Coupling of Glass Fiber with Polypropylene by Gamma Radiation

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Synopsis

The effect of ^{60}CO gamma radiation on polypropylene and glass-fiber-reinforced polypropylene was investigated. The glass fiber content in the reinforced polypropylene was observed to influence the mechanical properties of the original polypropylene. The initial modulus and off-set yield strength (at 0.25% strain) increased with the dose and glass fiber content. The tensile strength showed an initial decrease with the increase in glass fiber content followed by an increasing trend. A similar behavior was found with increasing dosage. The extent of fiber breakage during processing was higher at higher glass fiber content. However, the fiber length distribution became narrower beyond 2.45% glass fiber content in the polypropylene.

INTRODUCTION

Glass-fiber-reinforced plastics are well known for their several advantageous properties compared to conventional materials. Thermosetting resins have been used for a long time for the production of glass-fiber-reinforced plastics. On the other hand, thermoplastic resins have been used recently for the same purposes. Glass-fiber-reinforced nylon is commercially the most successful material in this regard. Hydrocarbon polymers, such as, polyethylene and polypropylene have not been very successful for composite applications, the reason being the lack of reactive sites on the polymer backbone to form a strong bond with glass fiber. Some coupling agents[1-4] have been used for increasing the interfacial bond strength in such a product, but still very satisfactory products have not been achieved.

Hydrocarbon polymers like polyethylene and polypropylene when exposed to gamma radiation have been known to produce polymeric free radicals.[5] The free radicals so generated lead to degradation of chains and/or production of crosslinks between polymeric chains. In the present study, polypropylene mixed with glass fiber have been exposed to gamma rays with a view to develop strong bonds involving polypropylene and glass fiber. The composite material so produced have been evaluated to ascertain the extent of reinforcement.

EXPERIMENTAL

Materials

Isotactic Polypropylene. Grade M 0030 having melt flow index 10 as supplied by Indian Petrochemicals Ltd. (IPCL), Vadodara, India, was used.

Glass fiber chopped strand of approximately 12.5 mm length, grade No. FGP-17 was supplied by Fibreglass Pilkington Ltd., Bombay.

Mixing Glass Fiber with Polypropylene

Polypropylene and glass fiber in various proportions were tumble-mixed for about 20 min followed by extrusion, using a Superneck extruder having $L{:}D$ ratio 26.8:1, with a screw speed 28 rpm and extrusion temperature 230°C. The extruded strands were chopped into grannules using a cutting machine.

Injection Molding

The grannules obtained by previous procedure were injection-molded for making test specimen using an R.H. Windsor SP-1 machine. The conditions used were as follows: (i) temperatures: zone 1, 215°C; zone 2, 225°C; nozzle temperature, 85°C. (ii) pressure: injection pressure, 49.032×10^5 N/m²; auxiliary pump pressure, 49.032×10^5 N/m². (iii) locking force, 27 tons; (iv) shot weight, 8.45 g. The mold used was without temperature control.

Irradiation

Irradiation was carried out at room temperature using a gamma ray source (Model GC-900) provided by Bhabha Atomic Research Centre (BARC), Bombay, at a dose rate of 0.32 Mrad/h for various dose values.

TESTING OF THE SAMPLES

Stress–Strain Analysis

These measurements were made over an Instron, floor model TTC-1, at 20°C and RH 65%, using a crosshead speed of 0.5 cm/min.

Fiber Length Measurements

The samples having various proportions of glass fiber were burnt in a muffle furnace at 450°C for 5 h followed by cooling and weighing of the residual glass fiber. A magnifying microscope of projectina model was used to evaluate the statistical distribution of lengths of fiber at a magnification of 100.

RESULTS AND DISCUSSION

Fiber Length Analysis

The properties of a composite material mainly depend upon the aspect ratio of the fiber, orientation of the fiber, fiber content, and extent of bonding between fiber and matrix.[6-12] For an efficient reinforcement, the aspect ratio of the fiber should be above a certain critical value, and it is, therefore, important that the fiber length not be excessively reduced during processing.[4,11,13]

From the histograms of various fiber lengths and their corresponding number fractions for original glass fibers and fibers incorporated in polypropylene in various proportions it was observed that the original fibers
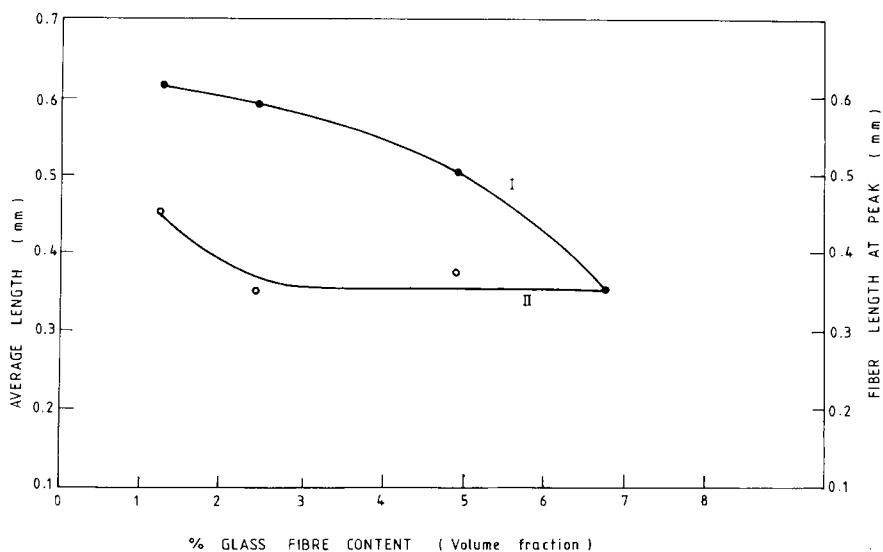
Fig. 1. (I) Plot of average fiber length with % glass fiber content (by volume) reinforced with polypropylene. (II) Plot of fiber length (at which maximum occurs in dFn/dL vs. L curve) with % glass fiber content (by volume) reinforced with polypropylene.

(i.e., fibers taken prior to mixing) were distributed in a range of 3–18 mm in length and concentrated at two fiber lengths, at 10.5 mm and 14.5 mm. During extrusion, the energy supplied and energy produced by friction fluidizes the polymer and leads to degradation in the length of the fiber. The extent of degradation with increasing glass fiber content is shown in Figure 1(I), in terms of average fiber length. The average fiber length linearly decreases with increasing glass fiber content. Jou and Wu[6] have also observed higher attrition at higher glass fiber content. Figure 1(II) represents the fiber length (l_{max}) (at which maximum occurs in the dFn/dL vs. L curve*) variations with glass fiber content. Initially l_{max} goes down and later on levels off with the increase in glass fiber content. Such behavior clearly indicates that there is a certain length of fiber ($\simeq$ 0.35 mm) below which the fiber breakage is least affected by increase in the glass fiber content during extrusion.

The dispersion of the length distribution has been taken into account by measuring the hald-width of dFn/dL curve (Fig. 2). Half-width shows an increase initially followed by a linear decrease with glass fiber content. Glass-fiber-reinforced polypropylene containing 2.45% glass fiber by volume exhibits the maximum dispersion of fiber lengths.

Mechanical Behavior

Effect of Irradiation on Initial Modulus

The mechanical behavior of polypropylene and glass-fiber-reinforced polypropylene have been investigated by a number of workers.[7–10,14] The

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* Here Fn stands for cummulative fraction and L for fiber length. dFn/dL vs. L curve is considered to smooth the length distribution curve.
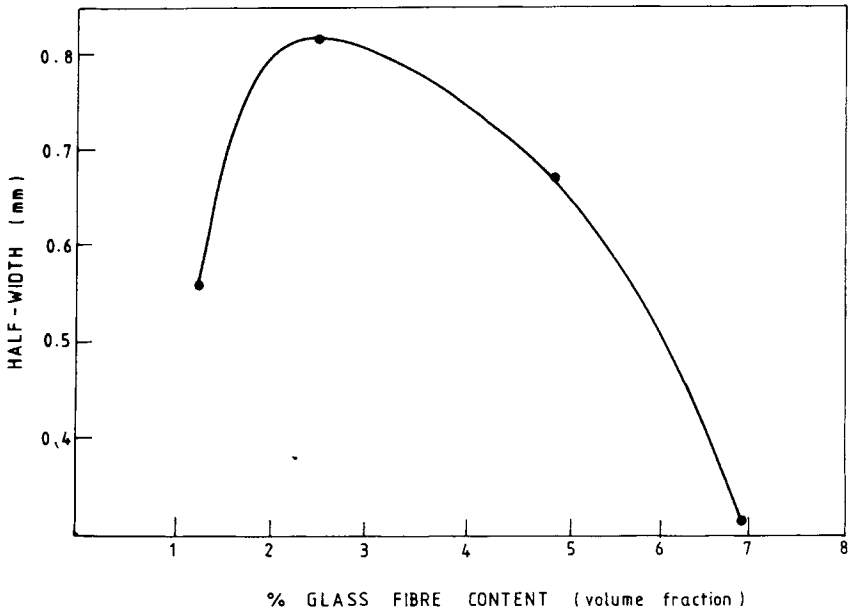
Fig. 2. Plot of half-width (evaluated by dFn/dn vs. L curve) vs. % glass fiber content (by volume) reinforced with polypropylene.

changes in initial modulus with percent volume fraction of glass fiber are shown in Figure 3. An increase in the incorporated glass fiber content results in higher initial modulus. This is quite understandable because the
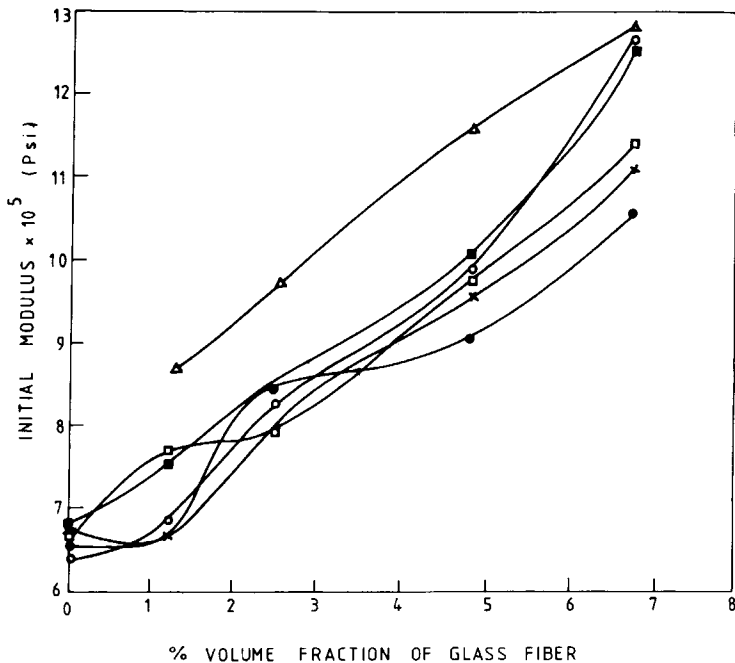

Fig. 3. Plots of initial modulus vs. % glass fiber content (by volume) in composite: (○) unirradiated; (X) irradiated to 1.6 Mrad; (●) irradiated to 3.2 Mrad; (□) irradiated to 4.8 Mrad; (■) irradiated to 8.0 Mrad; (△) irradiated to 14.4 rad.

composite modulus is contributed by glass fiber as well as by matrix, provided the volume fraction of glass fiber is above the critical amount.

Figure 4 represents the irradiation induced changes in the initial modulus for polypropylene and glass-fiber-reinforced polypropylene. The initial modulus of extruded polypropylene shows an increase of $\simeq 15\%$ over the original polypropylene. It appears that the polypropylene becomes more crystalline during extrusion, and so the initial modulus is enhanced. However, more work is necessary to ascertain the exact reason in this regard.

With the increase in total dose there is a slow but steady increase in the initial modulus of polypropylene. Polypropylene is a hydrocarbon polymer; it can undergo a chain-scission and/or hydrogen abstraction reaction to form free radicals.[5,15-19] The free radicals so produced can either destroy each other by mutual interaction, thus limiting the degree of polymerization at a lower value than the original (chain scission) or can form crosslinks by interacting with a free radical present in the adjacent polymeric chain. If the later phenomenon occurs, then the material becomes more brittle, and the initial modulus show an increase. It appears that such a mechanism is operating here.

With the incorporation of glass fiber in the matrix the initial modulus understandably shows an increase. When polypropylene and glass fiber mixture is exposed to irradiation, then irrespective of the glass-fiber content a further increase in the initial modulus is observed with dosage as shown in Figure 4. This behavior may be attributed to the two reasons, one the
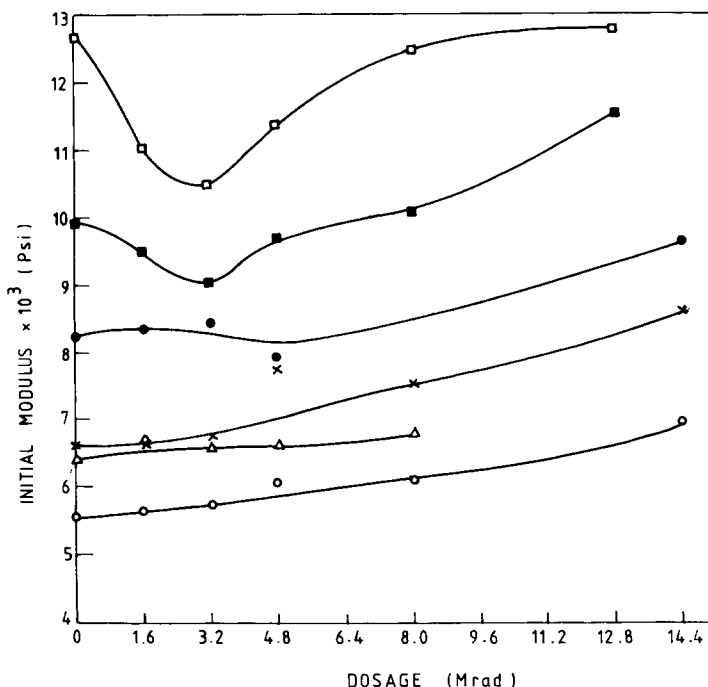

Fig. 4. Plots of initial modulus vs. dosage: ($\bigcirc$) plain polypropylene; ($\triangle$) extruded polypropylene; (X) polypropylene reinforced by 1.23% glass fiber; ($\bullet$) polypropylene reinforced by 2.45% glass fiber; ($\blacksquare$) polypropylene reinforced by 4.82% glass fiber; ($\square$) polypropylene reinforced by 6.75% glass fiber.

formation of crosslinks (which is evident from polypropylene curve in Fig. 4) and other an increase in interfacial bond strength between the glass fiber and polypropylene. The increase in the interfacial bond strength may be due to chain termination of free radicals formed on polypropylene by the abstraction of a suitable moiety from the glass fiber surface and thus bonding them together through a covalent bond. This view gets support from Figure 4, where the slopes of different curves show a mòre or less increasing trend with increasing glass fiber content.

When the glass fiber content is equal to or more than 2.45% by volume, in the initial stages there is a decrease in the initial modulus followed by an increase (Fig. 4). It appears that the glass fiber or the chemical entity present as coupling agent on the glass fiber is acting as a retarder to chain reformation reaction. As the total dose increases and surpasses a certain dose value (3.2–4.8 Mrad), the retarder gets exhausted and then the chain reforming reaction can take place without any hindrance. In the initial stages, the concentration of that chemical being very low, the effect is not noticeable, but with increasing the amount of glass fiber the effect becomes more evident.

The initial modulus value reaches a minimum at a total dose of 3.2 Mrad and then again increases as the total dose increases and ultimately surpasses the initial modulus value of unexposed sample when the total dose is more than 8.0 Mrad.

Effect of Irradiation on Tensile Strength

The tensile strength has been calculated on the basis of maximum stress withstood by specimen. Figures 5 and 6 present the effect of glass fiber
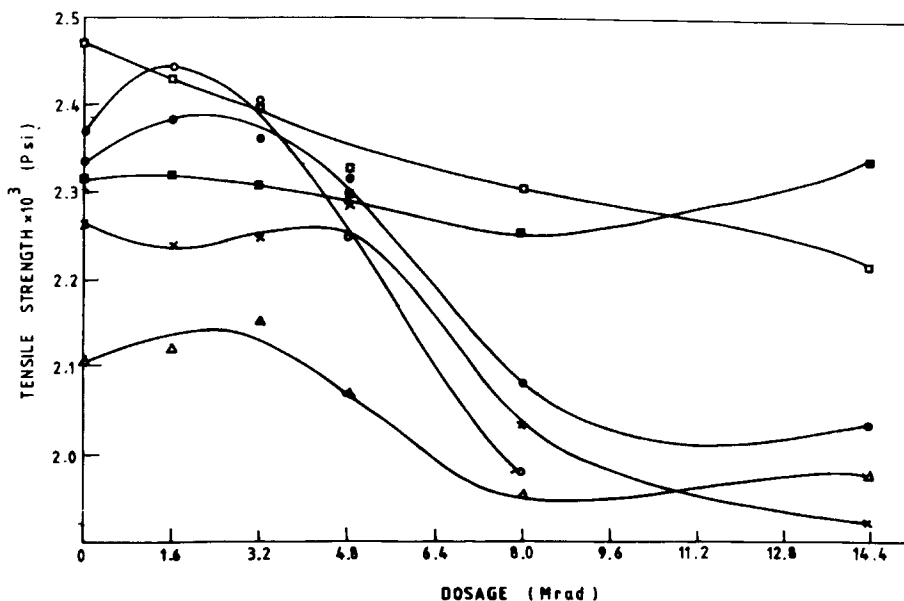

Fig. 5.   Plots of tensile strength vs. dosage: (△) plain polypropylene; (○) extruded polypropylene; (X) polypropylene reinforced by 1.23% glass fiber; (●) polypropylene reinforced by 2.45% glass fiber; (■) polypropylene reinforced by 4.82% glass fiber; (□) polypropylene reinforced by 6.75% glass fiber.
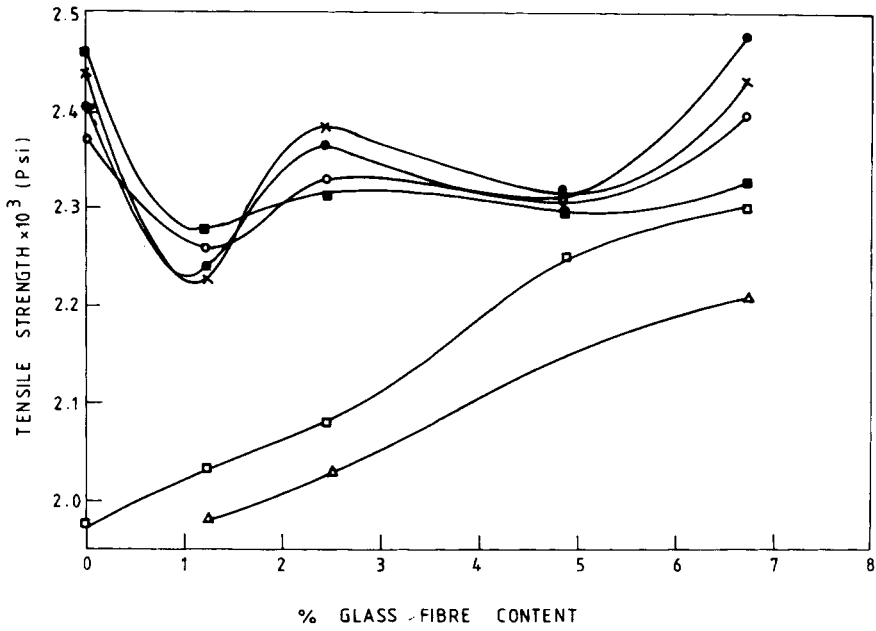
Fig. 6. Plots of tensile strength vs. % glass fibre content by volume: (●) unirradiated; (X) irradiated to 1.6 Mrad; (○) irradiated to 3.2 Mrad; (■) irradiated to 4.8 Mrad; (□) irradiated to 8.0 Mrad; (△) irradiated to 14.4 Mrad.
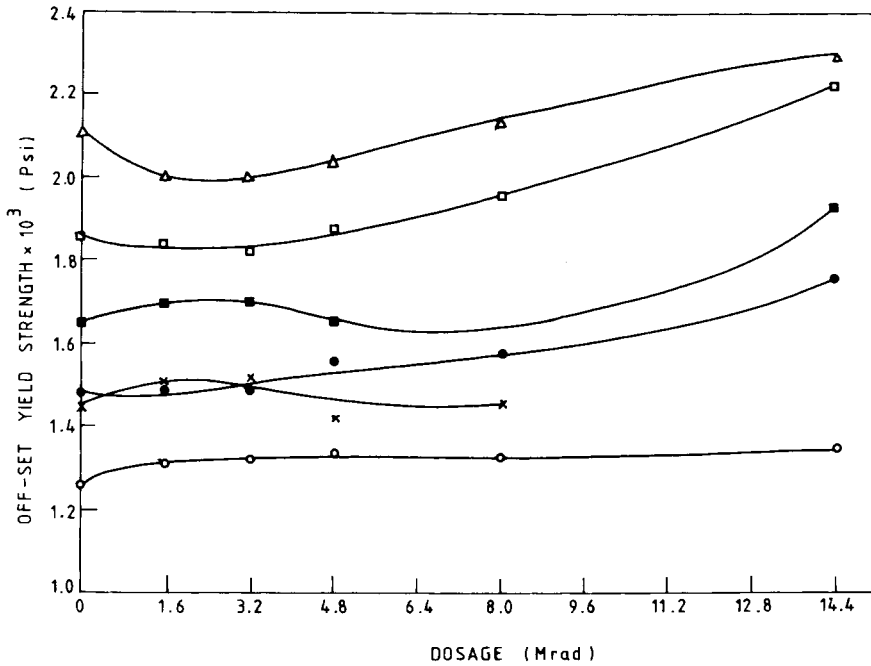

Fig. 7. Plots of off-set yield strength at 0.25% strain vs. dosage: (○) plain polypropylene; (X) extruded polypropylene; (●) polypropylene reinforced by 1.23% glass fiber; (■) polypropylene reinforced by 2.45% glass fiber; (□) polypropylene reinforced by 4.82% glass fiber; (△) polypropylene reinforced by 6.75% glass fiber.
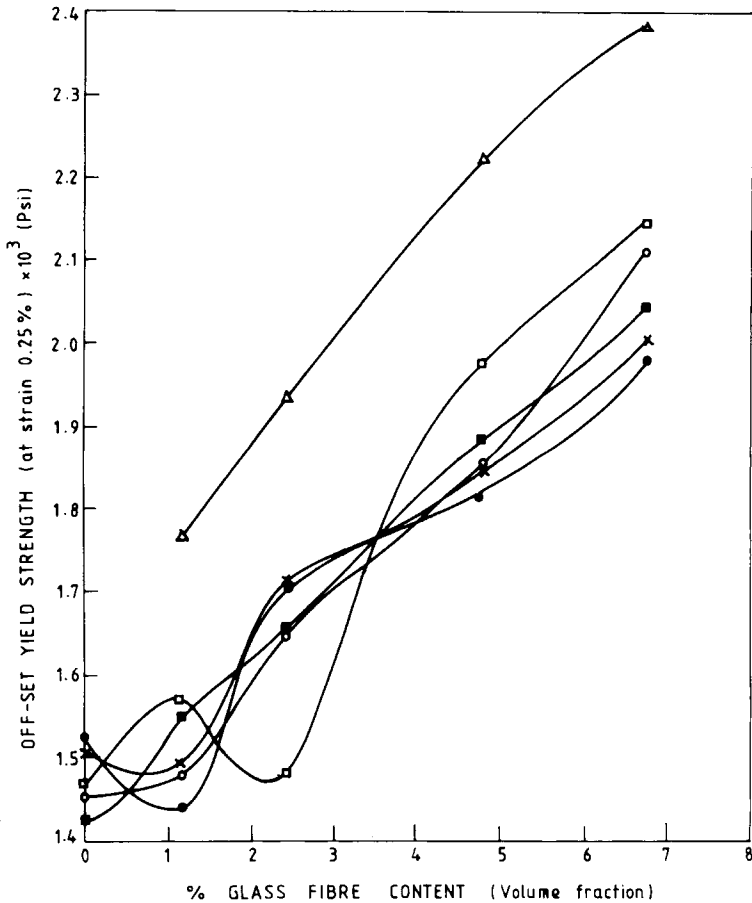
Fig. 8. Plots of off-set yield strength at 0.25% strain vs. % glass fiber content by volume: (○) unirradiated; (X) irradiated to 1.6 Mrad; (●) irradiated to 3.2 Mrad; (■) irradiated to 4.8 Mrad; (□) irradiated to 8.0 Mrad; (△) irradiated to 14.4 Mrad.

content and of radiation dosage, respectively, over composite tensile strength. A sharp reduction in tensile strength is observed for polypropylene and glass-fiber-reinforced polypropylene, containing 1.23% and 2.45% glass fiber samples on exposing them to gamma rays up to ≃9.6 Mrad, beyond which tensile strength becomes either constant or shows a slightly increasing trend. Almost same behavior has been reported by Hegazy et al. for polypropylene.[10] Noticeably, the reduction of tensile strength in glass fiber–polypropylene composites containing glass fiber 4.82% and 6.75% by volume is not very sharp. The probable causes have already been discussed in the previous section.

In irradiated samples as shown in Figure 6, the tensile strength initially goes down followed by an increase with increasing glass fiber content. This behavior is well known for a composite material, but, in the range of 2.45–4.82% glass fiber content, the glass-fiber-reinforced composite exhibits the behavior which is unlikely to be that of a standard composite. It can be attributed to sharp reduction in the average fiber length and broader length distribution of the fibers as discussed in the fiber length analysis section.

Effect on Off-Set Yield Strength

The variations of off-set yield strength (at 0.25% strain) with dosage and with % glass fiber content reinforced with polypropylene are shown in Figures 7 and 8, respectively. It is evident from Figure 7 that the off-set yield strength calculated at strain 0.25% linearly increases after a slight initial decrease. With irradiation also, off-set yield strength shows an increasing trend but for the sample irradiated to 8.0 Mrad a typical trend is observed at 2.45% glass fiber content by volume.

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