Performance of Polyamide-6 /Wollastonite / Kaolin Hybrid Composites

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ABSTRACT

The aim of this paper was to find the effect of flake-like kaolin and needle shaped wollastonite fillers on microstructural changes of polyamide-6 and its mechanical properties derived from tensile, izod impact and three point bending tests. One or more than one filler with varying weight percentage between 10 to 30wt.% were added to polyamide-6. Of the mechanical properties, tensile strength and modulus, flexural strength and modulus increased, whereas impact strength and elongation at break decreased with increasing filler content for single and mixed composite. The maximum improvement in mechanical properties is recorded with the addition of 20 to 30wt.% filler ratios. Scanning electron microscope has been used to reveal microstructural properties of the kaolin and wollastonite filled polyamide-6 composites. In addition, the microstructural changes of kaolin and wollastonite filled polyamide-6 composites were discussed in relation to its mechanical properties.

Key words: polyamide-6, kaolin, wollastonite, hybrid composites

1. INTRODUCTION

Many commercial polymer materials are composites, because they provide reduced cost and improved mechanical, thermal and electrical properties that cannot be obtained from simple polymers. A great variety of the physical properties can be obtained through alternating compositions of the polymer composites /1/. Polyamides are one of the most widely used engineering thermoplastics, especially in automotive, electric/electronic, packaging, textiles and consumer applications because of their excellent mechanical properties /2-4/. Mineral fillers are defined as additives in solid form that differ from the plastics matrix with respect to their composition and structure. Fillers are generally low-cost additives that increase the bulk. Furthermore, they produce specific improvements in certain mechanical and physical properties. It has been reported that fillers in polyamides produce the following improvements in properties: increase in stiffness, tensile strength, modulus of elasticity, hardness and heat deflection temperature. However, it is observed that elongation, impact strength and melt index are decreased. In general, mechanical properties of mineral filled polymer composites depend strongly on the size, shape, distribution of filler particles in the matrix polymer and good adhesion at the interface surface /5,6/. Many studies have been done for the utilization of polyamides as matrix resins for composites by adding inorganic fillers such as glass beads /4, 7-10/, kaolin /4, 7,11-13/, fly ash /14/, wollastonite /4, 7, 13-15/, mica /16-18/, talc /4, 7, 13, 19, 20/, alumina trihydrate /13/, calcium carbonate /13, 22/, clays /13/, silica /13/, and whiskers /22-24/. Among these fillers, wollastonite of needle-shaped structure provides many processing and performance benefits, including increased stiffness and strength, improved heat distortion temperature, low coefficient of expansion, ease of processability, and good reinforcement at considerably low costs. Kaolin has a plate-like structure and provides an optimal balance of impact strength, dimensional stability, heat deflection temperature and a good surface appearance. In this study, firstly, polyamide-6 composites filled with different contents of kaolin and wollastonite fillers have been investigated. Then, the preparation of the polyamide-6 composites with mixed kaolin and wollastonite fillers has been investigated. The goals are to reduce the cost of the polyamide-6 composites and to obtain good mechanical properties, because mineral fillers such as kaolin and wollastonite are cheaper than reinforcements such as glass fibers and carbon fibers. Therefore, the effects of various filler ratios on the mechanical properties of polyamide-6 composites were examined.

2. EXPERIMENTAL

2.1. Materials and Sample Preparation

In this investigation, the matrix material polyamide-6 (Domamid) with 1.14g/cm^3 density and melt flow index (2.16 kg, 230°C) 4 g/10 min., was obtained from Domopolymers in Belgium. The kaolin powder grade Dorkafil 601 (average particular size 1,7 μm), with a density of 2.6 g/cm^3, was supplied by Dorfner. The other filler was wollastonite of powder grade Nyglos M15 (average particle size 46,8 μm, aspect ratio:10/1, density: 2.9 g/cm^3), that was supplied by Nyco Minerals. Both kaolin and wollastonite fillers are surface treated with amino-silane coupling agent. In the sample preparation process, the kaolin and wollastonite were added separately or in combination to polyamide at 10, 15, 20, 25 and 30 wt.% ratios. The composite granules were prepared using twin-screw extruder NR II-75. The temperature profiles of the barrel were 240, 260, 265, 260,255, 250, 240, 240, 240, 250, 250°C from hopper to die. The extrudate was pelletized, dried, and injection molded into standard test samples for mechanical properties tests. The injection molding temperature and pressure were 230°C and 12 MPa, respectively.

2.2. Mechanical and morphology properties

In this experimental study, tensile tests of the specimens were carried out on a Zwick machine of Model Z020 at 5mm/min cross-head speed. Samples were measured according to ISO 527. Notched impact strength was tested according to ISO 180/1A and a Zwick testing machine was used. The flexural strength and flexural modulus of the specimens were measured by a Testometric Micro 350 machine and samples were measured according to ISO 178. All tests were performed at ambient temperature (23±2°C) with constant humidity. At least five samples were tested for each composite composition, and the average value is reported. The morphology of the fracture surface of the specimens was examined by a scanning electron microscope (Camscan S4). The fractured surface of tensile specimens were coated with a thin layer of gold before observation.

3. RESULTS AND DISCUSSION

3.1. Mechanical Properties

To optimize the ratio of fillers, the mechanical properties of the composites filled with a single filler of kaolin and wollastonite were investigated. With the increase of kaolin and wollastonite content in the polyamide-6 polymer, tensile strength increased. Furthermore, tensile strength exhibited a maximum value at a weight ratio of 20% for single fillers (Figure 1). For mixed state, it increased up to 20wt.% filler ratio, above which no significant changes was observed. Tensile strength values of polyamide-6/kaolin and polyamide-6/wollastonite composites are close to each other. For single fillers, the highest increase in strength is about 17% and 18% for kaolin and wollastonite respectively. As for mixed fillers, the increase in tensile strength is about 20% for kaolin/wollastonite mix.

Figure 2 demonstrates the variation of modulus of elasticity of polyamide-6 with respect to filler content for both the single and mixed fillers. The result shows that the modulus of elasticity follows a similar increasing pattern for kaolin and wollastonite fillers. The modulus of elasticity of polyamide-6/wollastonite composites containing 30wt.% wollastonite is 140% higher than that of polyamide-6 polymer. Moreover, the modulus of elasticity of polyamide-6/kaolin/ wollastonite composites containing 15wt.%

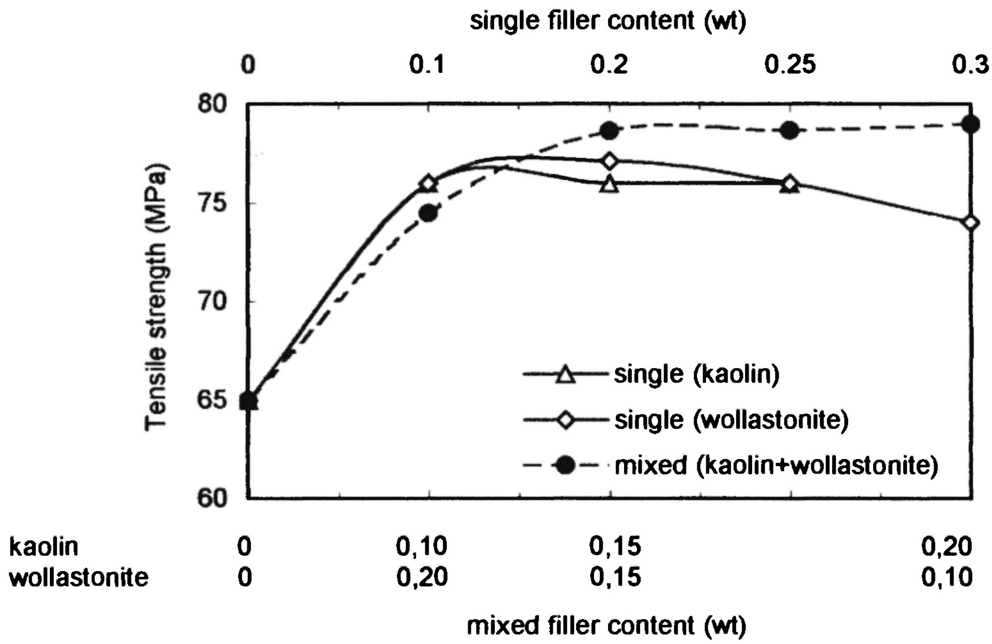

Fig. 1: Tensile strength-filler content wt curves for polyamide-6 composites

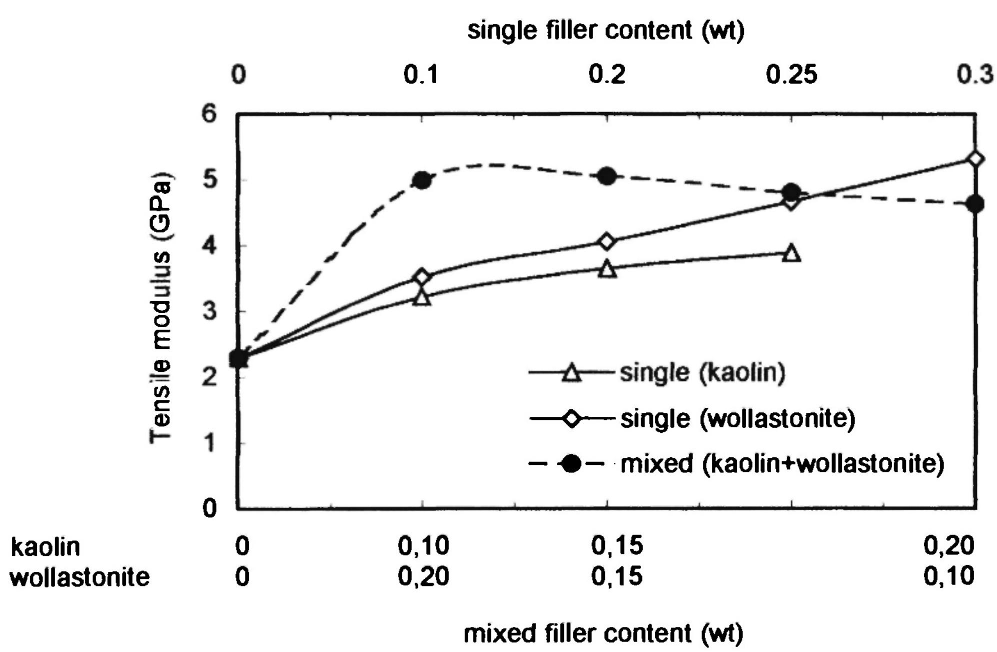

Fig. 2: Tensile modulus-filler content wt. curves for polyamide-6 composites

kaolin+15wt.%wollastonite fillers is 132% higher than that of polyamide-6 polymer. As expected, the modulus of elasticity increases with increasing kaolin and wollastonite fillers content as shown in Figure 2. This is due to the fact that firstly, the polyamide is substituted largely by the more rigid filler particles and secondly, the filler presence restricts the mobility and deformability of the polyamide matrix by introducing a mechanical restraint.

Figure 3 shows the variation of elongation at break with filler content for single and mixed fillers. The results show that for single and mixed fillers, the elongation at break decreases linearly with the increase in filler ratio up to 10wt.% and no significant changes occur above this ratio. For kaolin and wollastonite filler and filler combinations the decrease is from 88-96%. This shows the equal influence of the addition of these fillers as single and as mixed state. This can be explained by immobilization of the polymer chains due to the presence of the filler in the polymer matrix which increase the brittleness of the polymer.

Figure 4 illustrates the variation of notched izod impact strength with filler content for single and mixed fillers. The result shows that polyamide-6 possesses very high toughness. In the case of single filler, the izod impact strength decreases linearly with the increase in filler content. This decrease is about 23%. As for mixed filler, there is a decreasing and an increasing profile. Having said that, there is an increase of almost 3% in impact strength of the polyamide composite. This suggests the lower degradation in impact strength by the mixed fillers addition. The degradation in impact properties can be attributed to the immobilization of the macromolecular chains by the filler, which limits their ability to deform freely and hence making the material less ductile /25/.

Figures 5 and 6 display the variation of the flexural strength and flexural modulus with the addition of single and mixed kaolin and wollastonite fillers respectively. It is clear from Figure 5 and Figure 6 that the flexural strength and flexural modulus are increasing almost linearly with the increase in the filler ratio up to 10wt.%. As for mixed state, they increase to a maximum value at 15wt.%kaolin+15wt.%wollastonite filler ratio. The result shows that flexural strength follow a similar increasing pattern for kaolin and wollastonite fillers. The flexural strength of polyamide-6/kaolin composites containing 25wt.% kaolin is 105% higher than that of polyamide-6 polymer while the flexural strength of polyamide-6/wollastonite composites containing 30wt.% wollastonite is 75% higher than that of polyamide-6 polymer. The flexural strength of polyamide-6/kaolin/wollastonite composites containing 15wt.% kaolin+15wt.% wollastonite is 130% higher than that of polyamide-6 polymer. As can be seen in Figure 6, the flexural modulus of polyamide-6/kaolin and polyamide-6/wollastonite composites is 196% and 244% higher respectively than that of polyamide-6 polymer. In the case of mixed fillers, the increase in flexural modulus is about 312% for 15 wt.%kaolin+15wt.% wollastonite.

3.2. Morphology

The SEM micrographs of the polyamide-6/kaolin/wollastonite composites with different wollastonite and kaolin contents are shown in Figures 7a and b. It can be noted from Figures 7a and b that the wollastonite particles are randomly oriented in the polyamide-6 matrix with poor adhesion. Furthermore, Figures 7a and b reveal the existence of aggregates. This is clearer for higher kaolin percentage in polymer composites. In general, the kaolin particles are strongly inclined to agglomerate compared to those of wollastonite, because of the nature of their surface. According to Griffith's theory, a large aggregate is a weak point, which lowers the stress required for the composite to fracture. As shown in Figure 7a, large aggregates can be distinguished in the micrograph of the fracture surface and it is also clear that the crack propagated through it.

4. CONCLUSIONS

Polyamide-6/kaolin, polyamide-6/wollastonite and polyamide-6/kaolin/wollastonite composites are studied for the purpose of producing cost-effective composites and improving mechanical properties of the materials. Tensile measurements show that tensile strength and

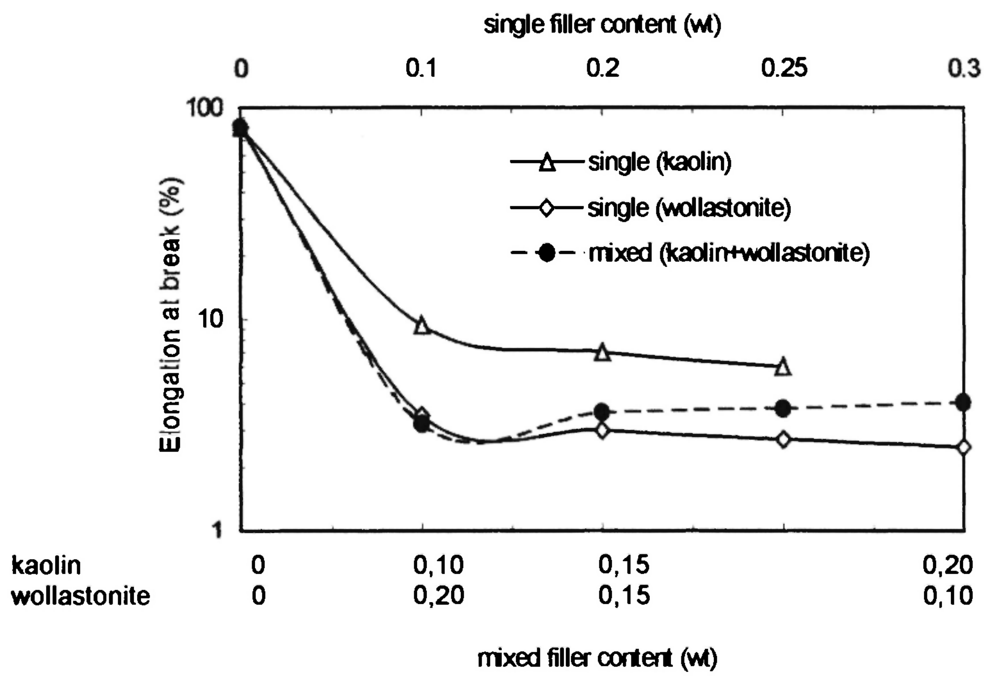

Fig. 3: Elongation at break-filler content wt. curves for polyamide-6 composites

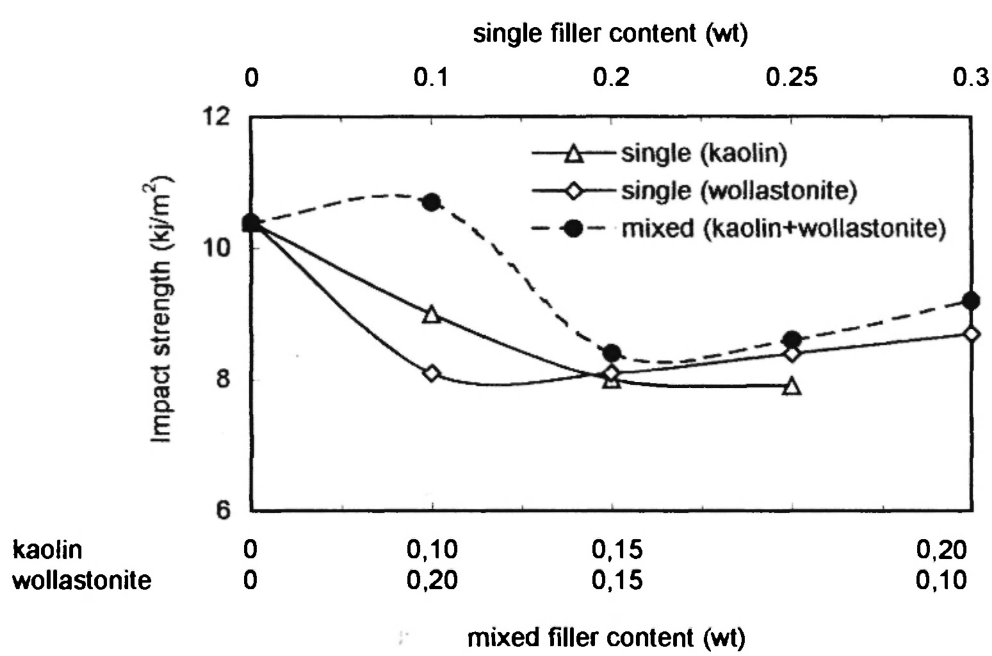

Fig. 4: Impact strength-filler content wt. curves for polyamide-6 composites·

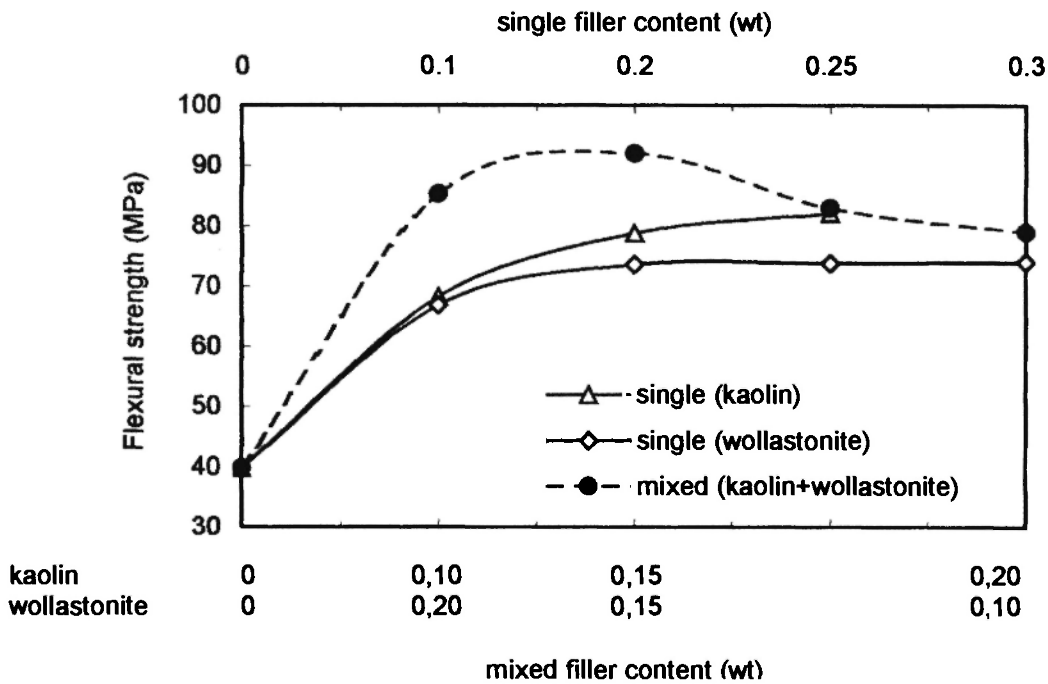

Fig. 5: Flexural strength-filler content wt. curves for polyamide-6 composites

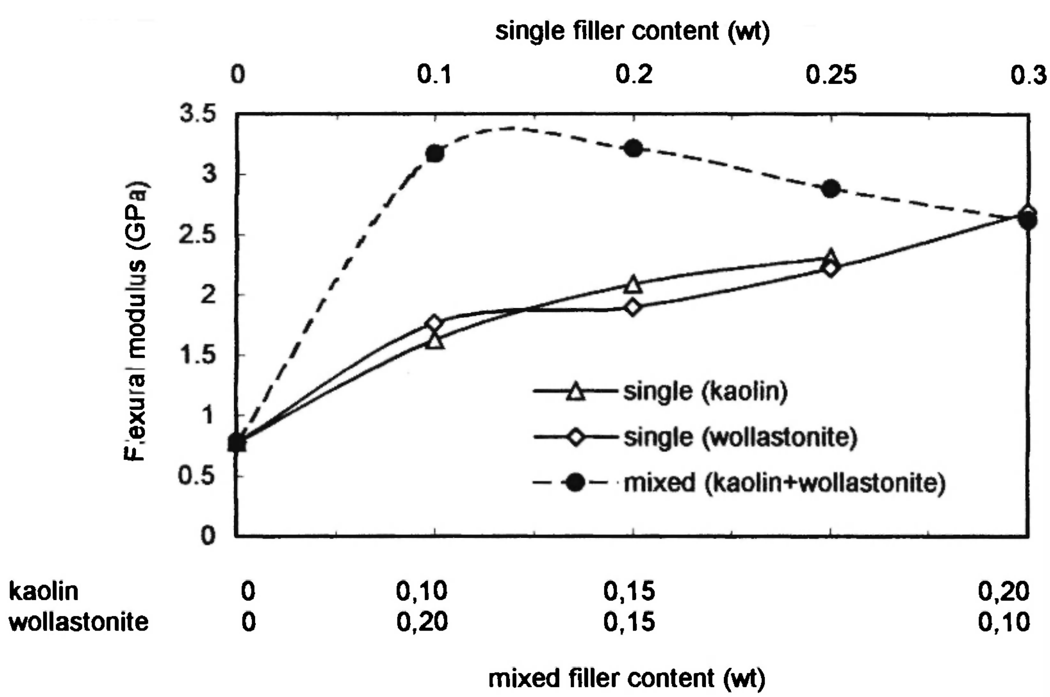

Fig. 6: Flexural modulus-filler content wt. curves for polyamide-6 composites

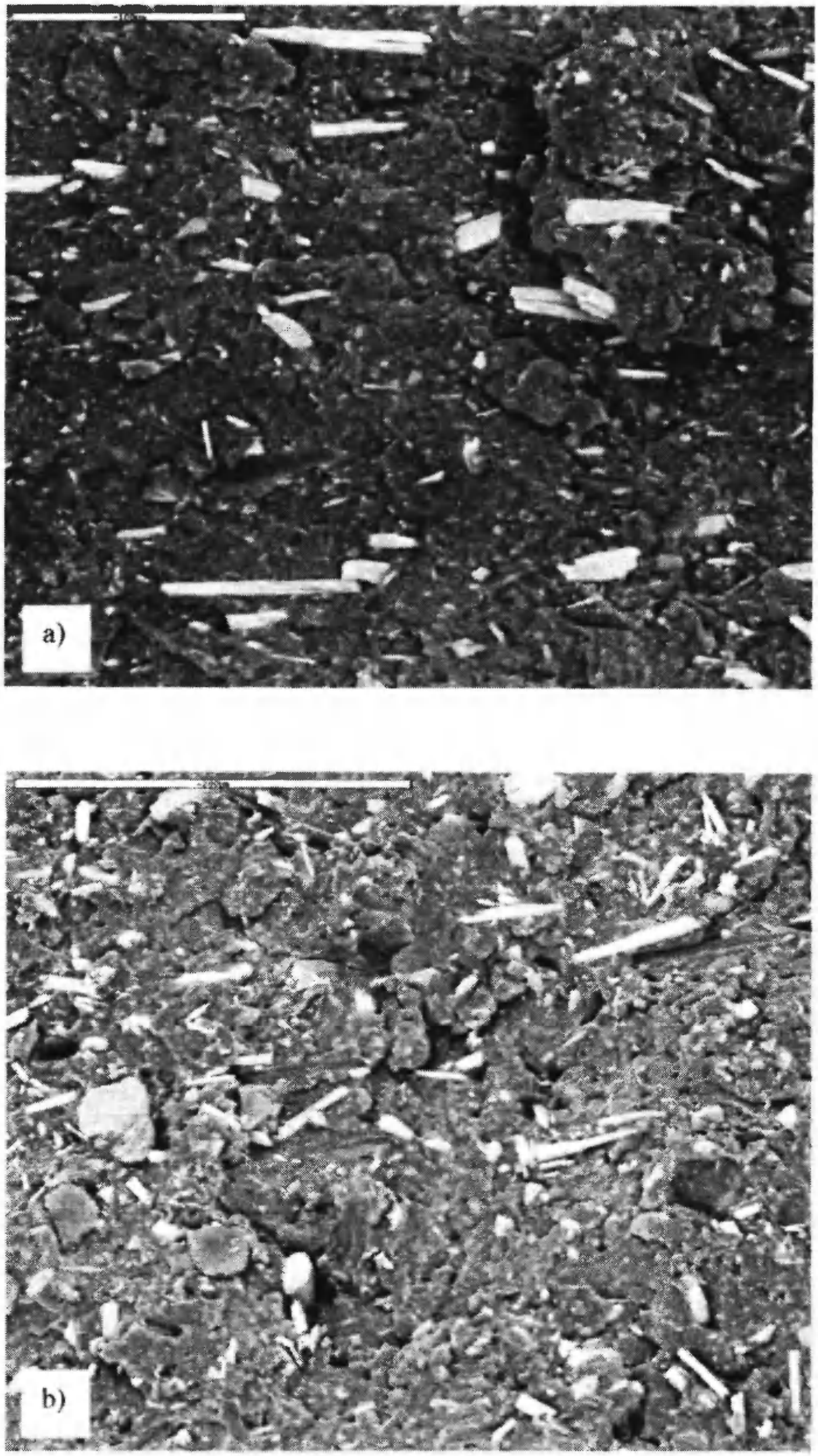

Fig. 7 a and b: Scanning electron micrographs of tensile fracture cross-section for the polyamide-6 with a) 10wt.%wollastonite+20wt.% kaolin b) 20wt.% wollastonite+10wt. %kaolin modulus of elasticity of the composites tend to increase with increasing kaolin and wollastonite content. It is apparent that the wollastonite and kaolin are favorable for improving tensile strength and modulus. Moreover, in terms of mechanical properties, the most suitable content of kaolin and wollastonite in polyamide-6 is 20wt.%. In addition izod impact tests indicated that the addition of wollastonite and kaolin fillers to polyamide-6 decreases the impact strength and ductility of the composite. Furthermore, except for notched izod impact strength values, the best improvement in properties of polyamide-6 was obtained either with 30wt.% wollastonite filler or with 15wt.% kaolin+15wt.% wollastonite mixed fillers. In all cases, elongation at break dramatically decreases for polyamide-6/kaolin and polyamide-6/wollastonite composites 10wt.% fillers above which, there is a little change in elongation. In the light of this study, mechanical properties of the composites can be improved using one or more fillers.

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