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Multicomponent Polymeric Engineering Materials

Manuel Panar and Bennett N. Epstein

It is only within the past century that synthetic plastic materials have been known at all, and it is only within the past several decades that they have had a significant level of application. For most of this period they found nonstructural uses, where their strength and mechanical durability were not fully tested. However, since the discovery, about 1940, of synthetic fibers which were stronger and more versatile than natural ones, structural applications of polymers have increased and engineering plastics of great strength and durability have replaced metals in many applications.

In spite of this shift in usage, the science needed to optimize application of these materials did not exist until very recently. Applications were, in fact, inhibited by the substitution of resin for metal without designing for the physical properties of the plastic, which led to many ill-advised applications and resulted in a stereotyped image of plastics as having poor quality. The problem was aggravated by the use of some plastics to do jobs for which they were unsuited. Within the past 25 years, recognition of the uniqueness of synthetic plastics has gradually advanced. The earlier materials have given way to highly engineered multicomponent systems with high levels of strength and durability, and a body of data has been accumulated which permits the design of structures based on the special characteristics of polymers.

Compared to metals, plastics have advantages in resistance to corrosion, ease of fabrication, adaptability to single-part fabrication of multifunction components, and lower energy content. Even though synthetic resins are produced from hydrocarbon feedstocks, the energy equivalent of the hydrocarbons which end up as polymer is far less than that consumed as fuel to produce metals. In addition, the amount of fuel used in producing polymer is low (*1*). As a result, the energy expended in producing a given volume of structural material is significantly less for plastics than for metals. Synthetic resins are less expensive on a volume basis than most metals, and this differential is expected to increase through this decade (*1*). The rapidly increasing strength of synthetic systems is overcoming this earlier advantage of metals; many synthetics already have strengths per unit volume (specific strengths) equivalent to or greater than those of metals.

As the use of polymers has become more sophisticated, polymers themselves have evolved rapidly. During the past few decades they have come to be treated as starting materials for the new multicomponent systems which are rapidly replacing metals in many applications, and the science is reaching the point where specific compositions can be designed for specific applications.

In this article we discuss (i) the applications of polymeric structural materials, (ii) the available classes of plastic resins and how they are modified for improved strength, (iii) concepts of molecular orientation and the high-strength polymeric fibers based on them, and (iv) the combination of fiber and resin to form new high-performance composites, and the mechanical properties of these systems. Finally, we review new research directions and discuss missing science, with particular reference to areas of opportunity for basic research.

Applications

Polymeric materials are rapidly moving into roles which were exclusively filled by metals only a few years ago. Nylon chain sprockets for bicycle and

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motorcycle chains would have been inconceivable prior to the recent revolutionary advances in multicomponent systems, yet are now in commercial use. Polymers have long replaced metals for instrument chassis and bodies of portable tools, and current uses involve larger scale and, more important, critical load-bearing applications. In the transportation industry the changes which will be distinguishing features in the 1990's can be seen not only in research and development laboratories but also in strikingly new products.

The most dramatic new area of use is in primary—that is, critical load-bearing—aerospace applications. In recently announced commercial aircraft, polymeric materials constitute a significant fraction of the total weight (*2*). These materials were initially used as interior panels and frames to save weight, but are now being used as ailerons, flap fairings, rudders, and so on. One manufacturer has announced an all-composite aircraft in which the reduction in weight results in a 2.5-fold improvement in fuel usage and a 60 percent increase in range over that of a comparable aluminum airplane (*3*). One estimate of the assembly advantages of a composite airplane includes a 5-fold decrease in the number of airframe parts and a 12-fold decrease in the number of fasteners used in assembly. Rocket motors and the NASA space shuttle are made considerably lighter through the use of polymeric materials. These examples of the high end of the use range illustrate the currently realistic potential. Many applications which are more cost-sensitive will benefit as newer processes bring the price for the products down.

Summary. The great advances made in polymeric structural materials over the past decade have led to their replacement of conventional materials in a wide range of uses including high-performance aerospace applications. This shift in choice of materials is based on economic advantages, simplified fabrication, freedom from corrosion, and lower weight. The trend toward use of polymeric materials will grow as the materials science of this new technology is developed. Better insight into such fundamental problems as the mechanisms by which forces are transferred to the polymer molecule, the surface interactions between polymer phases or polymer and fiber, and the molecular processes by which energy is absorbed during fracture will greatly stimulate the development of this field.

Resins

Thermoplastic resins are ones in which the individual molecular chains are free to move independently. As a result, they can be melted and thus be fabricated in a molten or softened state and be reused. Both amorphous and semicrystalline thermoplastics are used in engineering applications. The five most commonly used chemical types are nylon; acetal and polyester resins, which are semicrystalline; and polycarbonate and blends of polystyrene with polyphenylene oxide (PPO), which are amorphous. The "engineering plastics" have flexural moduli over 2 GPa [300,000 pounds per square inch (psi)], tensile strength about 70 MPa (10,000 psi), and elongation to failure over 25 percent. Lower volume, more expensive resins with greater resistance to chemical or thermal degradation are available. These polymers include fluoropolymers, polyphenylene sulfides, polysulfones, polyimides, and polyether ketones.

Semicrystalline plastics have greater resistance to solvents and to fatigue than the amorphous materials. The amorphous materials shrink less in molding, which simplifies mold design. Unmodified engineering plastics have useful properties and will continue to play a role, but they are increasingly being used as base resins in more sophisticated high-performance multicomponent systems.

In cross-linked resins covalent bonds are formed between the individual molecules, usually by a thermal treatment following initial fabrication. In these thermoset resins the cross-links greatly inhibit the mobility of the polymer network. The resulting polymers are infusible and stiff but brittle. Flexural moduli are about 500,000 psi and elongation the order of 1 percent. Applications are primarily restricted to reinforced structures. Thermosets are heavily used for the high-performance composite structures discussed below. Although the volume of reinforced thermosets produced is much greater than that of reinforced thermoplastics, the latter are more versatile and more easily fabricated, and the growth rate is higher.

Toughening (Blends)

Blends of two or more polymers are becoming of increasing importance among plastics which are considered to be of engineering or structural quality (*4*). Polymer pairs that are miscible on a molecular scale are comparatively rare since the entropy of mixing is low because of connectivity, which sharply reduces the number of possible positions available to each atom in the polymer. In some systems, such as polyester-polycarbonate blends, the enthalpy of mixing is sufficiently favorable that a large contribution from the entropy term is not required to achieve miscibility. Of the five major chemical types used in plastics, the polystyrene-PPO system is the only homogeneous blend.

Heterogeneous systems have the advantage that properties of the blends can be designed to take advantage of the desirable property of the continuous phase without the averaging effect which frequently occurs in homogeneous systems. For example, the transition temperatures of the continuous phase, both melting point and glass transition temperature, tend to be comparatively unaffected in heterogeneous blend systems.

Rubber-toughened amorphous polymers are among the more useful and interesting of these systems. Toughened polystyrene and acrylonitrile-butadiene-styrene were two of the first such materials. Rubber-toughened amorphous polymers are generally thought to absorb energy by craze formation, in which voids are opened at some distance from an advancing crack. The void formation is accompanied by stretching of ligaments of the parent material across the void. This deformation of material is the energy-absorbing mechanism. At modest levels of toughness enhancement, with perhaps three- to fourfold improvement in energy absorption, it has been calculated that the energy absorption occurs in the rubber particles themselves (*5*). Because of the limitations of styrene-based materials in terms of temperature and solvent resistance, and the comparatively poor combination of mechanical properties, the rubber-toughened polystyrene materials have not generally been used in engineering applications to replace metals. It was therefore an important objective to toughen the semicrystalline engineering plastics, which have a good combination of mechanical properties and solvent resistance.

Heterogeneous phase toughening of semicrystalline plastics to provide true ductility with good retention of stiffness was achieved in the latter half of the 1970's (*6*). The behavior of these materials has been described and partly elucidated (*7, 8*). It has been suggested that the heterogeneous inclusions in nylon cause localized shear yielding of the matrix. Modeling of the system depends on the assumption that interacting shear stress fields between neighboring particles are required to achieve the stress levels necessary to promote shear yielding at some distance from the advancing crack. Inherent in the analysis is the assumption that energy absorption occurs primarily in the matrix and that the role of the second phase is to promote this energy absorption. The observation (*7*) that the fracture surface in highly toughened nylon appears to run around the rubber particles and in the matrix supports this assumption. A very recent development is the introduction of toughened polyacetal, in which interparticle spacing is also important in heterogeneous toughening (*9*).

Reinforcement

Reinforced structures can be classified by the ratio of length to diameter (aspect ratio) of the reinforcing agent. Materials with a low aspect ratio such as clay, talc, and mica confer increased stiffness, generally by about a factor of 2, but not increased strength. Glass fibers used for reinforcement have larger aspect ratios, in the range 20 to 100. Engineering resins with this kind of reinforcement have substantially increased tensile strength as well as stiffness when the strength of the bond between the reinforcement and the matrix approaches the shear strength of the matrix. These materials can be injection-molded very conveniently. The combination of good mechanical properties and easy fabrication has led to broad industrial application. The strength of material reinforced with short-fiber agents is generally in the range 140 to 200 MPa (20,000 to 30,000 psi) with modulus 7 to 14 GPa (1 million to 2 million psi).

Molecular Orientation
(High-Strength Fibers)

Understanding the development of high strength and rigidity in polymeric materials requires some molecular concepts which are not found in other branches of materials science. Thermoplastic polymers are long, essentially linear molecules that, if fully extended, would have dimensions of about 0.6 nm by 100 to 10,000 nm. A molecule contains 200 to 20,000 covalent bonds along the chain. Since segments of the molecule can rotate about each of these bonds, it is only in very rare cases that they remain largely extended (*10*). However, the degree to which the chains are extended and the degree to which they are oriented in a single direction, extended or not, have a major influence on the physical properties of the polymer.

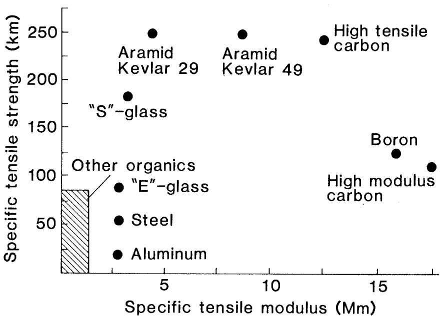

Fig. 1. Specific tensile strength and specific tensile modulus of reinforcing fibers (tensile strength or modulus divided by density).

Orientation and elongation are important because the major element in the ultimate strength of a polymer is the strength of the molecular chain itself. A hypothetical polymer in which all chains were oriented and elongated so that all carried the load would have a tensile strength three to seven times that of the strongest available polymeric fibers. However, attainable moduli, >200 GPa, may be much closer to the theoretical maximum, within a factor of 2 or 3. Although theoretical estimates are difficult to make and are controversial, polymeric materials are within an order of magnitude of the ultimate, and judging by recent advances in the field, a close approach to the ultimate goal appears achievable.

Much of the technology, present and foreseen, for improved high-performance polymeric systems hinges on ingenious routes to manipulating bulk polymers in a way that influences the molecular orientation. The bulk polymer can be manipulated by extension of the solid or by exerting shear and elongational forces on a melt or solution. Although the conditions under which the forces applied to the bulk will be transmitted effectively to the individual molecules are only now being elucidated, there are many examples of the phenomenon. One of the best examples is the conversion of polyethylene, which is normally not a high-strength material, to a high-modulus form (*11*). Slow, repeated drawing under controlled conditions converts chains that are normally random coils to highly extended rods, increasing the polymer stiffness manyfold.

Spinning of moderately concentrated solutions (gels) is another route to highly oriented polymers (*12*). This method has also produced high-strength polyethylene fibers (*13*).

A special case of orientation in the fluid state takes advantage of the anisotropic solution and melt phases known as liquid crystals. In a polymeric anisotropic phase formed from rigid molecules which are relatively elongated, the chains are spontaneously oriented in a single direction over micrometer-size volumes. Shear and elongational forces can orient these larger volumes more readily than uncorrelated molecules. The most dramatic example of the results of such orientation is found in the ultrahigh-strength fibers formed by spinning anisotropic phases (*14*). The aramid fibers are the only examples in current wide usage. In these fibers the morphology is such that a very high proportion of the molecules are load-bearing (*15*). Similar properties have been reported for polybenzimidazoles and polybenzthiazoles (*16, 17*).

Fibers made by these methods have specific strengths far greater than that of steel (Fig. 1). Considering that polymer science is relatively new and developing rapidly, further substantial improvements can be expected.

Composites

As we discussed above, it is in the nature of polymer molecules that their ultimate strength is unidirectional, and the strongest form in which they can be fabricated is fibers. The high-performance composites which have been developed over the past decade take advantage of the extremely high strength of new classes of fibers which are used as the load-bearing elements, the matrix serving to maintain shape and to help in distributing the load uniformly to the fibers (*18*). The most important of these fibers are aramids and carbon. The mechanical properties of high-performance composites in the fiber direction are in the range of those of high-strength steel, with tensile strength over 700 MPa (100,000 psi) (at one-fifth the density of steel) and specific stiffness as much as seven times that of steel.

Most composites are formed currently by impregnating bundles of fibers or fabrics with a liquid thermosetting resin, that is, one which can be further polymerized (cured). Two processes account for the bulk of the high-performance materials. In one, prepregs, which are mats of fiber impregnated with the resin, are assembled into the desired form and then polymerized with heat and pressure. The second process is filament winding, in which the fiber is continuously coated with resin as it is wound into the final form. Both processes, which are relatively time-consuming, are being highly automated. These fabrication routes allow the designer to place the fibers so that the resulting part has maximum strength in the desired directions. Ultimate strength must be traded for the desired balance of properties.

Strength and Durability

Commercial development and understanding of the chemistry and morphology of polymer blends has outpaced the description of mechanical behavior of these materials. Detailed understanding of the science underlying mechanical behavior will lead to improved utilization through optimization of multicomponent materials and more sophisticated design.

Modern study of fracture mechanics began with the work of Griffith (*19*). His work rested on two basic assumptions: that fracture is initiated at flaws which are inherent in all materials, and that the energy per unit area required to propagate the flaw is a characteristic of a material, called the fracture strength. Griffith's work on glass was extended to steel by Irwin (*20, 21*) and Orowan (*22*). This work led to a standard test method (*23*) which is useful for rigid materials including reinforced plastics, but it is not applicable to very tough materials which have very large damage zones rather than small, brittle cracks. A method was formulated (*24*) and developed (*25, 26*) to deal with the circumstance. This approach shows promise for defining fracture strength or toughness of very tough resin systems (*27, 28*).

As more sophisticated multicomponent structures with improved properties come into use, durability is becoming a matter of increasing concern. One measure of durability is the rate at which a crack can be driven through a specimen under conditions of steady crack growth (*29*). This rate of failure is an order of magnitude lower for semicrystalline polymers than for amorphous polymers (Fig. 2) (*30*). The higher resistance to damage of the semicrystalline resins is not completely understood but is consistent with greater resilience. In most toughened systems, fatigue performance by this test is not improved, but toughened nylon is a useful exception (*31, 32*).

Modes of failure in reinforced materials are very complex and are under intensive study. Concepts of fracture mechanics are being extended to the high-performance composites. Failure may be influenced by residual stresses from fabrication induced by differential rates of shrinkage between fiber and matrix (*33*). It can occur in the reinforcing material, at the interface, or in the matrix.

As in the example of material behavior above, one sign of the growing sophistication of polymeric materials is that material selection is being increasingly based on parameters more technically complex than simple strength and stiffness. Another good example is the distinction between use properties of aramid-epoxy and carbon-epoxy composites designed for load-bearing structures in aircraft and other transportation applications (*34, 35*). Reflecting the properties of the fibers, the carbon composites have greater compressive strength, and might be expected to be most valuable in an application which requires retention of strength in, for example, a crash. However, the mechanism by which the aramid composite fails is such that it is far "tougher." There are many interpretations of toughness, a parameter which is not as self-evident as tenacity and rigidity. A common definition is the energy absorbed in failure (the area under the stress-strain curve). When normalized for density, an aramid composite absorbs two to three times as much energy as a carbon fiber composite during failure because of its greater elongation to break. A second definition of toughness, which is more intuitive and possibly more meaningful, although more difficult to quantify, is the ability of a material to continue to function in the presence of localized damage and the ability to sustain catastrophic failure in such a way that, for example, occupants of an airframe are protected during the failure. Aramid-containing composites fail much like metals in that the damaged part retains a significant amount of mechanical integrity. In contrast, carbon composites fail in a brittle fashion in which all structural integrity is lost at once on failure (*36*). This failure, however, takes place at a higher compressive stress, so that to take advantage of the failure mode of the aramid composite, the designer must compensate for the lower compressive strength.

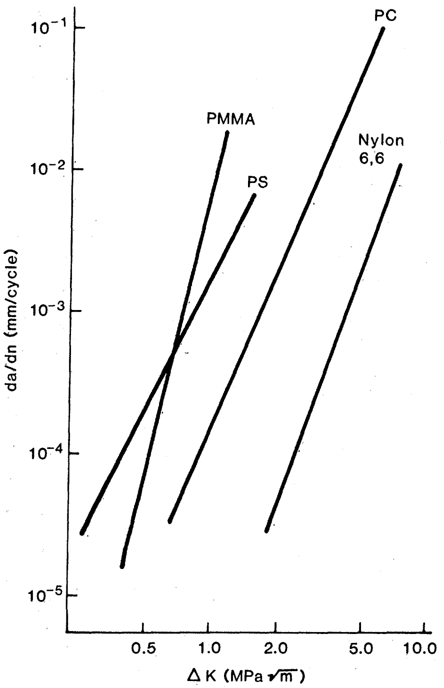

Fig. 2. Fatigue crack propagation behavior in selected crystalline and amorphous polymers; *da/dn* is a measure of the rate of crack propagation and ΔK is a measure of the applied stress. Data are from (*30*). Abbreviations: PMMA, polymethyl methacrylate; PS, polystyrene; and PC, polycarbonate.

The design of polymeric structures is increasingly based on hybrid composites in which two or more fibers are combined to take advantage of the best properties of each. For example, carbon and aramid fibers both confer strength. The carbon can be placed in the structure to maximize the overall stiffness, and the aramid fibers placed to take advantage of their fracture mode, which confers durability under repeated impact. The strength of hybrids can be greater than that which would result from the same total amount of either component (*18, 37*).

This trade-off of properties emphasizes a point made earlier: the maturing of polymeric materials has been based not only on the improvements in the materials themselves but also on the recognition that the term "plastics" represents not a single type of material but a broad class of materials with different but complementary properties. These materials must be used with an understanding of their strengths and weaknesses. This understanding has developed rapidly over the past decade, and their range of utility will expand at an increasing rate as the details of their functioning are elucidated. In the last section of this article we will cover some of the underlying scientific questions which must be answered so that this field can move forward as expeditiously as possible.

Future Materials

Current research and development activity is leading to new concepts that will further improve properties and simplify fabrication. One such concept involves continuous fiber composites formed from thermoplastic, rather than thermoset, matrix resins. Thermoplastic resins are capable of being thermally reshaped and are stable during storage. Composite shapes such as sheets, I-beams, and rods could be fabricated by the supplier and then be formed and joined into structures by the user. Advantages in durability are expected.

Another new concept receiving much attention involves the development of materials having properties similar to those of high-performance composites together with the economics of fabrication (for example, by injection molding) associated with short fiber-reinforced polymers. This would be done by taking advantage of the self-orienting characteristics of anisotropic melts and solutions (liquid crystals) discussed above. However, they would be used as self-reinforcing resins which, because of their high level of orientation, do not need the fiber component (*38–41*). Recently published work presents a promising route to the application of these systems (*42, 43*). Blends of anisotropic (liquid crystalline) polymers and more conventional resins are formed in which the anisotropic reinforcing component is dispersed either microscopically or molecularly.

The joining of polymeric structures is an active subject. Large structural parts must be assembled in such a way that localized high-stress areas are not formed. Structural adhesives have become standard parts of structures in recent years, and will be modified and optimized for use with polymeric systems. Welding of plastic parts will be rapidly extended to large structural shapes.

Missing Science

The mechanism by which shear and elongational forces are transferred to the polymer molecule and cause it to become extended and oriented has been examined. However, it is still poorly understood. More has been done to devise new ways to do the job than to explain the science underlying the process. Better insight into these phenomena would greatly stimulate the development of improved processes. Similarly, a molecular explanation of how mechanical forces applied to a solid polymer are transmitted to the individual chains to orient them would be of great value.

Many properties of blends and composites are assumed to be strongly influenced by surface interactions between polymer phases or between polymer and fiber (*44*). Much remains to be clarified about this interface. The effective thickness of the layer modified by surface forces has received considerable attention; however, the implications of the existence of this layer for mechanical properties are not understood. The effect of deliberately adding interphases (fiber coatings) to affect the micromechanics has received considerable attention (*44*). An improved understanding of these interphases would provide a powerful tool for modifying and optimizing adhesion and the sharing of stresses between the components of a composite.

Although the rheology of two-phase melts has been studied for years, many important questions remain. The orientation of short-fibers in a flowing viscous medium is not yet fully predictable in spite of much published work. Better control over this process would greatly extend the ability to form moldings with optimum orientation of reinforcing fibers.

The micromechanics of continuous fiber composite properties must be extended to the more ductile thermoplastic matrices. New understanding is needed to deal with the micromechanics of in situ reinforced blends in which the "reinforcing fiber" has dimensions approaching the molecular.

The molecular processes by which energy is absorbed during fracture are not yet understood, even though progress has been made in defining the morphological characteristics that confer toughness. Similarly, in spite of some correlations with relaxation phenomena (*45*) and phenomenological approaches (*46*), there is no molecular explanation of why some polymers are inherently more difficult to fracture. The need to thermally join thermoplastic materials raises fundamental questions about the diffusion of molecules at molten interfaces. This subject has been studied largely in the context of crack healing and is being extended to the fusion of preformed surfaces.

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