

EFFECT OF NUCLEAR RADIATION ON MAGNET INSULATION IN HIGH-ENERGY ACCELERATORS*

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Summary

Magnet coil insulation consists of inorganic materials such as glass fiber, fillers, and organic thermosetting resins. Most of the usual insulation systems are damaged if subjected to high radiation levels such as those anticipated at the Stanford Linear Accelerator, where the expected dose rate may be as high as 10^{11} rads per year. This paper describes tests carried out at SLAC in which several commercially available insulations were found to deteriorate severely at total absorbed doses of as low as 10^{10} rads. Changes in molecular structure induced by radiation effects and degradation of electrical and mechanical properties are discussed. The development and testing of an organic insulation using pure Al_2O_3 filler which retained about 25% of its original strength at an absorbed radiation level of 3.25×10^{14} ergs·gr⁻¹ will be described. This insulation required minor changes in the usual winding and potting technique prevalent in the industry.

I. Introduction

Radiation damage to organic materials, cables, and coil insulations is well known in high energy accelerators with high intensity beams. The sensitive parts in accelerator magnets are the coils, electric leads, and water connections. Magnet coil insulation consists of a supporting structure, i.e., glass fiber cloth, an organic binder, and frequently in addition to the glass and binder, additional electric insulation such as mica fleecy.

The glass mica structure is impregnated with suitable thermosets such as epoxies, silicones, or unsaturated polyesters; this impregnation can be accomplished under vacuum and pressure, or the binder can be contained in a pre-impregnated tape (B-stage thermosetting). Application of heat and pressure leads to a hard and compact structure which withstands mechanical and magnetic forces, thermal and electrical stresses, and the influence of moisture, chemical vapors, and dust. The insulations of high current magnet coils are stressed close to their rupture limit and fatigue over the magnet lifetime.

In addition to these stresses, the magnets are placed in a radiation environment which influences the mechanical, thermal, and electrical characteristics of the insulation. Conventional water connections from the manifolds to the individual cooling passages through the magnet must be flexible and provide the necessary insulation for the magnet. These connections consist of rubber hoses or other thermoplastic tubes which, under the influence of radiation, change their elasticity and become brittle so that they have to be replaced by ceramic tubes and stainless steel bellows (see Fig. 1) since radiation effects in polymers are irreversible.

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Changes in organic thermosets may be classified under:

- a. Permanent changes in appearance, color effects.
- b. Chemical changes, such as double bond formation cross linking, oxidative degradation, polymerization, depolymerization, and gas evolution.
- c. Physical changes, such as electrical and thermal conductivity, heat distribution.
- d. Mechanical changes, such as changes in tensile and flexural strength, Young's modulus, hardness, elongation, and flexibility.

Several reactions may occur simultaneously under irradiation, but the initial effect is a "curing" process which improves tensile strength and hardness by increasing cross links. Radiation and local heat build-up are so efficient that the end result is always a binder so highly cross linked as to be fragile and crash sensitive. At this point hardness and Young's modulus may be great, but the binder cannot withstand any mechanical stresses. The binder finally becomes brittle and disintegrates into a black powdery substance. Irradiation makes the binder more susceptible to oxidation and moisture absorption.

Irradiated glass reinforced insulation shows the following characteristics:

- Stage I. Discoloration of the organic binder.
- Stage II. Degradation and deterioration of the organic binder.
- Stage III. Disintegration of mica.
- Stage IV. Disintegration of any inorganic fillers.
- Stage V. Degradation of the glass cloth.

The different stages occur at different absorbed irradiation doses and have to be treated separately.

The organic binder consists primarily of carbon, hydrogen, oxygen, and perhaps nitrogen and halogen atoms linked together in various ways by covalent forces. Covalent forces can be ruptured by addition of radiation energy to the structure. Where covalent forces are broken, new bonds are formed and the structure is changed. Weaker bonds are ruptured first, such as bonds between C-H and O atoms. Resonant bonds are the most stable. With increase in irradiation dose, bonds of equal strength remain in the structure. The contribution to insulation strength from the resin is apparently small after decarbonization. It seems that there is some contribution from the intrinsic strength of the resin at intermediate states, which is obvious from retardation effects observed in experiments.

Glass fiber reinforcement and filling of the resin with a mineral filler improve radiation resistance considerably. This effect is not clearly understood, but an explanation based on fact and speculation is given in the following sections.

A variety of thermosets were tested under radiation at Stanford's Mark IV linear accelerator and in a pool type reactor at the Lawrence Radiation Laboratory in Livermore. Epoxies, polyesters, and silicones were prime candidates. Detailed test results have been published.¹ A thorough investigation of irradiation effects on elastomers and organic compounds has been done by J.R. Kircher and R.E. Bowman.² Detailed results on properties of glass fibers and fillers may be obtained from S. O'Leesky and G. Mohr.³

This report is concerned primarily with epoxies, and only test results of some other thermosets are given.

Table I gives the relative radiation resistance of thermosets at room temperature without and with glass fiber cloth reinforcement, unfilled and mineral filled.

composition with 2-3 times strength of one epoxy blended with aromatic curing agent above. These mixtures have also high radiation resistance, i.e., Epon 828 and 1031 (1:1 ratio) and curing agent Z (Shell Chemical Corporation) or DEN 438 and DER 332 LC (1:3 ratio) and curing agent MPDA and MDA (2:3 ratio) has a radiation resistance of 2 times higher than Epon 828 or DER332LC. The blend of epoxies result in a higher viscosity of the mixture and for many applications this increase leads to difficulties in a void free impregnation. The addition of mineral fillers such as granulous pure Alumina (10-20 microns grain size) improves the radiation resistance, but reduces the initial strength of the mix.

Adding Dow Corning's Z6040 (1 part per weight of the epoxy) restores the mechanical strength. In order to keep the filler in suspension it seems

TABLE I

RELATIVE RADIATION RESISTANCE OF THERMOSETTING RESINS AT ROOM TEMPERATURE

Resin	Radiation dosage (ergs.gr ⁻¹) required for:			
	threshold damage	25% damage	50% damage	90% damage
<u>Epoxy</u>				
Unfilled ^{a)}	2×10^{10}	3.2×10^{11}	10^{12}	7×10^{12}
Laminated, glass fiber ^{b)}	2.5×10^{11}	2.65×10^{12}	7.4×10^{12}	$\approx 2 \times 10^{13}$
Mineral filled ^{c)}	7×10^{10}	5×10^{11}	3×10^{12}	$\approx 10^{13}$
Mineral filled and laminated, glass fiber	8×10^{11}	5×10^{12}	1.25×10^{13}	3.5×10^{14}
<u>Polyester</u>				
Unfilled ^{d)}	5×10^7	1.2×10^8	5×10^9	-
Laminated, glass fiber ^{b)}	8×10^{10}	5×10^{11}	10^{12}	-
Mineral filled	9×10^9	1×10^{11}	4×10^{11}	-
<u>Silicone</u>				
Unfilled ^{e)}	10^{10}	4×10^{10}	2×10^{11}	-
Laminated, glass fiber ^{b)}	1×10^{11}	10^{12}	6×10^{12}	-
Mineral filled	1×10^{11}	-	-	-

a) Epoxy: DER 332 LC and curing agent MPDA and MDA. (Dow Chemical Corp., Freeport, Texas.)

b) Medium weave, Volan A treated fiberglass. (Owens-Corning Corp.)

c) Alumina 900 mesh. (10-20 microns grain size max.)

d) Unsaturated, low pressure, low viscosity polyester resin.

e) Silicone resin: R-7521, curing agent dicumyl peroxide and zircon filler. (Dow Corning Corp., Midland, Michigan.)

A classification of polymers with respect to their stability against radiation is given in Table II.

We conclude that polymers containing benzene ring formation in side groups are the most stable ones. "Aromatic Compound" lend stability to polymers and "Aliphatic Structures" shows poor radiation resistance. Aromatic hardeners is therefore preferred in combination with epoxies.

A distinct connection between radiation resistance and the "strength of char" of different epoxies blended with aromatic amines result in charred

necessary to add a colloidal silica, such as Cab-c-sil (Godfrey L. Cabot, Inc.) to the mix, (1 part per weight of the epoxy).

In some cases, particularly when long pot life is required the above mentioned hardeners are not suitable. A substitution is to use Lewis acid BF₃ MEA (Shell Chemical Corporation) which results in a pot life of several days but reduces the radiation resistance of the epoxy mix. (Figure 2.)

TABLE II
POLYMER UNITS AND THEIR RESISTANCE TO RADIATION

Simplified Polymer Unit	Threshold Radiation Value (ergs/gr)		Characteristics
	Unfilled resin	filled	
$\begin{array}{c} \text{H} \quad \text{H} \\ \quad \\ -\text{C} - \text{C}- \\ \quad \\ \text{O} \quad \text{H} \end{array}$	$10^{10} - 10^{11}$		Stable polystyrene with benzene ring in side chain (thermoplast)
$\begin{array}{c} \text{O} \\ \\ \text{O}-\text{CH}_2-\text{CH}-\text{CH}_2 \\ \\ \text{O}-\text{CH}_2- \end{array}$	$(4.0-8.8) \times 10^{10}$	2×10^{12}	Epoxy chain with aromatic type curing agent
$\begin{array}{c} \text{H} \quad \quad \text{O} \\ \quad \quad \\ \text{C} - \text{C} - \text{C} - \\ \quad \quad \\ \text{H} \quad \quad \text{H} \end{array}$	$10^7 - 10^8$	10^{11}	Polyester chain
$\begin{array}{c} \text{CH}_3 \\ \\ \text{Si} - \text{O} - \\ \\ \text{CH}_3 \end{array}$	10^{10}	2×10^{11}	Basic silicone chain (dimethylsilane)
$\begin{array}{c} \text{OH} \quad \text{H} \\ \quad \\ \text{C} - \text{C} - \\ \quad \\ \text{H} \quad \text{H} \end{array}$	$10^8 - 10^9$	4×10^{10}	Phenol formaldehyde polymer (increased cleavage due to radiation)
$\begin{array}{c} \text{F} \quad \text{F(Cl)} \\ \quad \\ -\text{C} - \text{C}- \\ \quad \\ \text{F} \quad \text{F} \end{array}$	2×10^6	-	Teflon (poor radiation resistance)

II. Expected Irradiation Dose in a High Energy Accelerator

This problem has to be solved for particular applications and particular types of particles separately. For the SLAC electron linear accelerator the expected radiation dose in magnet coils operating over a period of 10 years is calculated by Brechna.¹ The calculation under simplified assumptions showed that radiation dose up to 2×10^{13} ergs·gr⁻¹ can be expected in some areas where the magnet is located near collimators.

III. Radiation Effects on Coil Insulations

1. Mechanical Properties

a. Bond Strength. The bond strengths of the insulation to the conductor did not change appreciably up to an absorbed radiation dose of 7×10^{10} ergs·gr⁻¹ (Fig. 2).

The initial bonding strength to the conductor was measured for mineral filled Epoxies: $\sigma_{b0} = 62$ kg·cm⁻²; for glass fiber reinforced, mineral filled thermosets: $\sigma_{b0} = 183$ kg·cm⁻²; glass fiber reinforced ceramics: $\sigma_{b0} = 22$ kg·cm⁻² and for laminated and filled silicon: $\sigma_{b0} = \sim 70$ kg·cm⁻².

The best results achieved were with medium weave, glass fiber tape Volan A treated, impregnated under vacuum with epoxy DER332LC (100% parts per weight) and hardener MPDA + MDA (18% parts per weight), 10-20 microns grain size pure alumina granulate (120% parts per weight), and a thixotropic material such as colloidal silica (cab-o-sil) (1 part per weight). At an absorbed radiation dose of 3.25×10^{14} ergs·gr⁻¹, the insulation still had a bond strength of 20.3 kg·cm⁻².

b. Compression and Impact Strength. The compression strength changes as a function of irradiation dose in the same rate as the bond strength. Unfilled, mineral filled, and glass fiber reinforced epoxies were tested and their compression strength as a function of radiation dose is shown in Fig. 3. Laminated, mineral filled DER 332LC and MPDA + MDA hardener retained their compression strength up to 3.6×10^{11} ergs·gr⁻¹. The initial compression strength value of unirradiated samples was 3250 kg·cm⁻².

Measurements of the impact strength, performed with the Baldwin pendulum cantilever beam impact testing machine of the Izod type show that glass fiber reinforced and mineral filled epoxies lose 24% of their initial value at 4.9×10^{12} ergs·gr⁻¹. The impact strength is practically zero at irradiation doses of $> 10^{14}$ ergs·gr⁻¹.

2. Electrical Properties

a. Volume Resistivity. The volume resistivity of alumina filled glass fiber reinforced DER 332LC and hardener MPDA and MDA changed from 5.86 $\times 10^{14}$ ohms·cm to 1.755×10^{11} ohms·cm when exposed to 1.1×10^{14} ergs·gr⁻¹, and to 7.8×10^{10} ohms·cm when exposed to the absorbed radiation dose of 3.25×10^{14} ergs·gr⁻¹. (Figure 4.)

b. Dielectric Strength. Samples tested were exposed to 500 volts dc continuously during irradiation. The leakage current was measured and recorded as a function of irradiation dose. The speculation that insulation materials become semi-conductors when exposed to high radiation doses could not be verified. However, it is interesting to note that even after complete breakdown of mechanical properties, the insulation resistance of dry irradiated samples is still more than 50% of the original value prior to radiation. Upon exposure to moisture, a rapid degradation of the insulation resistance occurs.

c. Moisture Absorption. Moisture absorption is accelerated by irradiation. The greater affinity of the glass fiber reinforced epoxy system toward moisture absorption is probably due to breaking of the binder around and along the glass fibers.¹

IV. Dosimetry

The test setup was chosen such as to simulate the actual coil design. Hollow copper conductors were insulated with glass fibers and impregnated under vacuum with suitable thermosettings. The water was passed through the hollow conductor; the water temperature rise due to absorbed irradiation energy and was measured and recorded (Fig. 5). Measuring the amount of water per unit time cooling the samples and the water temperature rise provides a fairly accurate way of determining the absorbed radiation dose. Most tests were performed at the Mark IV accelerator with an electron beam energy of 60-70 MeV and an average beam current of 16-30 μ a at 360 pulses per second, corresponding to an average beam power of 960-2100 watts. The measured weight of the irradiated part of the sample was approximately 15 grams. The irradiation flux corresponds to a maximum of 4.66×10^8 ergs/gr/sec.

V. Metallographic Techniques

It may be seen from Table I and Fig. 2, that glass fiber reinforced and mineral filled thermosets are more radiation resistant than unfilled thermosets. An explanation for this phenomena can not be given with certainty, but the following reasons are possible:

- a. Formation of a more rigid structure.
- b. Better distribution of the beam flux.
- c. Devitrification of the glass fiber.
- d. Intrinsic strength of the resin.
- e. Sintering of the filler.

The effect of electron beam irradiation on the insulation was investigated by metallographic techniques and X-ray diffraction methods. Microscopic observation showed a bright fringe around the glass fiber in irradiation composites (Fig. 6), which may be related to devitrification of the glass fiber. The epoxy had changed into a black powder, but no change in the lattice and crystal structure of the alumina filler was evident. A sintering effect due radiation energy on the filler and glass fiber could not be seen at this stage. We may conclude¹ from the test that the present reinforcement effect of glass fibers as insulation in radiation environ-

ments is to a certain degree correlated to a healing of the "flaws" and "cracks" on the glass surface, devitrification, and the intrinsic strength of the epoxy used.

VI. Manufacturing Problems

The necessity of using highly filled thermosets which have an initial viscosity of 1000-3000 cP at impregnation temperature initiates difficult manufacturing problems. In medium and tight weave glass fiber cloths, the filler may filter and block the paths of the resin through the fiber.

Three methods have been investigated and prove to be adequate impregnation techniques: wet winding, vacuum impregnation, and B-staged tapes.

a. Wet-Winding. The filled thermoset is applied to the insulation by means of brushing and spraying at room temperature. The excess of material is pressed out during the uniform all-side compression of the coils. The coils are heated, impregnated under vacuum with the same filled resin used for wet winding, pressed to final specified dimensions, and cured. This technique required a long pot life of the resin system, which can be obtained by using BF₃MEA hardener.

b. Vacuum Impregnation. Conductors are wound with dry, medium or tight weave fiber glass and impregnated with a suitable thermoset under vacuum. This technique requires less binder viscosity (less than 1000 cP) and delicate maneuvering of the vacuum and heat during impregnation and curing. A post application of pressure prior to curing improves the quality of the impregnant. The best result was achieved by using DER 332LC epoxy, MPDA and MDA hardener, and 10-micron alumina granulates.

c. B-Staged Tapes. B-staged tapes make use of partially cured epoxies contained in the glass fiber tape. This so-called binder has a pot life of several months at 10°C or less. The binder can be filled with a mineral filler and brought into a B-stage phase to the tape. After wrapping, the coil insulation is pressed to final dimension and cured.

VII. Conclusions

From a number of different thermosets, epoxies have been found to be adequate impregnants for magnet insulation. From various epoxy systems tested we may conclude the following:

(a) Glass fiber reinforced and mineral filled pure epoxy and aromatic amines show the highest radiation resistance.

(b) Systems with the highest equivalent of epoxy per hardener and aromatic hardener give best results. Excess of epoxy stabilizes the system to radiation.

(c) Epoxies with a high heat distortion temperature are more stable.

(d) In order to achieve optimum results, 25-30% of the insulation volume should be occupied by glass fiber. Up to 50-70% of the volume is filled by the filler, mica fleece, and the rest of the volume (20-30%) by the epoxy system.

(e) Specific epoxy systems such as DER332LC and hardener MPDA and MDA, 18 parts per weight of

the epoxy, and Dow Corning Z6040 (1 part per weight) filled with granular alumina, 120 parts per weight of the epoxy, retained 25-30% of their original mechanical properties at the absorbed radiation dose of 2×10^{14} ergs \cdot gr $^{-1}$.

References

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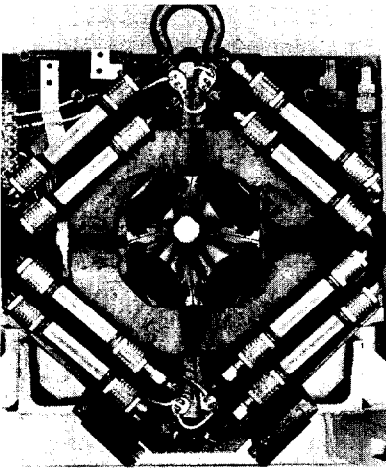


Fig. 1. 8 cm quadrupole.

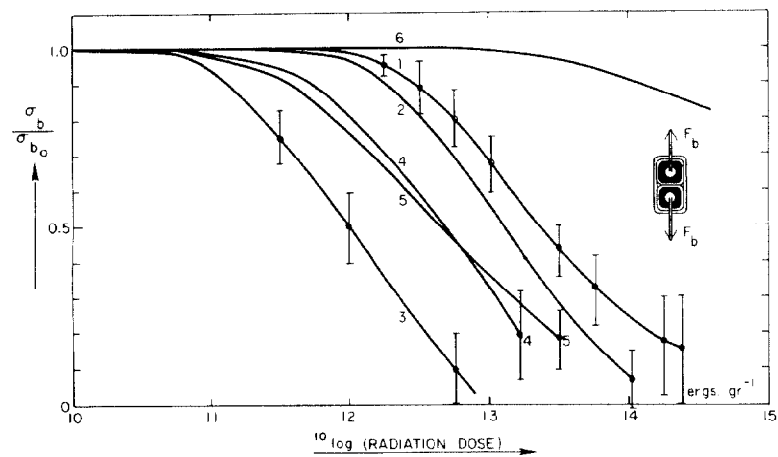


Fig. 2. Bond strength of glass fiber reinforced and mineral-filled thermosettings and ceramics.

1. DER 332 LC and curing agent MPDA and MDA wetting agent Z6040 ($\sigma_{b_1} = 183 \text{ kg cm}^{-2}$).
2. Epon 828/1031 curing agent NMA and BDMA ($\sigma_{b_1} = 189 \text{ kg/cm}^2$).
3. Emerson and Cuming 2850 FT no glass fiber reinforcement ($\sigma_{b_1} = 62 \text{ kg cm}^{-2}$).
4. DER 332 LC and curing agent BF_3MEA ($\sigma_{b_1} = 175 \text{ kg cm}^{-2}$).
5. Dow Corning R-7521 silicone and curing agent dicumyl peroxide; zircon filler ($\sigma_{b_1} = 70 \text{ kg cm}^{-2}$).
6. Eccoceran part A and B ($\sigma_{b_1} = 22 \text{ kg cm}^{-2}$).

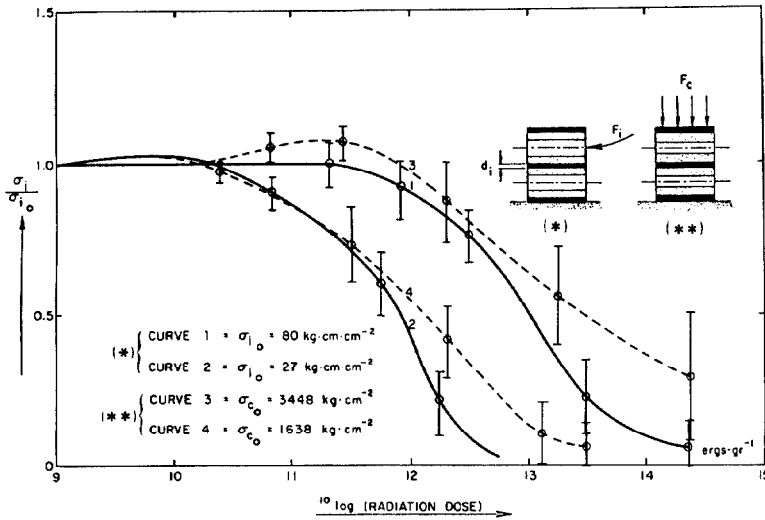


Fig. 3. Relative compression strength of glass fiber reinforced, mineral-filled thermosettings as a function of absorbed radiation dose.

1. DER 332 LC and hardener MPDA and MDA (Al_2O_3 filler).
2. R-7521 silicone resin hardener dicumyl peroxide (zircon filler). Relative impact strength of glass fiber reinforced, mineral-filled thermosettings as a function of absorbed radiation dose.
3. DER 332 LC and MPDA and MDA (Al_2O_3 filler).
4. R-7521 silicone resin (zircon filler).

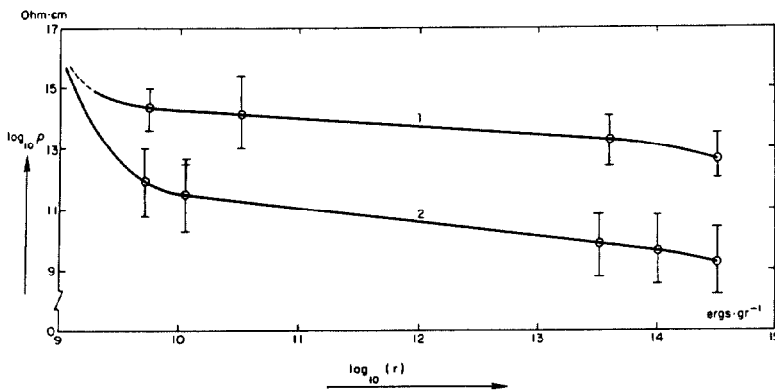


Fig. 4. Volume resistivity of glass fiber reinforced, mineral filled thermosettings as a function of absorbed radiation dose.

1. DER 332 LC hardener MPDA and MDA (Al_2O_3) filler.
2. R-7521 silicone resin hardener dicumyl peroxide (zircon filler).

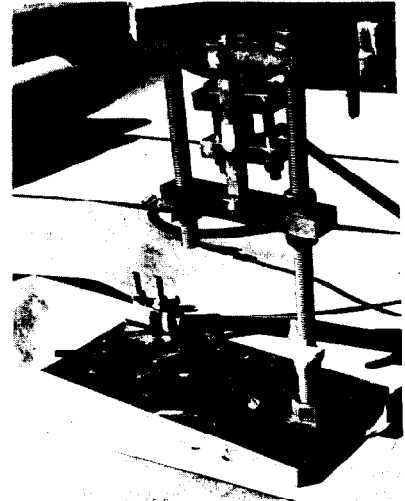


Fig. 5. Sample holders and sample for radiation tests.



Fig. 6. Irradiated sample (magnification 300). Absorbed radiation dose $2 \times 10^{13} \text{ ergs}\cdot\text{gr}^{-1}$ (Fringe in glass surface top).