Crystalline organization and toughening: example of polyamide-12

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Abstract

Improving the impact resistance of plastics is a key to many applications. Today, dispersing rubber and inorganic particles into semicrystalline polymers is widely used to increase their impact strength without greatly altering other interesting properties such as elastic modulus or chemical resistance. Yet, the underlying mechanisms controlling such toughening are controversial. Hitherto it has been often suggested that a critical distance between particles which controls the brittle-to-tough transition is an intrinsic property of the polymer. On the contrary, we demonstrate here that differences in crystalline organization of the matrix can induce dramatic changes in toughening efficiency. A thermal treatment and microscopic observations strongly suggest that crystalline orientation, size of crystalline grains and molecular organization at grain boundaries play a determinant role in the toughening mechanisms. These observations may have important implications for designing and manufacturing tough plastic materials.

Keywords: Toughening; Polyamide; Semi-crystalline polymers

1. Introduction

The impact resistance of semicrystalline polymers such as polyolefins or polyamides, is dramatically affected by the presence of defects or sharp notches which act as stress-concentrators and favor brittle fracture. In the past 50 years, great research efforts have been carried out to reduce the defect-sensitivity and improve the impact strength of these materials. Within this context, it was found that semicrystalline polymers can be considerably toughened by dispersing a second phase—usually rubber—in the semicrystalline matrix [1,2]. For instance, incorporating 20% of ethylene–propylene rubber particles into a polyamide-6 matrix can lead to a 10-fold increase in the impact resistance of notched specimens [3]. Such toughened semicrystalline plastics are now widely used in a large variety of demanding industrial applications ranging from automotive parts to off-shore pipes. Here, we present microscopic observations of toughened systems which bring a new insight into the mechanisms underlying the toughening of semicrystalline polymers.

In a very schematic way, the toughening strategy consists in modifying the polymer material to dissipate the impact energy by other means than crack propagation. In semi-crystalline polymers toughened with particles, both experimental [4–11] and theoretical [12–16] studies show that toughening results from a multiple step mechanism where cavitation and plastic deformation are crucial. When a crack propagates through an efficiently toughened system, voids are created inside or around the particles due to strong stress concentration at the front of the advancing crack. Particle cavitation and void formation generates a new stress distribution in the material which facilitates the initiation of plastic deformation of the matrix. As a consequence, the matrix ligaments confined between cavitated particles can undergo extensive plastic deformation which dissipates large amount of impact energy. The succession of particle cavitation, the initiation step, and matrix plastic deformation, the dissipative one, are now well-admitted mechanisms. However, the molecular parameters controlling their activation have not yet been clearly identified. The ability to predict toughening efficiency remains a controversial and challenging issue.