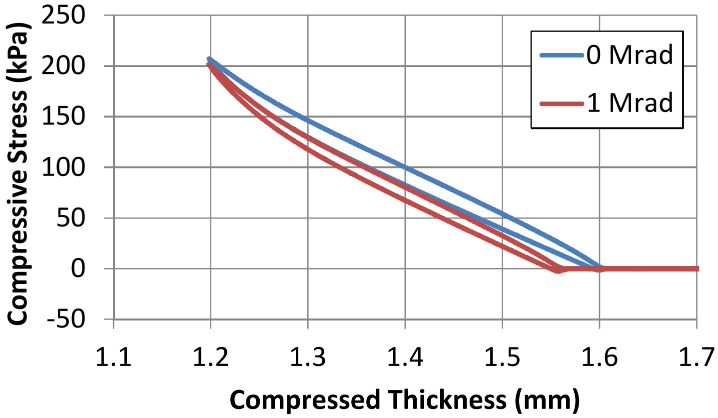
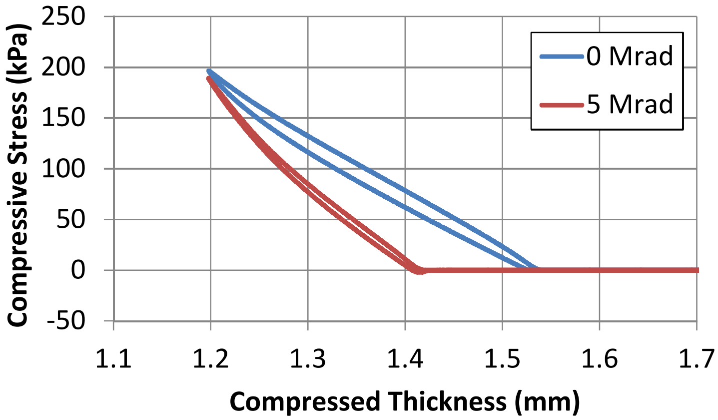


Fig. 3. Average (a) compression set (relative to deflection) and (b) load retention of the SE 1700 DIW porous specimens and M9763 cellular silicone as a function of radiation dose (n=4). Error bars represent the standard deviation.

(a)



(b)



(c)

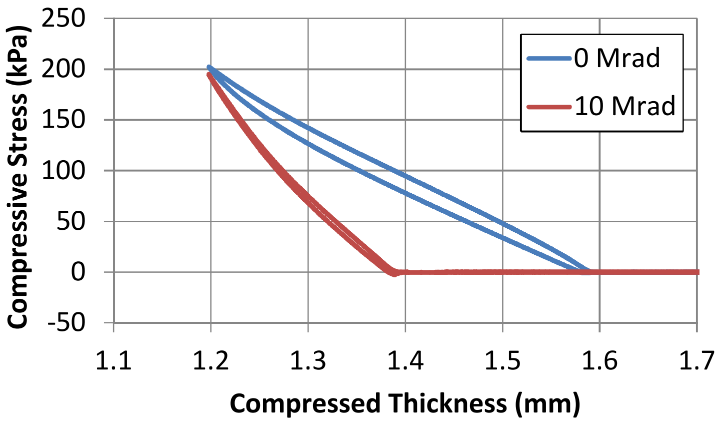
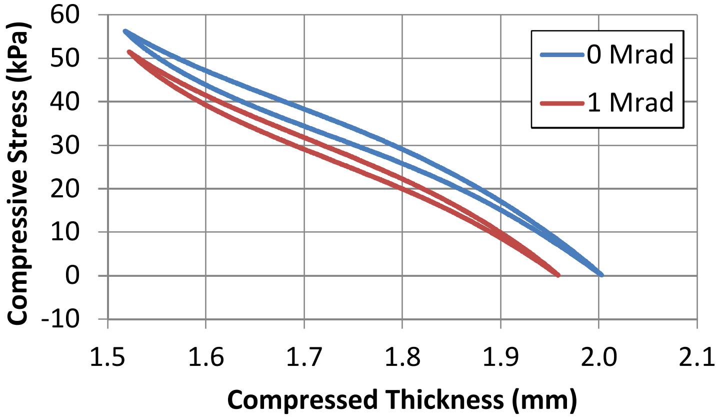


Fig. 4. Representative stress vs. compressed thickness curves for SE 1700 DIW porous specimens irradiated at (a) 1 Mrad, (b) 5 Mrad, and (c) 10 Mrad. The third load-unload cycle is shown. The pre-aging (0 Mrad) curves are shown for comparison.

(a)



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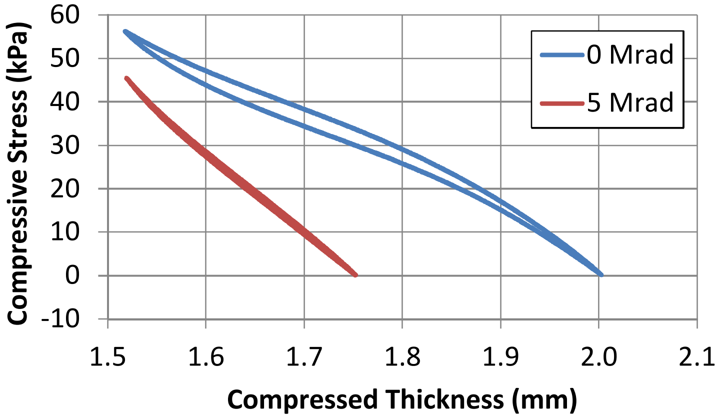

(c)

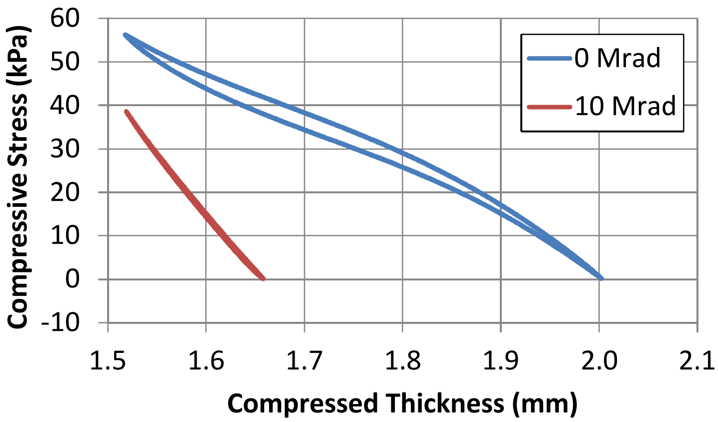

Fig. 5. Representative stress vs. compressed thickness curves for M9763 specimens irradiated at (a) 1 Mrad, (b) 5 Mrad, and (c) 10 Mrad. The third load-unload cycle is shown. The pre-aging (0 Mrad) curves are shown for comparison.

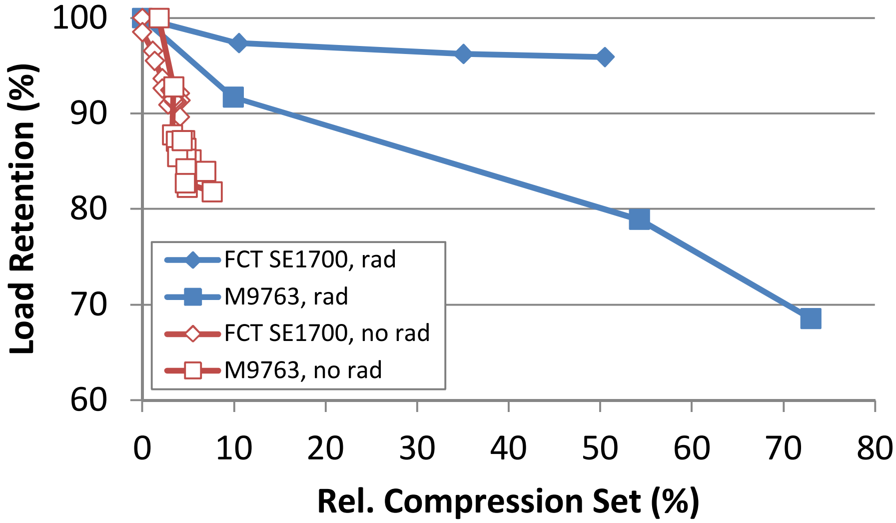

Fig. 6. Load retention vs. compression set for DIW SE 1700 and M9763 aged under ~25-30% compressive strain at room temperature with and without radiation exposure. Data out to 1 year is shown for the non-irradiated materials (the M9763 was aged for 2 years, but only the first year data is shown here).