

# Effect of Carbon Coating on the Properties of Gamma Irradiated Ultra-High-Molecular-Weight Polyethylene Specimens

*D. P. Mukherjee, A. L. Ogden, and R. H. Mayeux*

Department of Orthopaedic Surgery, Louisiana State University Medical Center, 1501 Kings Highway, Shreveport, LA 71130

*U. Siriwardane and H. Patel*

Department of Chemistry, Louisiana Tech University, Ruston, LA 71272

**ABSTRACT:** We reported the effect of carbon coating on the changes in properties of the ultra-high-molecular-weight polyethylene standard material (Hospital for Special Surgery) after gamma irradiation in air and storage for 2 years. The coating showed a slight improvement in crystallinity (X-ray) and tensile properties (under cyclic loading) over the uncoated and irradiated control group. The oxidation level as measured by Fourier-transform infrared spectroscopy was unaffected by coating.

## I. INTRODUCTION

The generation of wear particles from ultra-high-molecular-weight polyethylene (UHMWPE) liners in total hip and knee arthroplasties and the biological interactions of these particles are subjects of a number of investigations.<sup>1-3</sup> The wear of UHMWPE parts after gamma sterilization in air was higher than those sterilized by either ethylene oxide or gas plasma as well as those gamma sterilized while packaged in an inert atmosphere.<sup>4</sup> Further examination of the gamma-irradiated UHMWPE samples showed the presence of subsurface white band about 2 mm below the surface.<sup>5,6</sup> The examination of the gamma-irradiated and aged UHMWPE parts by Fourier-transform infrared spectroscopy (FTIR) showed the presence of oxidative products. The Electron Spin resonance (ESR) studies of these samples showed the presence of free radicals.<sup>7</sup> From these studies the picture has emerged that free radicals are formed during gamma irradiation, which react with the available oxygen resulting in some of these changes in the UHMWPE samples.

It is apparent that two basic approaches may be taken to reduce the wear of irradiated UHMWPE specimens: (1) the reduction of the concentration of free radicals and (2) removal of oxygen. The cross-linking at higher levels of radiation followed by heat treatment is showing some promise to reduce the wear of UHMWPE parts.<sup>8,9</sup>

We hypothesized in this study that the application of a thin carbon coating to UHMWPE might provide a nonoxidative atmosphere in the gamma-irradiated samples, thereby reducing the available oxygen. It may also reduce the generation of free radicals.

## II. MATERIALS AND METHODS

Disks made from the reference material Poly Hi Solidur, Hoechst, Celanese, UHMWPE (Hospital for Special surgery) were cut and machined to a thickness of 10 mm and 76.2 mm diameter. The disk samples were then sent to Lawrence Livermore National laboratory (Livermore, California) for carbon coating using their proprietary chemical vapor deposition process. These coated samples along with the uncoated controls were gamma irradiated at the nominal dose of 25 kgray at Isomedix (Morton Grove, Illinois). These samples were then stored for approximately 2 years before sectioning.

Sectioning of each sample required machining a channel though each side of the disk. A 9.5-mm ball endmill with a 7.9-mm radius was used in a milling machine and driven to a depth of 3 mm into each side of the disk and machined across the diameter. The disks were then placed in a microtome and sliced perpendicular to the channel yielding specimens of "dog-bone shape". The specimens' dimensions were 76.2 mm long, 10 mm wide and 4 mm wide at the "dog-bone" section. The thickness of microtomed sections ranged from 0.005 to 0.005 mm.

X-ray diffraction and FTIR spectroscopy were performed on the "dog-bone" specimens. The powder diffraction patterns of two samples of each of the groups were obtained using a Scintag X series (X-1/PAD5 MZ4) diffractometer with a goniometer radius of 250 mm. The range of the scan was from 2.00 to 79.98 degrees with a scan rate kept to 2.00 degrees/min for recording accurate intensity peaks. All scans were run continuously.

FTIR characterization was conducted using a Galaxy 2025 FTIR instrument. The range of the spectra was between 400 to 4000  $\text{cm}^{-1}$ . The resolution was set at 4  $\text{cm}^{-1}$ .

The tensile tests were carried out in an Instron (Canton, MA) Mechanical Testing Machine (Model 4202), with a 10 kN load cell. The gage length of each sample was kept to 30 mm. Each tensile test was carried out at a rate of 25 mm per minute. Each specimen was first cycled ten times between 10 to 20 N tensile force.

The specimens, which survived the cycling, were then taken to failure. All data was recorded on a Gateway 2000 P5-90 computer running LabVIEW 4.1 at a sampling rate of 10 per second. The results were tabulated using Microsoft Excel '97.

## III. RESULTS

The data obtained from the X-ray diffraction and IR spectroscopy are as follows:

**TABLE 1**  
**X-Ray Diffraction Average Intensities**

UHMWPE Sample	Average Intensities and Range		
	Peak 1 (19.3°–19.9°)	Peak 2 (21.4°–22.1°)	Peak 3 (24.0°–24.6°)
Gr1: uncoated and nonirradiated	1447.85	2617.80	1088.8
Gr2: uncoated and irradiated	1882.95	4577.25	1847.5
Gr3: carbon-coated and nonirradiated	1544.40	3305.00	1258.7
Gr4: carbon-coated and irradiated	1747.10	3908.30	1495.2

**TABLE 2**  
**FTIR Spectroscopy: (Galaxy 2025 FTIR)**

UHMWPE	FTIR characterization at 1717 cm <sup>-1</sup> wave number		
	Specimen no.	Observation	Absorbance (%)
Gr1: uncoated and nonirradiated	1	No peak	—
	2	No peak	—
Gr2: uncoated and irradiated	1	Peak	0.15
	2	Peak	0.17
Gr3: carbon-coated and nonirradiated	1	No peak	—
	2	No peak	—
Gr4: carbon-coated and irradiated	1	Peak	0.16
	2	Peak	0.15

**TABLE 3**  
**Mechanical Properties**

UHMWPE	Failure strength (N)	Failure elongation (mm)
Gr1: uncoated and nonirradiated	28.6 ± 5.4	20.6 ± 7.5
Gr2: uncoated and irradiated	24.41 ± 5.2	23.5 ± 6.1
Gr3: carbon-coated and nonirradiated	29.0 ± 3.9	24.8 ± 4.4
Gr4: carbon-coated and irradiated	23.7 ± 5.4	24.2 ± 6.0

#### IV. DISCUSSION

The X-ray data showed that the peak intensities (indicating crystallinity) at the 21.4 to 22.1° Bragg angle were in the following order: Group 2 (4577) > Group 4 (3908) > Group 3 (3305) > Group 1 (2618). The FTIR data showed that irradiated coated and uncoated samples showed approximately the same degree of oxidation as seen from the absorbency of the peak intensities at 1717  $\text{cm}^{-1}$  wave number. The nonirradiated samples of both coated and uncoated samples showed the absence of this carbonyl peak, indicating no oxidation. The mechanical properties showed that the average failure strengths of the nonirradiated, but coated and uncoated specimens were 29.0 N and 28.6 N, respectively. The average failure strengths of the irradiated but coated and uncoated specimens were 23.7 N vs. 24.4 N, respectively. These differences were not statistically significant. More insight may be obtained from the survival frequency data under the cyclic load that showed a very distinct behavior (Table 4). The uncoated and nonirradiated samples were the best (100%), whereas the uncoated and irradiated samples were the worst (40%). The frequency of survival for the gamma-irradiated samples after cyclic loading improved with coating (65%). The cyclic loading data would indicate that carbon coating reduced the development of the brittle nature in these specimens after irradiation when compared with the uncoated and irradiated specimens. The X-ray data also confirm this trend showing a lesser degree of crystallinity when compared with the uncoated and gamma-irradiated specimens. The data taken collectively would indicate that the carbon coating reduced the development of higher crystallinity and improved the tensile properties but did not show any improvement of the oxidation.

#### V. CONCLUSION

The carbon-coated UHMWPE specimens showed some trend toward improvement of crystallinity and frequency of failure under cyclic tensile loads but the oxidation level did not change when they were irradiated.

**TABLE 4**  
**Survival Frequency During Cyclic Test**

UHMWPE	Survival (%)
Gr1: uncoated and nonirradiated	100
Gr2: uncoated and irradiated	40
Gr3: carbon-coated and nonirradiated	90
Gr4: carbon-coated and irradiated	65

## REFERENCES

1. Shanbag, A. S., Jacobs, J. J., Black, J., Galante, J. O., and Glant, T. T., Effects of particles on fibroblast proliferation and bone resorption in vitro, *Clin. Orthop.*, 342, 205-217, 1997.
2. Jasty, M., Goetz, D. D., Bragdon, C. R., Lee, K. R., Hanson, B. A., Elder, J. R., and Harris, W., Wear of polyethylene acetabular components in total hip arthroplasty. An analysis of one hundred and twenty-eight components retrieved at autopsy or revision operations, *J. Bone Joint Surg.*, 79-A, 349-358, 1997.
3. Wimmer, M. A., Andriacchi, T. P., Nataraajan, R. N., Loos, J., Karlhuber, M., Petermann, J., Schaeider, E., and Rosenberg, A. G., A striated pattern of wears in ultra-high-molecular-weight polyethylene components of Miller-Galante total knee arthroplasty, *J. Arthroplasty*, 13(1), 8-16, 1998.
4. Collier, J. P., Sutula, L. C., Currier, J. H., Wooding, R. E., Williams, I. R., Farber, K. B., and Mayor, M. B., Overview of polyethylene as a bearing: comparison of sterilization methods, *Clin. Orthop.*, 333, 76-86, 1996.
5. Li, S. and Burstein, A. H., Current concept review: ultra-high molecular-weight polyethylene, *J. Bone Joint Surg.*, 76-A, 1080-1090, 1994.
6. Rimnac, C. M., Klein, R. W., Betts, F., and Wright, T., Post-irradiation aging of ultra-high-molecular-weight polyethylene, *J. Bone Joint Surg.*, 76-A, 1052-1056, 1994.
7. Jahan, M. S., Stovall, J. C., Davidson, J. A., and Hines, G., Long-term effects of gamma-sterilization on degradation of implant materials, *Appl. Radiation Isotopes*, 26(6-7), 637-638, 1995.
8. McKellop, H. and Shen, F.-W., Tribology and current status of crosslinked polyethylene for bearing articulation, *Proc. 66th Annual Meeting, American Academy of Orthopaedic Surgeons*, Anaheim, CA, 1999, 309-312.
9. Harris, W., Electron beam irradiation and melting, *Proc. 66th Annual Meeting, American Academy of Orthopaedic Surgeons*, Anaheim, CA, 1999, 464-465.