

# *Waste Materials Cured and Modified by Irradiating and their Use in Concrete*

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## **16.1 Introduction**

Transformation from a conventional consumption-based society to a sustainable society has great importance for several reasons such as lowering the pollution of the natural environment, prevention of exhaustion of natural resources and slowing down filling of final waste-disposal facilities. Recycling has the potential to reduce the amount of wastes disposed of in landfills and to preserve natural resources. Recycling, one of the strategies in minimizing waste offers three benefits: (1) reduces the demand for new resources; (2) cuts down on transport and production energy costs; (3) utilizes waste that would otherwise go into landfill sites. One novel alternative for recycling materials is gamma radiation, which has proved its efficacy for modification and improving of structural and physicochemical properties.

Concrete is one of the principal materials for structures and it is widely used for many applications all over the world. Thus, it is compelling to use life-cycle and sustainable-engineering approaches to concrete technology.<sup>1</sup> Concrete containing waste and recycled materials can support construction sustainability and contribute to the development of the civil engineering areas, minimizing the consumption of natural resources and producing more efficient materials.<sup>2</sup>

This chapter is divided into three sections, in the first one the modifications of waste materials by using gamma radiation are described; such materials are PET bottles, Tetra Pak packages and rubber tires. The second section describes the effects of these waste materials on the mechanical properties of concrete; and in the last section those effects provoked by gamma radiation in concrete containing these waste materials.

## 16.2 Waste Materials Cured and Modified by using Gamma Radiation

When using gamma radiation on polymers, three main processes occur: crosslinking, scission, and grafting of chains. The likelihood of each process occurring depends on the specific polymer properties. The gamma-rays have neither mass nor charge, and they are emitted from different sources,  $^{137}\text{Cs}$  or  $^{60}\text{Co}$  with 0.66 MeV and 1.33 MeV of energy, respectively.<sup>3</sup> The gamma-radiation exposure can provoke either lateral chain fracture and produce radicals combining with each other in order to bond adjacent molecules (crosslinking) or can break the main chain (scission), reducing the mean molecular weight (degradation).<sup>4</sup> Whether both of these processes occur in similar proportions or whether one or the other dominates is dependent on the chemical composition of the polymer. The application of gamma radiation for recycling polymers has increased its acceptance as a current technology due to the ecologic and economical features and mainly to the capacity of ionizing radiation to alter and improve the structure and properties of practically any polymeric material.

The Tetra Pak packages recycling is focused on their components: cellulose (75%), polyethylene (20%), and aluminum (5%); which are recovered in three different ways: (i) generation of energy through incineration, (ii) the recovery of aluminum in pyrolysis ovens, and (iii) the processing of the mixture of PE and aluminum to obtain high-end plastic lumber products.

With the exception of cotton, the main components of natural fibers are cellulose, hemicellulose, and lignin.<sup>5</sup> Linear cellulose molecules are linked laterally by hydrogen bonds to form linear bundles, giving rise to a crystalline structure. The degree of crystallinity is one of the most important structural parameters in cellulose; because physical and mechanical properties depend on it. The crystalline regions are interrupted every 60 nm with noncrystalline amorphous regions; such crystalline regions may contain occasional kinks or folds in the polymer chain, called “defects”. When

increasing the crystalline/amorphous regions ratio the rigidity of cellulose fibers increases, but flexibility decreases.<sup>6,7</sup>

Gamma radiation causes break-up of cellulose to shorter chains, which are water soluble, and it most likely leads to an “opening of additional microcracks”, in which water molecules can easily penetrate. The softening temperature ( $T_s$ ), is 235 °C for untreated cellulose, which decreases linearly up to doses of about 300 kGy, where any changes in the degree of crystallinity are detected, but degree of polymerization value decreases according to the dose increase. Therefore, it is assumed that the  $T_s$  shift to lower temperature range would be due to the scission of chemical bonds such as a glucosidic bond. On the other hand, the  $T_s$  shift in the range exceeding 300 kGy would be due to degradation of crystalline structure.<sup>8</sup>

At 500 kGy a higher solubility is obtained due to depolymerization and destruction of hemicellulose. The side-chain constituents of hemicelluloses (galactans) are affected sooner and to a greater extent than xylans or mannans that represent the primary backbone of the hemicelluloses. Degradation of cellulose or lingocellulosic material produces huge numbers of water-soluble or insoluble oxygenated compounds. Although the water content of aqueous phases is high, it contains some valuable chemicals. One of them is phenolic compounds.

The two highest-volume polymers HDPE and LDPE are classified as materials that primarily undergo crosslinking when irradiated under an inert atmosphere; however, they are very susceptible to radiation-induced oxidative degradation (chain scission) at relatively low doses in the presence of air. The melting temperature of neat HDPE is 138.2 °C, with a crystallinity of 49.1%. No significant variations of these values are observed after irradiation, except for samples having received the highest irradiation dose (100 kGy), for which the crystallinity decreased to 40% due to crosslinking. PE is known to suffer from degradation and loss of mechanical properties after gamma irradiation that is induced crosslinking leads to a strong decrease of elongation at break and impact strength values, counter balanced by an increase of Young's modulus and yield stress values.

Many efforts for recovering recycled-LDPE samples through the improvement of properties have been made. The recycled PE required a higher dose than virgin PE to achieve crosslinking. Moreover, exploratory research in which the radiation-induced breakdowns of PE scrap was carried out to generate lower molecular weight micropowders.<sup>9</sup>

In the case of PET some authors report changes due to the chain scission process at low dose (up to 10 kGy), while others note them at high dose (above 120 kGy). PET irradiated at 25 and 50 kGy shows a small but meaningful difference in ester ethyl of terephthalic acid generation, less than 1 mg kg<sup>-1</sup> for nonirradiated PET samples and 2 mg kg<sup>-1</sup> after irradiating at 50 kGy; measurable quantities of low molecular mass are formed and they resist gamma irradiation. Irradiated PET (from 100 to 300 kGy) was studied; at 100 kGy it shows a low molecular weight and an increment in crystallinity, which influence the aliphatic chain longitude and the aromatic ring (which

increases the resistance to the harm by radiation); these phenomena are attributed to the breakage of polymer chains. Moreover, the estimated average size of defects in the crystalline zones and crosslinking are detected, as well as improvement on the stability after irradiating.

For gamma-irradiated PET at doses higher than 135 kGy the bands at  $871\text{ cm}^{-1}$  and  $1303\text{ cm}^{-1}$  disappear and the absorption increases in the 320–370 nm range, which is attributed to the free radicals produced in PET during the process, which react with air oxygen to form carbonyl and hydroxyl groups. In another study, fair stability in the physicochemical properties at high doses (900 kGy), with changes from crosslinking processes up to 35% from the starting values were seen.

In early studies, recycled PET was compared to virgin PET when irradiating at different doses. It was found that random scission reactions are produced mostly in the main chain, with the consequent decrease in the polymer molecular mass; this was proved by means of fluidity and intrinsic viscosity level tests, performing by DSC. Gamma irradiation effects on thermal properties and the dyeing capacity of virgin and waste PET mixtures show an increase in thermal stability. In addition, color intensity was studied, for a mixture with 80/20% of virgin and recycled PET, the color intensity was improved 53 and 98% after submitting at 30 and 50 kGy dose, respectively.

A broad range of possibilities should be pursued using gamma radiation and recovered scrap rubber in an effort to create useful structural engineering materials. Some studies include ground tire rubber (GTR) and its mix with other materials, where gamma irradiation can vulcanize such thermoplastic/elastomer blends and to evaluate the possibility of recycling GTR and to obtain a product with specific properties.

Some work on GTR mixed with different materials has been conducted, including: (a) Recycling end-of-life GTR powder as functional filler in a thermoplastic matrix that offers an opportunity to design second-generation materials that would be recyclable; (b) GTR and nonrecycled resins, to avoid phase separation and lead to improvement of mechanical properties; (c) GTR and HDPE, allowing compatibilization of both recycled materials and improvement of their mechanical properties. Elongation at break and Charpy impact strength of the blends are significantly increased for irradiation doses ranging from 25 to 50 kGy, thanks to the involvement of GTR dispersed phase in material deformation. The moduli of elasticity of the blends are only slightly decreased by this compatibilization due to the fact that in the same time, the radiation induced crosslinking of the HDPE matrix. Nevertheless, for higher irradiation doses (100 kGy) such crosslinking leads to lower mechanical values.

In a GTR/PE blend gamma-radiation-induced chain scissions within the rubber phase will reduce the incompatibility with the thermoplastic matrix; and provokes crosslinking of polyethylene when using an inert atmosphere, nevertheless, in the presence of air oxidation occurs, leading to lower crosslinking. In general, for rubbers, chain scission dominates and it is enhanced under an air atmosphere.

Blend compatibilization between GTR and HDPE was confirmed by SEM, observations performed during a microtensile test. For a strain of 13% along the vertical axis, a lack of cohesion is observed below the GTR particles, while for 17%, the GTR particle does not participate in blend deformation. After irradiating HDPE/GTR blends at 25 kGy the elongation of the GTR particles increases.

### 16.3 Recycled and Waste Materials used in Concrete

Separation of the paper, aluminum and polyethylene used in packaging cartons is based on plasma technology. The heat transforms the plastic into paraffin and the aluminum is recovered in its pure form.<sup>10</sup> During the separation the first devolatilization takes place between 200 and 400 °C (maximum peak=360 °C), it is associated with the decomposition of the cardboard layer, including degradation of lignocellulosic material (up to 95% of total degradation). The second devolatilization occurs between 400 and 510 °C (maximum peak=475 °C) that corresponds to decomposition of polyethylene. Above 700 °C light paraffins and olefins are obtained.

Degradation of cellulose and PE are independent during pyrolysis, the presence of carbonyl group in wax may be related to both interaction between degradation products from cellulose and PE and the presence of some tarry impurities from cellulose degradation. The pyrolysis of PE at moderate temperature gives the mainly waxy product, yields of oil (C4–C15 hydrocarbons) and gases (C1–C4 hydrocarbons) are low. Moreover, the aluminum foil has no effect on the thermal degradation of both PE and cellulose.

Another polymeric waste material is polyethylene terephthalate (PET) from bottles (Figure 16.1), which represents one of the most common plastics in solid urban waste.

Some investigations already confirmed the potential of PET waste in replacing aggregates in concrete (Figure 16.2), which represents a better option than landfill.<sup>11–13</sup> Concrete with PET has a compressive strength up to 28 MPa,<sup>14</sup> increases its thermal performance up to 18%,<sup>15</sup> the unit weight decreases and flow ability increases, which means a better workability. The use of lightweight concrete (desirable in earthquake-prone areas) with a lower density can result in significant benefits such as superior load-bearing capacity of elements, smaller cross sections and reduced foundation sizes.<sup>16</sup>

Fatigue failure is a common problem of asphaltic concrete that can lead to pavement damage. The use of low concentration of PET in stone mastic mixture increases the stiffness modulus of the mixture; conversely, high concentration diminishes stiffness. PET-reinforced mixtures exhibit significantly higher fatigue lives compared to the mixtures without PET.<sup>17</sup>

PET can be returned to fibers that are obtained from melted PET waste to form a roll-type sheet. Then, the sheet is cut into 50 mm long fibers and a deforming machine is used to change the fiber surface geometry. When

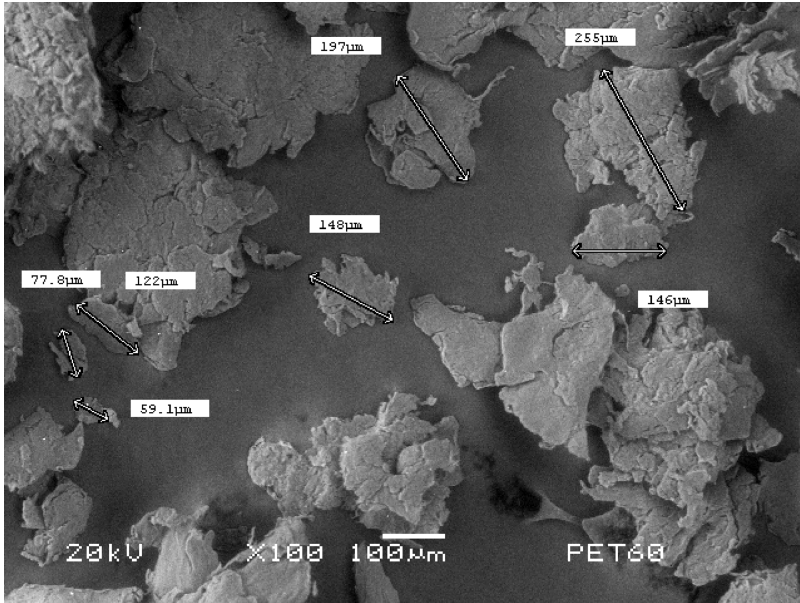


Figure 16.1 PET wastes after milling.

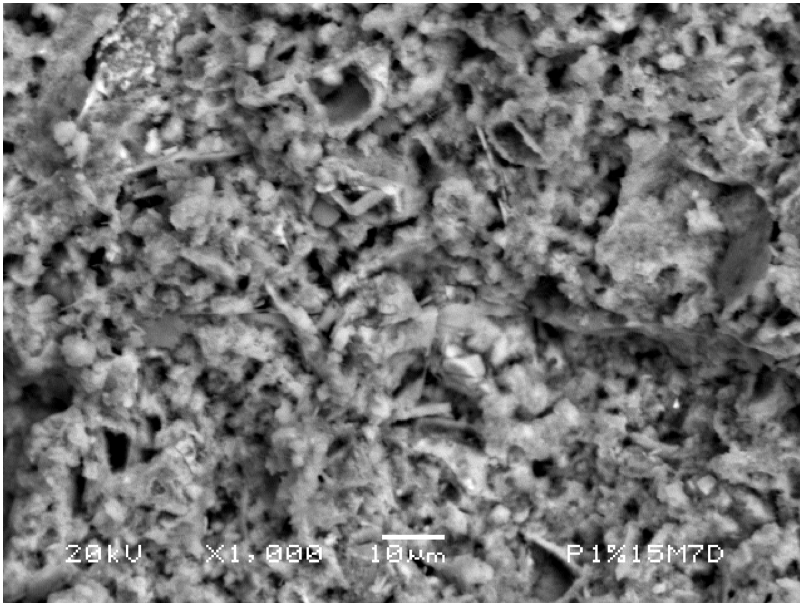
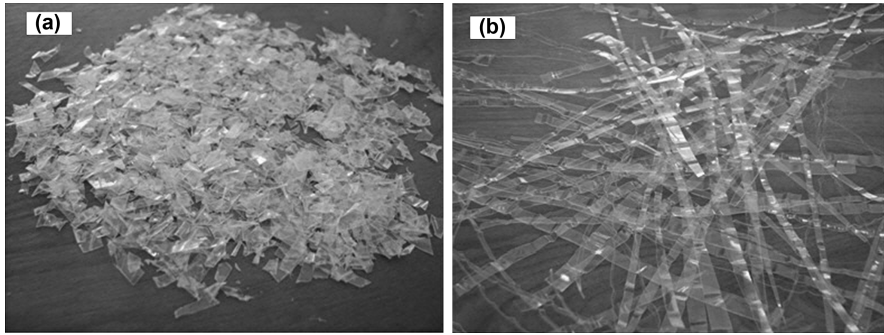
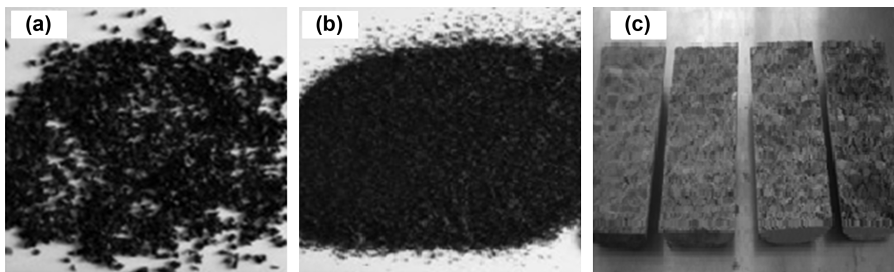


Figure 16.2 Granules of PET used as aggregate.

using PET-based fibers in concrete a better control on the plastic shrinkage is done. Moreover, depending on the surface treatment of PET fibers, these show a better performance of dispersion and bonding (Figure 16.3).<sup>18</sup>



**Figure 16.3** Recycled PET: (a) straight type, and (b) crimped type.



**Figure 16.4** (a) crumb rubber aggregate, (b) ground tire rubber aggregate, and (c) mixed into concrete.

The environmental damage caused by improper management of waste tires increased over the past years creating a relevant problem to be solved. In the field of civil engineering results show that it is possible to reutilize the rubber of the waste tires. The main markets for recycling currently are energy recovery (as kiln fuel in the cement industry) and raw materials recovery. Mechanical and chemical processes, such as tire shredding, pyrolysis and cryogenic reduction are used by the tire-recycling industry.<sup>19</sup> The current applications of recycling waste tires in civil engineering practices mainly are as follows: (a) Modifiers to asphalt paving mixtures, (b) as an additive to Portland cement concrete, (c) Light-weight fillers, and (d) in whole tires as crash barriers, bumpers, and artificial reefs.

Waste tire rubber is produced and classified according its size as follows:<sup>20</sup> (a) Fiber rubber aggregate (8.5–21.5 mm); Shredded or chipped rubber aggregate (>4.75 mm); (b) Crumb rubber (0.425–4.75 mm); (c) Ground tire rubber aggregate (0.425 mm), (Figure 16.4).

Some studies have been conducted on tire-modified concrete and mortars. Results have indicated that rubberized concrete mixtures show lower density, increased toughness and ductility, higher impact resistance, enough compressive and splitting tensile strength, more efficient sound insulation and increased thermal performance.<sup>21</sup>

Self-compacting concrete has a high potential for replacement of natural sand with tire rubber, and can be used in quantities of up to  $180 \text{ kg m}^{-3}$  of 64 mm crumb rubber aggregate replacement while providing acceptable fresh and hardened properties.<sup>20</sup> Resistance to chloride-ion penetration decreases with increasing waste tire content in concrete containing rubber aggregate. Chloride-ion penetration in concrete mixes after 28 day curing increases approximately 57% for a replacement ratio of the overall volume of aggregate of 25%.<sup>22</sup> Concrete containing rubber waste up to 15% has a high resistance to sulfuric acid attack.<sup>23</sup> The performance of concrete produced with waste tire aggregate is better than that of ordinary concrete under freeze–thaw cycles. High-strength concrete (HSC) shows a higher tendency of explosive spalling when subjected to rapid heating, as in the case of fire. This behavior is mainly due to the lower permeability of cement paste. HSC filled with solid fiber-shaped particles of recycled tire rubber reduce both the risk of HSC explosive spalling and its stiffness without a high loss of strength, enlarging the compatibility of deformation with other building elements.<sup>24</sup>

Bituminous materials have been used in buildings, construction, tunnels, on bridges, for hydraulic structures, and other purposes in civil engineering. The best known and important role of bituminous materials is in road construction and road safety. The accelerated deterioration and eventual failure of bituminous pavements has been partially solved by the addition of tire rubber to an asphalt concrete binder.<sup>25</sup> Waste tires provide a good mechanical behavior under static and dynamic actions. Also, recycled tire rubber-filled concrete can be used as a rigid pavement for roads on elastic subgrade when fatigue loads are applied.<sup>26</sup> A good compatibility and interaction between rubber particles and asphalt binder is achieved, leading to various improved properties of asphalt mixtures. Advantages include high resistance to temperature and to freeze–thaw cycles; better drainage; and diminution of cracking due to the elastic properties of rubber.<sup>27,28</sup>

## 16.4 Irradiated Concrete Containing Waste and Recycled Materials

The use of gamma radiation as a mechanism for reaction initiation and as an accelerator of the polymerization of a monomer in a ceramic matrix can bring considerable advantages to the process. Some studies cover the effects of ionizing radiation on polymer–ceramic composite materials, where the yield of polymerization increased with increasing radiation dose.<sup>29</sup> As this process is completed at room temperature there is much economy of heat energy, besides the reduction in costs to keep the system under pressure. The pressure is used to keep much of the monomer (usually with a high volatility) filling the interstices of the ceramic matrix during the conversion to ceramic–polymer composites.

Virgin materials are modifiers that can be used to improve the properties of road surfaces, nevertheless they are uneconomical and difficult to find.

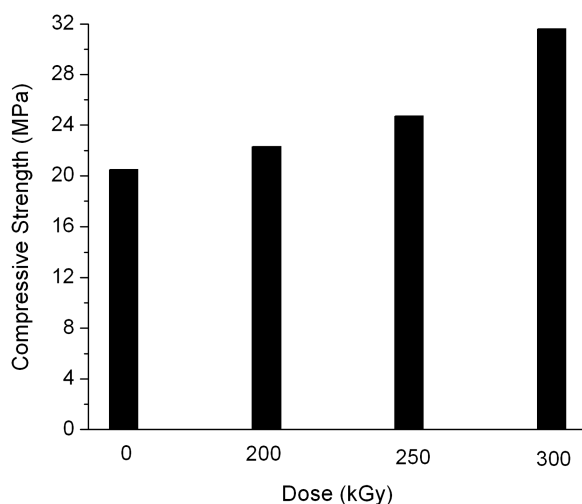


In the last decade many studies have focused on using waste polymers as a modifier in road surfaces; they can potentially help reduce material wastage and improve the performance of road surfaces at the same time.

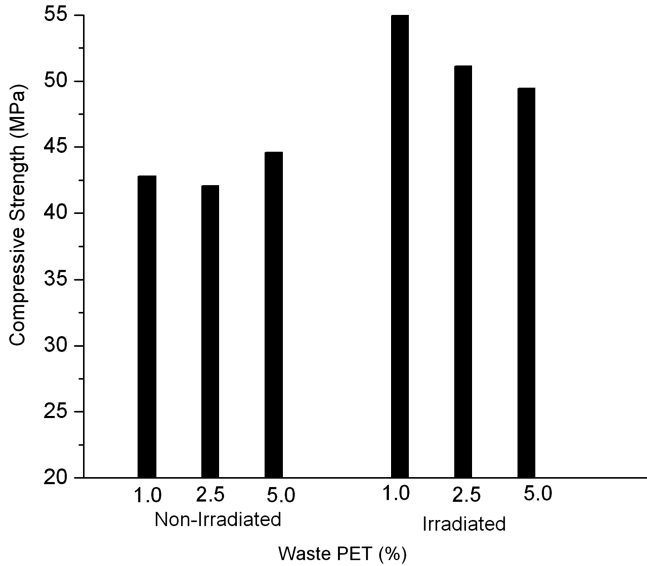
For concrete containing recycled cellulose (from packaging cartons) the compressive strength values gradually decrease when more concentration of cellulose is added; the strength depends on the amount of waste cellulose and water cement ratio (w/c). When irradiating concrete the highest compressive strength value is observed at 300 kGy at 28 days of curing time, which means an improvement of 45% respect to control concrete (Figure 16.5); moreover the values increase when the radiation dose increases too. As is known, many types of chemical reactions take place during gamma irradiation of polymeric materials, crosslinking and degradation by chain scission among others, but one or the other of these effects may be predominant in some materials.

Application of high-energy irradiation to cellulose creates free radicals by the scission of the weakest bonds; such radicals can react with some molecules in the cement matrix. The interaction between calcium silicate hydrate (formed during the hydration process) and the cellulose present in the pores during irradiation polymerization that enhances the interphase bonding, and as a result, an improvement of the mechanical strength takes place.

Improvements in modulus of elasticity values of concrete point out a predominant domain of crosslinking of polymer chains in cellulose. However, some shorter chains are produced which are water soluble and in consequence an increment in the solubility is obtained. In general terms, irradiated cellulose cover the sand particles, thus the zone around them is affected by a stress concentration. Therefore, if the distance between



**Figure 16.5** Compressive strength of concrete with waste cellulose at different irradiation doses.



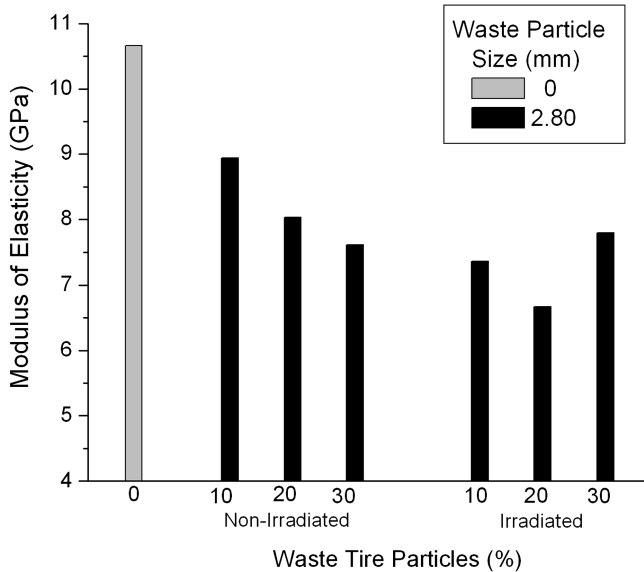
**Figure 16.6** Compressive strength of concrete at different PET volume concentration.

particles is small enough, these zones join together and form a percolation network, which generates good adhesion between the cement matrix and the cellulose and in consequence an increment in the modulus of elasticity is obtained.<sup>8</sup>

The compressive strength of concrete containing waste PET is shown in Figure 16.6. For nonirradiated concrete the values increase when PET concentration increases from 1.0 to 5.0 vol%. The highest value is 44 MPa for concrete with 5.0 vol% of PET. For irradiated concrete the values are up to 35% higher when compared to those of nonirradiated concrete, nevertheless they decrease progressively when increasing the PET concentration.

Compressive strain values of concrete containing waste tire particles decrease progressively according to the particle concentrations increase. Concrete with 10% of particles of 2.8 mm in size is 11% lower than those for control concrete (24 MPa). According to the particle size, the compressive strength values are higher for concretes with particles of 2.8 mm size than those with a size of 0.85 mm. For concrete with waste irradiated tire particles the compressive strength values decrease when increasing the particle concentrations. It is more convenient using bigger size particles instead of smaller ones.

The modulus of elasticity values of concrete are shown in Figure 16.7. They decrease on increasing the concentration of particles; also, concrete with particles of 2.8 mm size have higher modulus of elasticity values when compared to those with a size of 0.85 mm. For concrete with irradiated particles, moduli of elasticity values are higher than those for concrete with nonirradiated particles. Two “stages” are identified in concrete with



**Figure 16.7** Modulus of elasticity of concrete with waste-tire particles.

irradiated particles, the first one consists in a diminution of the values for concretes with 10% and 20% of particles, and the second increment for concrete with 30% of particles.

## 16.5 Conclusions

As is known, environmental problems caused by waste materials are in constant growth and as consequence different methods have been development, nevertheless some are consuming money and time. One novel alternative consists in using ionizing radiation to modify the physicochemical properties of the wastes and after using them as fillers or modifiers in composite materials as concrete. In this chapter, we have shown that some properties can be improved when using gamma radiation. As concrete's compressive strength is one of the most important structural design parameters used by engineers, waste PET particles can be a suitable material for construction. A small amount of PET could be used for substituting fine aggregate in the mix design to increase strength, and diminish strain. Thus, irradiation can become a useful tool and a suitable method for recycling waste PET. Moreover, in other cases both waste cellulose concentration and gamma radiation are adequate tools for improvement of the mechanical properties of concrete, where sand is substituted by waste cellulose. In general, compressive strength and modulus of elasticity values show an improvement when adding waste cellulose and applying certain gamma dose. Conversely, diminutions on the mechanical properties are seen for nonirradiated concrete.

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