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Microfailure behaviour of randomly dispersed short fibre reinforced thermoplastic composites obtained by direct SEM observation

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The microfailure behaviour of thermoplastic polyamide 6,6 composites reinforced with randomly dispersed short glass fibres was studied. Scanning electron microscopy was carried out on the surface of the composites under load to observe directly the behaviour. The microfailure proceeds following the steps (1) interfacial microfailure occurs at the fibre tips, (2) the microfailure propagates along the fibre sides, (3) plastic deformation bands of the matrix occurs from the interfacial one, (4) crack opening occurs in the band and the crack grows slowly through the band, (5) finally a catastrophic crack propagation occurs through the matrix with pulling-out fibres from the matrix. A model for the microfailure mechanism of the composites is proposed and some methods to improve the mechanical properties of the composites are discussed on the basis of the mechanism.

1. Introduction

Microfailure is closely related to the mechanical properties of the composites, thus its study is important for improving the properties as well as its reliability. As far as thermoplastic composites reinforced with short fibres are concerned, the microfailure mechanism of the composites does not seem to be well known.

The mechanism has been studied recently. Microscopic stress analysis was made in order to estimate the stress level of the fibre, matrix, and interface generated under a stress equal to or greater than the ultimate tensile stress. It showed that the interfacial microfailure would occur in the initial stage of the composite failure [1, 2]. The acoustic emission technique was applied to the composites under stress to detect the initiation of microfailure. The measurement proved that the microfailure begins to occur at a load much lower than the ultimate load, and that accumulation of the microfailure with the load leads to final failure of the composites [1, 3, 4]. Fractographic study using scanning electron microscopy (SEM) revealed the state of adhesion between the fibre and matrix [5], a characteristic crack propagation pattern starting from the broken fibre tips [6], fatigue crack propagation along the fibres [7–11], and energy dissipation mechanism in the composites was suggested [12]. These studies provide useful knowledge about the microfailure in the composites. Many of these are focused mainly on the fracture surface of the composites after failure. Detailed information about the process of microfailure seems not to have been given. If the process were observed directly, more useful information would be obtained.

The object of this study is to investigate the process of microfailure of thermoplastic polyamide 6,6 composites reinforced with randomly dispersed short glass fibres. The microfailure behaviour of the composites was studied by *in situ* SEM observation proposed in our previous studies [13, 14]. On the basis of these results a model for the microfailure mechanism of the composites was proposed and some methods to improve the mechanical properties of the composites were discussed from the viewpoint of the microfailure mechanism of the composites.

2. Experimental procedure

2.1. Specimen

The composites are thermoplastic polyamide 6,6 ones reinforced with randomly dispersed short glass fibres. The composite specimen was made of thermoplastics pellets and the chopped strand of glass fibres (13 μm in diameter) by conventional processes of compounding and injection moulding. The glass fibres were treated with a silane coupling agent. The content of the fibres was 30% by weight. The dimension of the moulded specimen was the same as a tensile test piece specified in ASTM D638. The average fibre length in the specimen was about 300 μm. The fibres were distributed randomly in the matrix with the exception of the surface layer. The fibres in the layer were slightly oriented along the flow direction of the moulding (the longitudinal direction of the tensile test piece).

2.2. *In situ* SEM observation

In situ SEM observation was carried out to study the behaviour of microfailure of the composites. The specimen for the observation was 50 mm in length, 13 mm in width, and 3 mm thick, and was obtained from the tensile test piece by cutting off both holder parts. The surface of the specimen (50×13 mm^2) was polished by a metallurgical polishing technique with a buff and alumina powder (0.05 μm), and was slightly coated with gold for SEM observation. The specimen was then subjected to a bending load with a small three-point bending device (45 mm of span length) and set on the sample stage of a SEM apparatus. SEM examination was carried out on the tensile surface of the specimen under load to observe initiation of microfailure occurring on the surface. The examination was focused mainly on the region of maximum deflection of the specimen. After the examination, the bending load was slightly increased and the SEM examination was carried out again to observe propagation of microfailure on the surface. This procedure was repeated with stepwise increase of the load until the specimen failed. The process of microfailure as well as the morphology of the microfailure under load was obtained by this observation.

3. Results and discussion

3.1. Process of microfailure of composites

3.1.1. Initiation of interfacial microfailure at fibre tips

The surface of the specimen for the SEM observation is shown in Fig. 1. As shown in the figure, any defect was hardly observed in the fibre, matrix, nor interface between them. When load was applied to the specimen, a cluster of microcracks was observed to occur in the matrix around the fibre tips. Fig. 2 shows a typical example of the cluster of microcracks at the fibre tips, which was obtained under a load of about 50% of failure load of the specimen. The microcracks were considered to be generated by tensile stress concentration around the fibre tips. As the applied load was slightly increased, separation at the interface between the matrix and fibre tip occurred and a microvoid was formed there. Fig. 3 shows a typical example of the microvoid formed at the interface of the fibre tips, thus, the initiation of the interfacial microfailure at the fibre tips was observed as the first stage of microfailure in the composites.

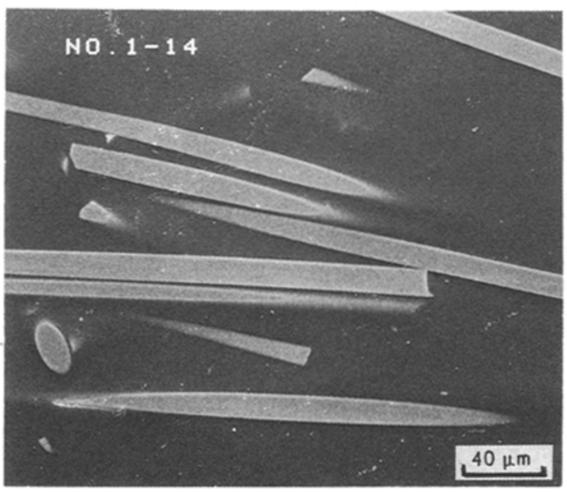

Figure 1 The surface of the specimen before loading, which was buffed and polished with alumina powder and coated with thin gold film.

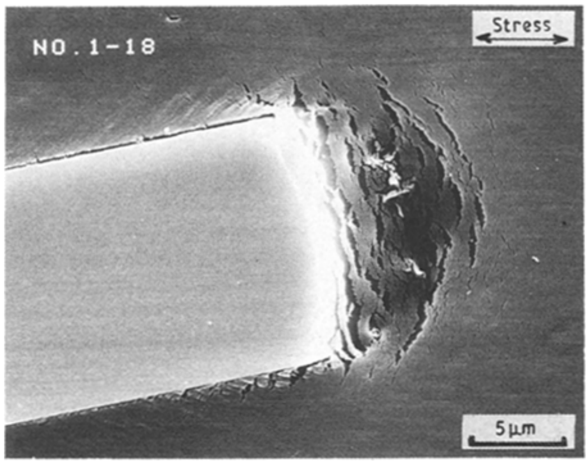

Figure 2 A cluster of microcracks occurring in the matrix around the fibre tip, which was obtained under about 50% of failure load.

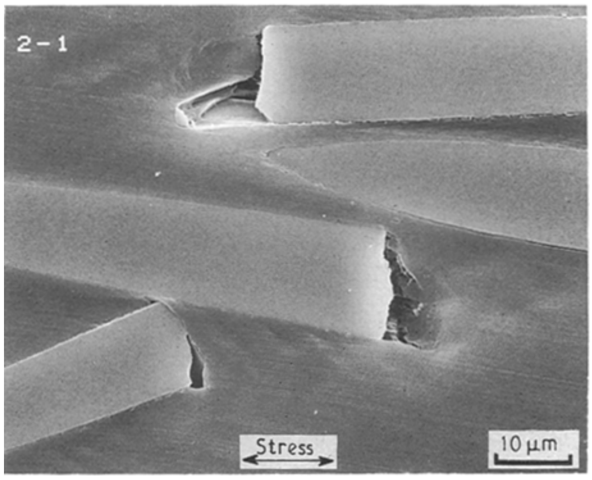

Figure 3 Microvoid occurring at the interface separating the fibre tip from the matrix, which was obtained under 50% of failure load.

3.1.2. Propagation of interfacial microfailure along fibre sides

As the applied load was further increased, the fringe pattern of shear mode and microcracks were observed to occur in the matrix along the fibre sides. The microfailure was then observed to propagate from the fibre tip along the interface of the fibre sides. Fig. 4 shows a typical example of the interfacial microfailure along the fibre sides, which was obtained under a load of about 75% of failure. The interfacial microfailure is considered to be generated by shear stress concentration along the fibre sides. The propagation of the interfacial microfailure along the fibre sides was observed as the second stage of microfailure.

3.1.3. Occurrence of plastic deformation band in matrix region

When the applied load was increased to near failure, bands of microcracks were observed to occur in the

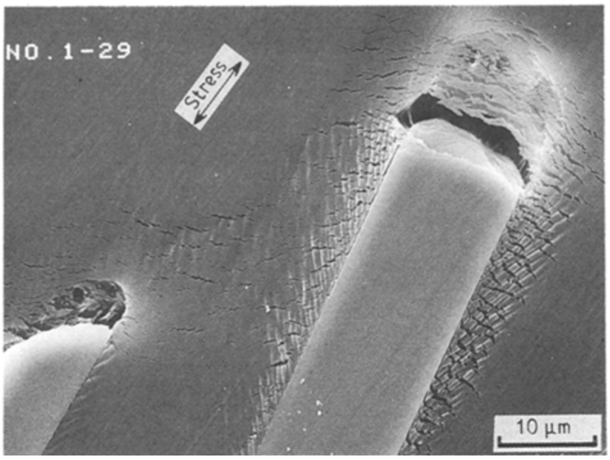

Figure 4 Interfacial microfailure along the fibre sides, which was obtained under about 75% of failure load.

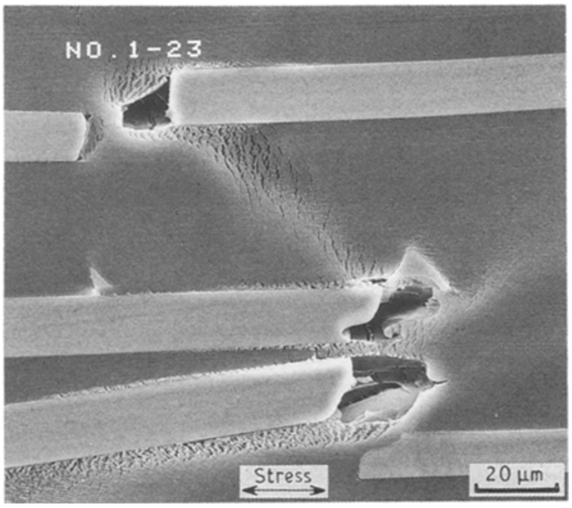

Figure 5 Band of microcracks in the matrix region, which was obtained under about 90% of failure load. The band indicates the occurrence of plastic deformation of the matrix beneath the surface.

matrix region propagating from the interfacial microfailure. Fig. 5 shows a typical example of the band of the microcracks running from one fibre tip to another one, which was obtained under the load of about 90% of failure one. A magnified view of the microcracks is shown in Fig. 6. The depth of microcracks was observed to be very small and the bottom was observed to be flat. From the morphology, it is considered that the apparent microcracks are not true failure of the matrix, but just failure of the gold film on the surface of the specimen. The microcracks are considered to be generated due to large deformation of the matrix beneath the film thus the band of the microcracks indicates that the part of the matrix is subjected to large deformation. Strain of the matrix beneath the microcracks could be approximately estimated from the separation between microcracks. The estimated strain was about 50 to 100%, which was much greater than the yield strain of the matrix and nearly equal to the failure strain of the matrix. The presence of the band of the microcracks indicates, therefore, the large plastic deformation of the matrix generated in the localized region.

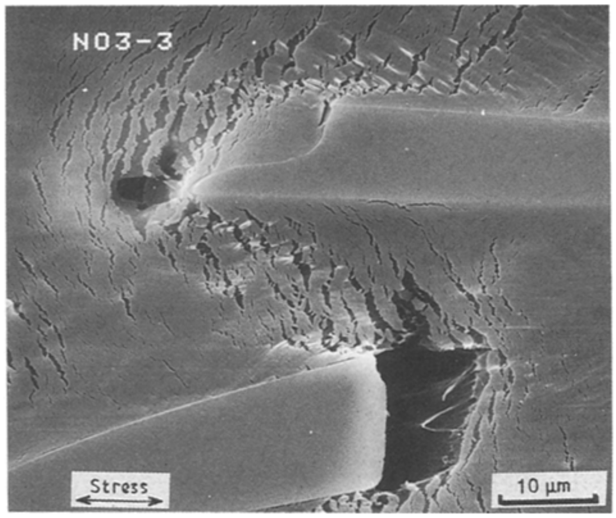

Figure 6 Magnified view of microcracks in the matrix region which was obtained under load. The microcracks are due to failure of the thin gold film on the surface, thus they indicate the occurrence of large plastic deformation of the matrix beneath the surface.

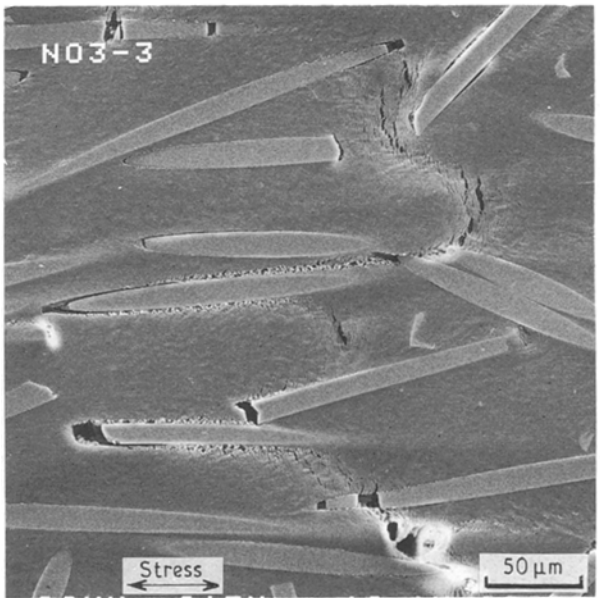

Figure 7 A large zigzag band of microcracks of the matrix, which was obtained under load just before failure of the specimen. The band indicates the occurrence of the long plastic deformation band of the matrix connecting the interfacial microfailure at the fibre tips and along the fibre sides.

The bands of the microcracks were observed to propagate connecting the interfacial microfailure along the fibre sides and at the fibre tips. They formed some long zigzag bands in the specimen. Fig. 7 shows a typical example of the long zigzag band, which was obtained just before the occurrence of failure of the specimen. As seen in the figure, the plastic deformation of the matrix is generated only in a narrow region of the matrix and hardly observed in the rest of the matrix. This fact indicates that only a localized region of the matrix is subjected to stress concentration, and that most of the matrix is in elastic deformation. The stress concentration is considered to be caused by the reduction of the load bearing capability of fibres due to the interfacial microfailure around the fibres.

The occurrence of the plastic deformation bands in the matrix region was thus observed as the third stage of microfailure of the composites.

3.1.4. Crack opening and slow crack propagation

When the load was increased very near to failure, the matrix in the plastic deformation bands was strongly stretched. Finally tearing of the matrix occurred, which was responsible for the initiation of crack opening in the matrix region. Fig. 8 shows a typical example of the crack opening obtained just before the failure of the specimen. The crack opening was sometimes generated from the interfacial microfailure at fibre tips or fibre sides. Fig. 9 shows another example of the crack opening which was generated from the interfacial microfailure along the fibre side. The cracks were observed to grow slowly through the plastic deformation bands leaving the dimples of the

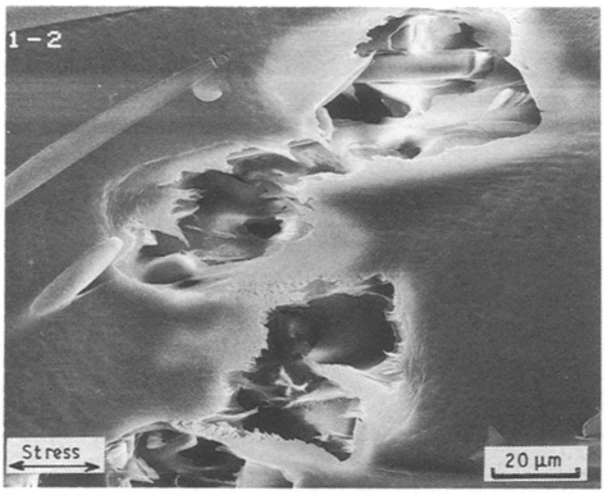

Figure 8 Crack opening occurring in the large plastic deformation band of the matrix, which was obtained under load just before failure.

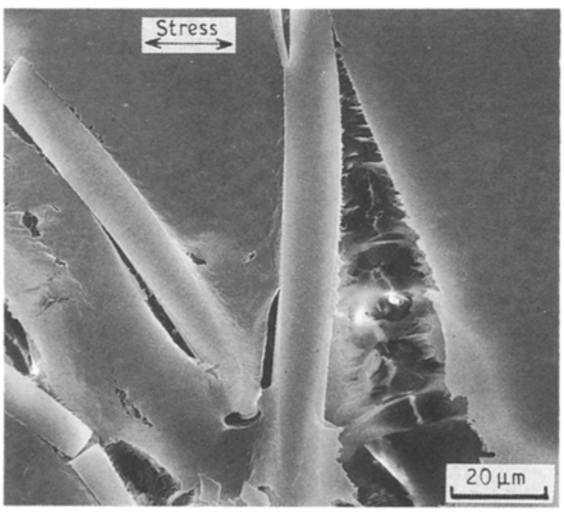

Figure 9 Crack opening grown from the interfacial microfailure along the fibre side, which was obtained under load just before failure.

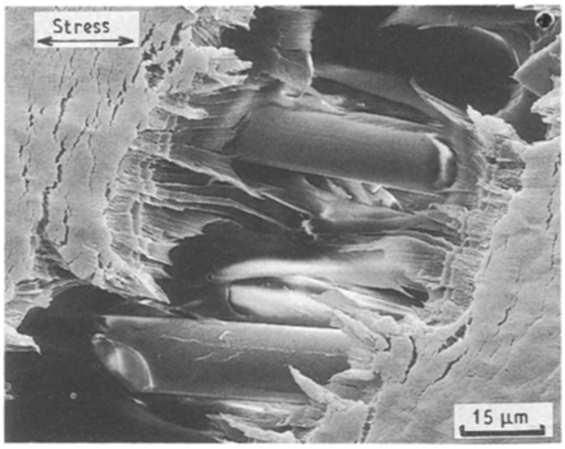

Figure 10 Fibre pull-out from the matrix in the period of the ductile crack propagation, which was obtained under load just before failure.

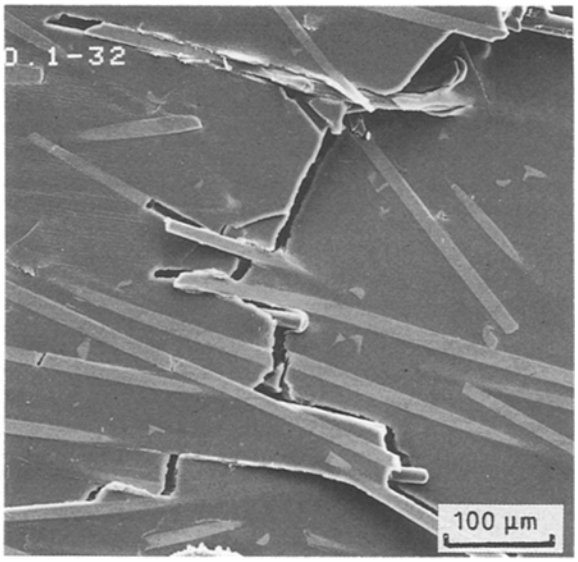

Figure 11 Fracture surface observed after failure, which shows the brittle crack propagation along the interface of the fibre sides.

stretched matrix on the crack surface. The fibres in the front of the crack were observed to be pulled out from the matrix. Fig. 10 shows a typical example of the fibre pull-out from the matrix. The crack opening and slow crack propagation of the matrix were observed as the fourth stage of microfailure of the composites.

3.1.5. Fast crack propagation

When the crack grew to some critical size (about 1 mm in length), a catastrophic crack propagated through the matrix leading to final failure of the specimen. Fig. 11 which was obtained after failure shows the brittle crack propagation along the fibre sides. Most of the fibres on the fracture surface were observed to be pulled out from the matrix as shown in Fig. 12. The fast crack propagation of the matrix with fibre pull-out was observed as the final stage of microfailure of the composites.

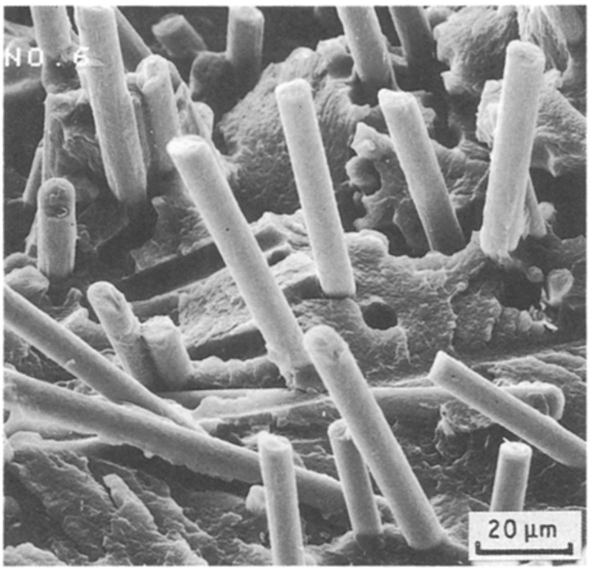

Figure 12 Fracture surface observed after failure. Most of the fibres were pulled out from the matrix.

3.2. Observation of tensile fracture surface

In order to study the analogy between the bending failure mode and tensile failure one, tensile fracture surface obtained by a tensile test was examined by SEM. Fig. 13 shows the tensile fracture surface of the specimen. Most of the surface was covered with the pulled-out fibres and a patchwork pattern of the matrix which is a characteristic brittle failure mode of the matrix. Only one region (about 1 mm of diameter) was, however, observed to be covered with the pulled-out fibres and stretched dimples of the matrix which is a characteristic ductile failure mode of the matrix. The ductile region was isolated in the brittle failure mode. From the morphology, it is considered that the small ductile region is the initiation of the crack and the crack propagates through the specimen in a brittle manner leading to final failure of the specimen. This failure mode in tension is considered to be essentially the same as that in the bend obtained in the previous section.

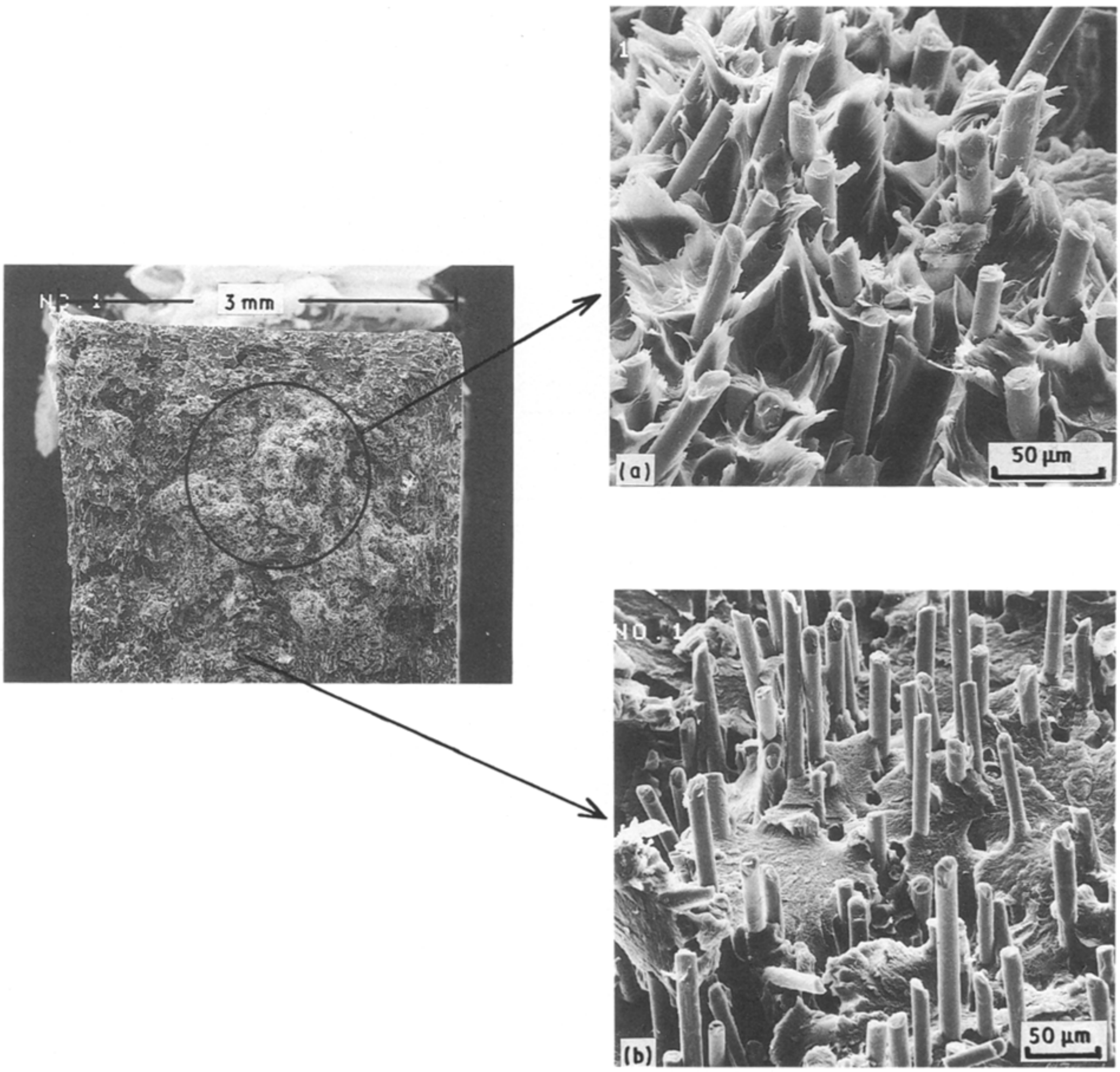

Figure 13 Tensile fracture surface. (*a*) Ductile failure mode observed in the only one region located near the centre of the fracture surface, (*b*) brittle failure mode observed all around the ductile one.

3.3. Stress–Strain curve of composite

The tensile stress–strain curve of the specimen was obtained in order to study the relationship between the macroscopic deformation behaviour and the microscopic failure behaviour of the composites. Fig. 14 shows a tensile stress–strain curve of the specimen. The composites showed linear deformation with the increase of load and non-linearity at high stress. Brittle failure occurred at the ultimate load. The change from the linear to non-linear deformation occurred at a load of about 80% of failure. In such a load range the interfacial microfailure around the fibres was generated in the composites as shown in Figs 4 and 5. The interfacial microfailure around fibres is responsible for the reduction of the load bearing capability of the fibres, therefore the composite is considered to exhibit non-linearity at higher stresses.

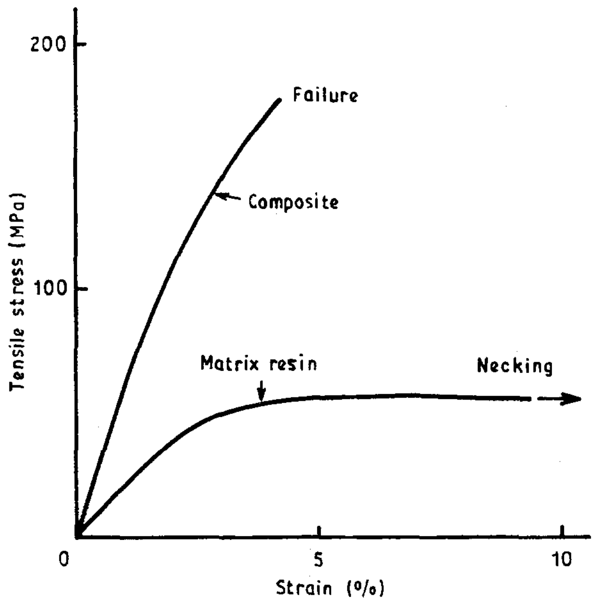

Figure 14 Stress–strain curve of the composite specimen obtained by a tensile test.

The brittle failure at the maximum load is considered to result from the restriction of the plastic deformation of the matrix. As shown in Fig. 7 the matrix in the composites exhibits large plastic deformation as the neat matrix resin. The occurrence of the plastic deformation is, however, localized only in a narrow region of the matrix subjected to stress concentration, and the extension of the deformation is restricted due to the presence of lots of fibres. When such composites are subjected to much greater load, the matrix is torn and a catastrophic crack propagation occurs. This is considered to be the reason why the composites fail in a brittle manner.

3.4. Model of microfailure mechanism

A model of the microfailure mechanism of the composites is proposed on the basis of the results obtained by the *in situ* SEM observation. Fig. 15 shows the process of microfailure of the composites, which consists of five stages. The first stage is the initiation of the interfacial microfailure at the fibre tips. The matrix around the fibre tips is subjected to tensile stress concentration, so that microfailure separating the matrix from the fibre tips occurs. The second is the propagation of interfacial microfailure along the fibre

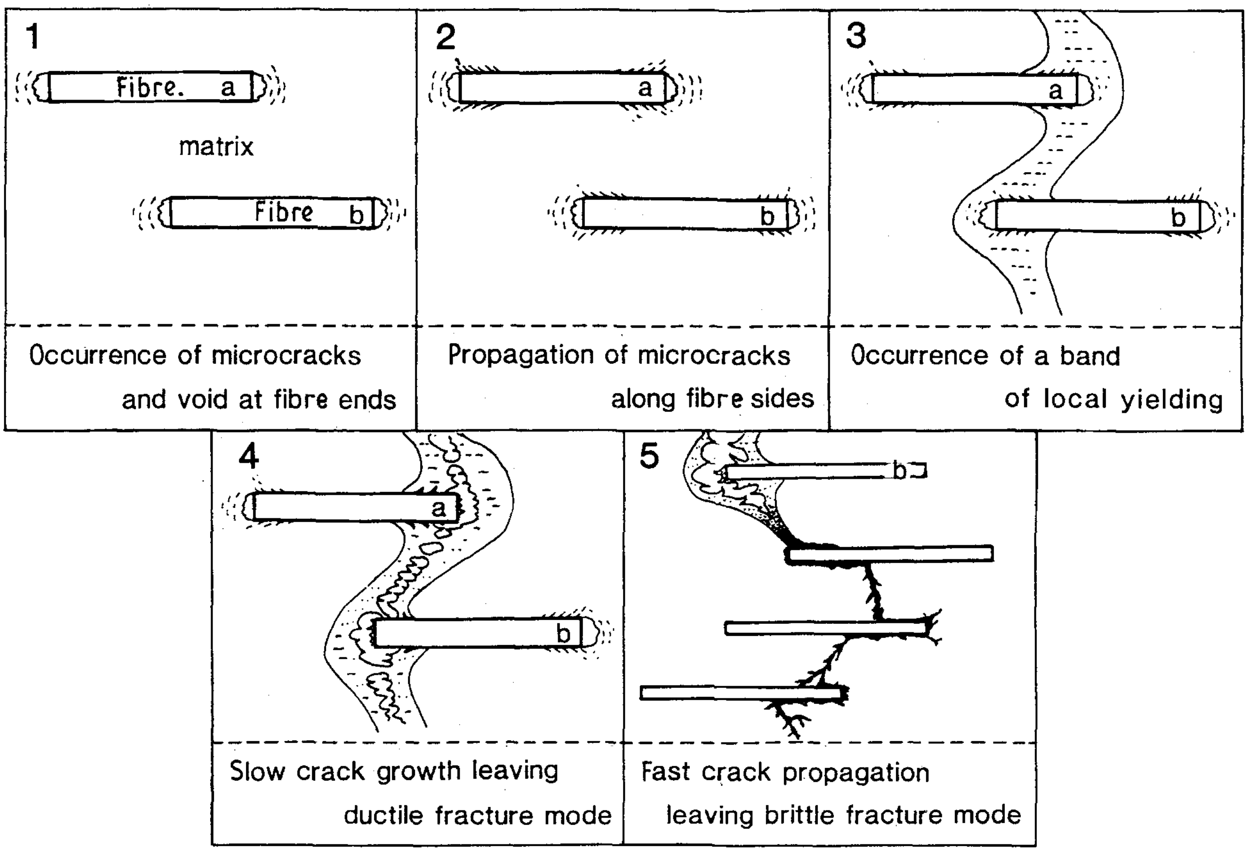

Figure 15 Model of microfailure process of composites reinforced with randomly dispersed short fibres.

sides. The matrix along the fibre sides is subjected to shear stress concentration. When the shear deformation reaches the critical value, the interfacial microfailure propagates from the fibre tips along the fibre sides. The third is the occurrence of the plastic deformation band of the matrix. The interfacial microfailure around the fibres results in localized stress concentration in the matrix region, so that the plastic deformation band of the matrix occurs. The band propagates along the interfacial microfailure along the fibre sides and fibre tips. The fourth is the crack opening and slow crack propagation. When the matrix in the band reaches the limit of deformation, tearing of the matrix occurs, which is the beginning of the crack opening. The crack grows slowly through the band connecting the interfacial microfailure along the fibre sides and at the fibre tips. In the period of the slow crack propagation, the fibres in the front of the crack tip are pulled out from the matrix. When the crack grows to the critical size, a catastrophic crack propagation occurs through the composites in a brittle manner. This is the final stage of microfailure of the composites. Most of the fibres are pulled out from the matrix in the period of brittle crack propagation, because the interface along the fibre sides has already been damaged in the previous stages.

3.5. Material design

The methods to improve the composite strength and toughness are briefly discussed on the basis of the microfailure mechanism. As observed, microfailure at the interface between the fibre and matrix is responsible for composite failure, so that good adhesion at the interface is necessary for improvement. Fig. 16 shows a magnified view of the pulled-out fibres. The fibres are observed to be covered with the thin film of the matrix. It indicates that the interfacial microfailure is caused by cohesive failure of the matrix very near the interface, not by simple debonding at the interface. This fact indicates that the adhesion at the interface is sufficient, and it is considered that further research of the fibre surface treatment for good adhesion is not an effective way to improve the strength and toughness in this composite.

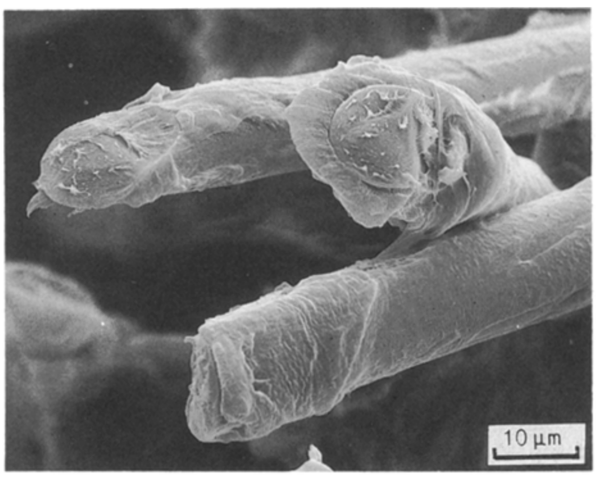

Figure 16 Magnified view of the surface of the fibre pulled-out from the matrix. The surface was covered with a thin film of the matrix, which indicates the cohesive failure of the matrix at or very near to the interface.

The initiation of the interfacial microfailure should be suppressed for the improvement of composite strength and toughness. Three methods to suppress the interfacial microfailure would be possible. The first one is to use the fibres whose diameter is smaller than those of the conventional ones. The smaller diameter fibres give many more fibres in the composites for the same weight fraction of fibres. In the composites reinforced with smaller diameter fibres, the stress concentration at the fibre tips is considered to be reduced due to closer packing of the fibres. The interfacial microfailure is consequently expected to be suppressed through the reduction of the stress. Experimental study of the method has already been made and has proved to greatly improve the composite strength and toughness [15].

The second is to use longer fibres than conventional ones. It is said that the fibre length strongly affects the mechanical properties of the composites. The fibre longer than the critical fibre length has high efficiency of the load bearing capability. The method which uses the long fibres is considered to effectively improve the composite strength and toughness. The method also has the effect to reduce the number of the fibre tips which act as the site of stress concentration. The glass fibres are, however, broken and shortened in the processes of compounding and injection moulding. New process free from the fibre breakage must be developed for this method.

The third is to introduce the interfacial layer at the interface between the fibre and matrix. If a soft layer such as rubber could be introduced at the interface, the initiation of the interfacial microfailure would be expected to be reduced due to a large deformation of the layer. On the contrary, if a rigid layer whose modulus is intermediate between that of the fibre and matrix could be introduced at the interface, the stress concentration along the fibre sides is considered to be lowered. The initiation of the interfacial microfailure is expected to be reduced through the reduction of the stress concentration. The introduction of the interfacial layer would be possible by making interpenetrating structure of polymer network and polymer alloying.

4. Conclusion

The microfailure behaviour of thermoplastic polyamide 6,6 composites reinforced with short glass fibres was studied by *in situ* SEM observation. Interfacial microfailure at the fibre tips and along the fibre sides were found to be the essential mechanism for microfailure of the composites. The interfacial microfailure was caused by the cohesive failure of the matrix very near the interface, not by the simple debonding at the interface. Fibres were not broken, but pulled out from the matrix in the failure of the composites. Matrix plastic deformation was generated from the interfacial microfailure around the fibres due to the stress concentration. Finally the crack opening occurred in the matrix starting from the interfacial microfailure. The crack propagated through the matrix connecting the interfacial microfailure, which led to final failure of the composites. The microfailure mechanism obtained in this study is considered to be essential for other types of short fibre composites.

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