Mechanical Properties of an Aromatic Polyamide-imide Composite Film Reinforced with an Aromatic Polyamide Fiber Cloth at High Temperature

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Synopsis

An aromatic polyamide-imide (PAI) film was reinforced with a plain cloth of aromatic polyamide fiber (Du Pont, Kevlar 49). The mechanical properties of the composite film were investigated by examination of the temperature dependencies of tensile dynamic mechanical properties, stress relaxation, and tensile stress-strain behaviors. The softening temperature of the composite film was lower than that of a homogeneous PAI film. At a high temperature, Kevlar fibers may act as a thermal conductor and lower the softening point of the PAI composite. The mechanical properties of the composite film at a high temperature are mostly controlled by the PAI matrix.

INTRODUCTION

For the purpose of producing a flexible composite film, an aromatic polyamide-imide (PAI) matrix is reinforced with an aromatic polyamide fiber (Du Pont, Kevlar 49). Thermally stable flexible films of an aromatic polyamide-imide are obtained by casting from solution in a polar solvent. The fairly flexible composite films of PAI are obtained with an aromatic polyamide fiber as a flexible reinforcement. The mechanical properties and thermal expansion behavior of this type of PAI composite films were investigated by Ohmiya and Kambe[1,2] with respect to the direction of fiber orientation. In unidirectionally reinforced films, the most remarkable effect of the reinforcement fibers was observed at the fiber direction. The elastic and thermal expansion of this film, however, was more isotropic than that of other fiber orientations. The anisotropy problem was solved by bidirectional reinforcement. As for a bidirectionally oriented cloth composite film, the elastic behavior was isotropic at an ambient temperature, but the thermal expansion behavior was anisotropic owing to the PAI matrix.

The thermal stability of the bidirectional cloth composite film is affected by the anisotropy of thermal expansion. The temperature dependence of tensile dynamic mechanical properties, stress relaxation, and tensile stress-strain of the bidirectional cloth composite film were measured, and the role of reinforcing fibers is discussed in the present paper.

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EXPERIMENTAL

Materials

An aromatic polyamide-imide varnish, produced experimentally by Film Laboratory of Toray Co., was kindly supplied us as a material. Dimethylacetamide was used as a solvent in varnish and also used as a diluent.

A commercial fiber of an aromatic polyamide, Kevlar 49, was obtained from E.I. du Pont de Nemours Co. in a form of 195 denier yarn and used as a reinforcement.

Reinforcement Texture

A bidirectional plain cloth of Kevlar 49 was used as a reinforcement of the composite film. Kevlar yarns were twisted clockwise by 220 turns per meter for the warp of a sample cloth. Twisted Kevlar yarns were sized by dipping in a 3% polyvinyl alcohol (PVA) solution. In a plain cloth of Kevlar, the density of the sized warp was 10 yarns per centimeter and that of the unsized woof was 8 yarns per centimeter.

Casting Films

The PAI composite film was prepared as follows. First, a plain cloth was fixed on a glass plate. In preparing the composite film, it was necessary to remove the PVA sizing from the fibers before immersing in a varnish. For this purpose the cloth of the glass plate was soaked in water overnight and dried in a desiccator to remove water completely. The cloth reinforcement was dipped in the PAI varnish together with the glass plate and dried in vacuum overnight. After leaving in air for several hours, the resulting film was heated at 140°C for 3 h at an ambient pressure. The film was removed from the glass plate and heated at 250°C in vacuum 3 h to get a sample film.

Composite Film Samples

By these procedures, the composite films obtained exhibited a considerable flexibility and an enhanced strength compared with the homogeneous PAI film. In the composite films, the yarns are compactly twisted and undulated in the matrix. The volume fraction of the reinforcement in the composite films was estimated at 9 ± 2 vol% from density of the film.

Measuring Methods

Tensile dynamic mechanical measurement was carried out with an Iwamoto viscoelastic spectrometer at a frequency of 10 Hz with a heating rate of 1.5°C/min from 30 to 300°C.

Tensile stress relaxation was measured with a homemade apparatus[3] coupled with a personal computer, at a temperature range of 20–200°C during a period from 0.1 to 10^4 s. The rectangular specimen strips of 5 mm in width and 25 mm in length were used. An initial strain of 0.4% was given on a strip instantaneously, and the stress change was followed automatically at a fixed interval.

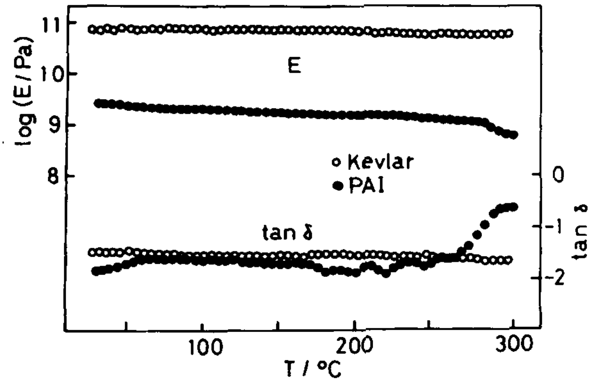

Fig. 1. Temperature dependence of dynamic moduli for Kevlar yarns and PAI film.

Tensile stress-strain measurement was conducted with a Toyo-Baldwin UTM instrument. The tensile stress of dumbbell-shaped specimens 10 mm in width and 20 mm in length was measured at a uniform extension rate of 5 mm/s up to 200°C.

All measurements were conducted in an ambient atmosphere.

RESULTS AND DISCUSSION

Dynamic Mechanical Properties

As shown in Figure 1, the dynamic storage modulus E' and $\tan\delta$ of Kevlar yarns show no significant temperature dependence in the measured range. The E' of PAI film shows a decrease above 280°C. A $\tan\delta$ peak appears at 290°C. The glass transition temperature of PAI film[4] was measured at 279°C by differential scanning calorimetry (DSC) at a heating rate of 16°C/min.

The dynamic storage modulus E' of the composite film at the various fiber orientation angles θ is shown in Figure 2. The E' of the composite film behaves similarly to that of Kevlar fiber at $\theta = 0°$. On the other hand, a small loss peak is observed at 270°C, which reflects the existence of the matrix. At

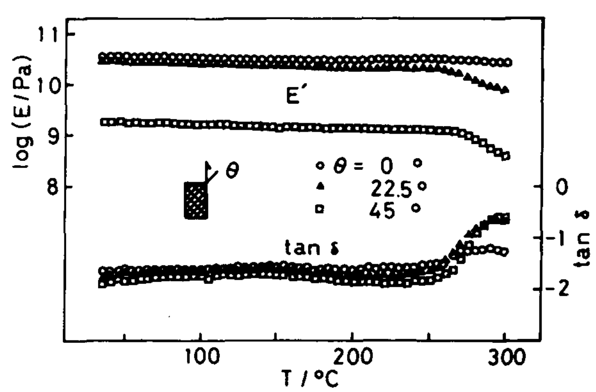

Fig. 2. Temperature dependence of dynamic moduli for composite films at various fiber orientations.

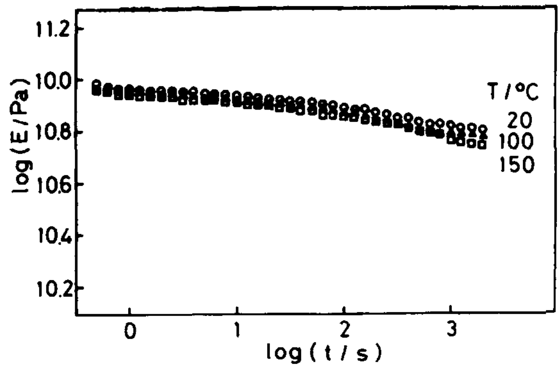

Fig. 3. Stress relaxation curves for Kevlar yarns at 20–150°C in air.

θ = 22.5 and 45°, the significant effects of the matrix are observed on E' and $\tan\delta$ above 260°C. The softening temperature of the composite film is lower than that of PAI matrix. In the composite, plain cloth reinforcement may act the role of a thermal conductor and the softening point of PAI matrix is lowered.

Stress Relaxation

As shown in Figure 3, the stress relaxation curves obtained for Kevlar yarns shows no marked temperature dependence in the measured range. The relaxation curves are linear for these plots, indicating a nonrelaxing behavior of this material.

In Figure 4 the stress relaxation curves of PAI film at various temperatures are shown. The curves are linear for these plots below 100°C. Above 125°C, PAI films show a nonlinear relaxing behavior. In particular, a very marked relaxation is observed at 175°C. This behavior may be caused by the glass transition of PAI matrix.

The stress relaxation curves obtained for the composite film at θ = 0° are shown in Figure 5 Although the relaxation curves were measured at the fiber direction, they showed relaxing behaviors at a long time range above 100°C. In Figures 6 and 7 the stress relaxation curves for the composite films at

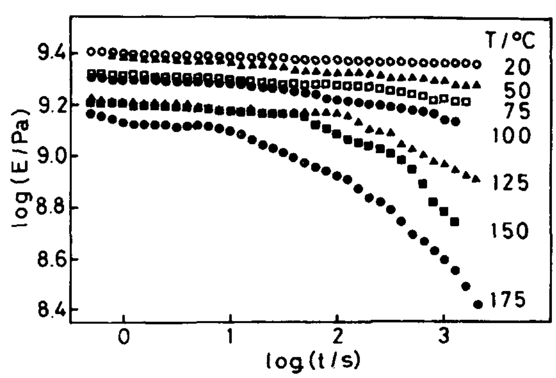

Fig. 4. Stress relaxation curves for PAI film at 20–175°C in air.

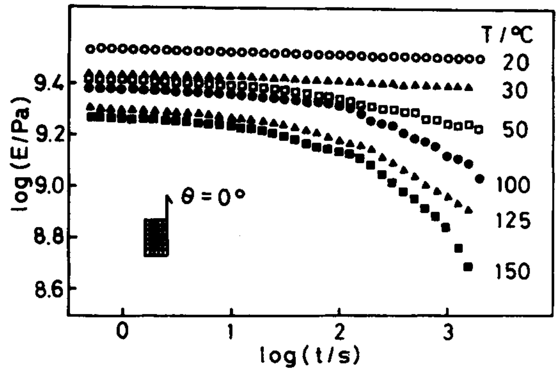

Fig. 5. Stress relaxation curves for the composite film at $\theta = 0°$ at 20–150°C in air.

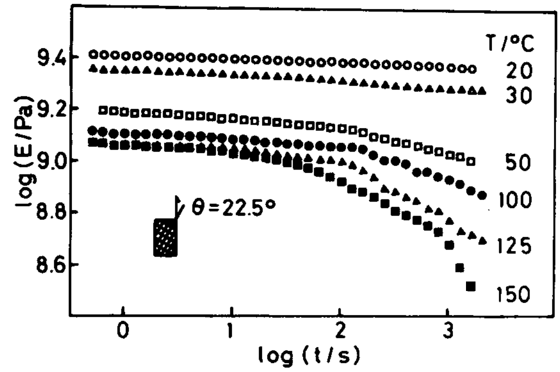

Fig. 6. Stress relaxation curves for the composite film at $\theta = 22.5°$ at 20–150°C in air.

$\theta = 22.5$ and 45° are shown. The relaxation curves above 100°C at a long time range show more significant relaxing behaviors. The relaxing temperature of the composite film is lower than that of PAI matrix. The plain cloth reinforcement may act the role of a thermal conductor to lower the softening point of the composite.

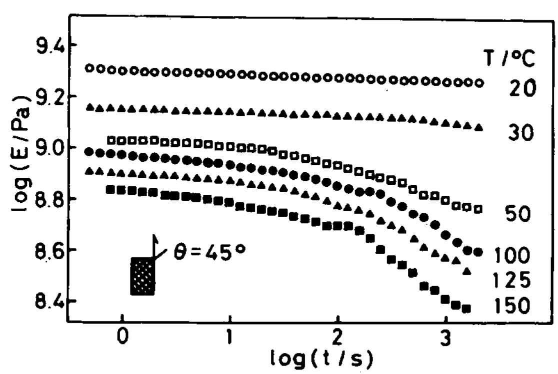

Fig. 7. Stress relaxation curves for the composite film at $\theta = 45°$ at 20–150°C in air.

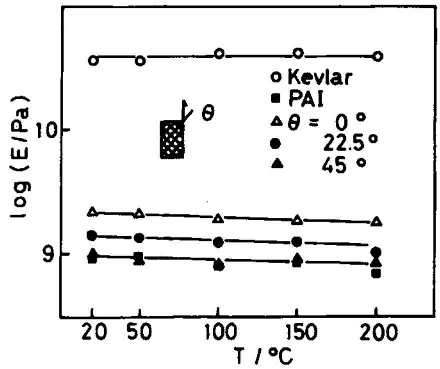

Fig. 8. Temperature dependence of Young's moduli for Kevlar yarns, PAI film, and composite films at various fiber orientations.

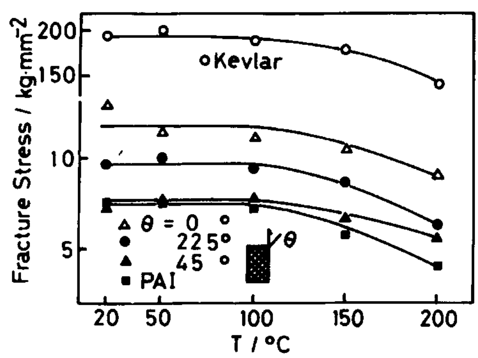

Fig. 9. Temperature dependence of fracture stress for Kevlar yarns, PAI film, and composite films at various orientations.

Stress-Strain Behaviors

The initial Young's moduli E obtained from stress-strain curves for Kevlar yarns, a homogeneous PAI film, and the composite films over a temperature range are shown in Figure 8. The E of PAI and the composite films show a gradual decrease in this temperature range.

The fracture stress of σ is plotted against temperature in Figure 9. For Kevlar yarns, σ retains up to 150°C but shows a small decrease at 200°C. PAI film shows a more significant decrease of σ above 100°C. The composite films show no marked dependence of σ and orientation angle. The fracture stresses of the composite films above 100°C show a more significant decrease of σ, reflecting a characteristic of the PAI matrix.

CONCLUSION

A flexible heat-resistant composite film was produced from PAI and a Kevlar cloth reinforcement. At an ambient temperature, the plain cloth reinforcements can stabilize the composite films. But at a high temperature, Kevlar yarns may act the role of a thermal conductor and lower the softening point of the composite. The plain cloth reinforcement enhances the unstability of the composite film.

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