Durability Study of Recycled Glass-Fiber-Reinforced Polyamide 66 in a Service-Related Environment

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ABSTRACT: A study of the mechanical properties in an accelerated service-related environment of recycled glass-fiber-reinforced polyamide 66 is reported. Material reinforced with 30 wt % of short fibers was reground and remolded up to seven times. Thermal aging in air at 140°C for up to 3000 h and coolant aging at 110°C for up to 1000 h showed no significant differences in behavior pattern. In addition to mechanical testing, the fiber length measured directly and the matrix stability measured by differential scanning calorimetry (DSC) were used to determine the influence of process-induced degradation on the durability of recycled samples compared with that of virgin samples. The results indicate that fiber length controls the initial properties. The differences in tensile strength and modulus between recycled and virgin samples were similar within the examined times of aging and could be explained by process-induced fiber shortening. The onset of embrittlement during both aging conditions is revealed first in a decrease in tensile elongation at break. Because of a lower degree of fiber reinforcement, the elongation at break of recycled samples was always as good as that of virgin reference samples. However, increasing the number of molding operations up to four to five times resulted in a faster deterioration rate in elongation at break of recycled samples. Further processing had less effect on the deterioration rate. The oxidative stability of the matrix as determined by DSC decreased as a result of repeated processing. The results suggest that matrix stability is related to changes occurring in elongation at break during accelerated aging of samples remolded up to about four times. © 1997 John Wiley & Sons, Inc. J Appl Polym Sci **65:** 1631–1641, 1997

Key words: glass-fiber-reinforced polyamide 66; recycling; durability; accelerated thermal aging; coolant aging

INTRODUCTION

Considerable environmental concern has arisen over the generation and disposal of solid wastes. The necessity for reducing the solid waste stream going to landfills and the introduction of environ-

mental regulations related to plastics disposal are now major concerns in the automotive industry. Recycling of engineering plastics therefore represents an important area for reducing the waste stream going to landfills. Short-fiber-reinforced thermoplastics have been used in the automotive industry for many years. Glass-fiber-reinforced polyamide 66 is a commonly used material for automotive under-the-hood applications. The engine compartment is a harmful environment for plas-

tics, since the functional requirements include dimensional stability and resistance to high temperatures, impact, and chemicals.

The radiator end-cap is one of the earliest large under-the-hood components manufactured from heat-stabilized glass-fiber-reinforced polyamide 66. The radiator in automobiles transfers the heat of the coolant from the engine to the ambient air. During service, the material in a radiator end-cap is exposed to coolant in addition to high temperatures and mechanical loading. Thus, the functional requirements include resistance to hydrolysis at elevated temperatures. Normal operating temperature of the coolant ranges from 85 to 110°C but can peak over 130°C. The coolant usually consists of a 1 : 1 mixture of water and ethylene glycol, which is the most common antifreeze.

Owing to a lack of knowledge concerning the durability of recycled polyamides and the critical requirements regarding reliability, only small quantities are currently being recycled in-plant or mechanically for engineering applications. More polyamides could be recycled for use in engineering applications if we possessed a better understanding about their performance during service. Recycled plastics will find widespread acceptance as engineering materials only if they can guarantee the required performance. It is crucial, therefore, to establish methods to permit quantitative studies of process-induced and service-induced degradation.

Process-induced fiber shortening and the decomposition of heat stabilizers during the recycling operation may influence durability. Reprocessing decreases the strength because of process-induced fiber breakage.[1,2] It has also been shown that the thermal stability of the polyamide matrix, as measured by differential scanning calorimetry (DSC), decreases as a result of reprocessing.[3] These results may serve as reliable indicators of the durability of recycled items. Previous studies of the performance of polyamide 66 in an oxidative environment showed that the degradation exhibited a strong position dependence and occurred primarily in the surface region of the samples.[4,5] This surface degradation was shown to be responsible for the embrittlement of thermally aged samples. Furthermore, reprocessing showed a negative influence during subsequent thermal aging, since recycled samples indicated a higher rate of degradation, especially in the surface region.[4]

This article discusses studies that were carried out in service-related environments to investigate
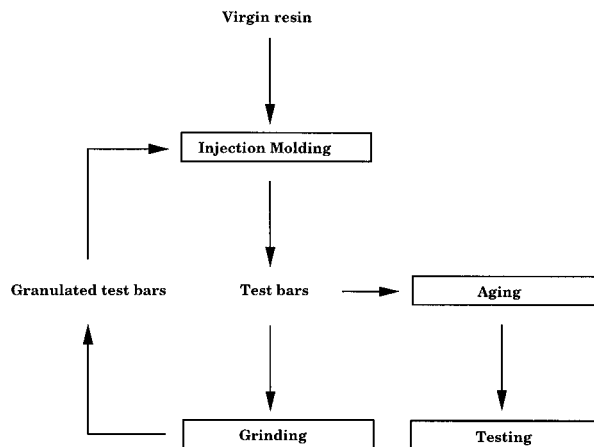

Figure 1 Description of the different operations and samples examined.

the durability of reprocessed glass-fiber-reinforced polyamide 66 compared with that of virgin material. To study the interaction between process-induced and service-related degradation, injection-molded test samples were exposed to accelerated test conditions to simulate long-term service life. The effects of thermal-oxidative degradation and coolant-induced hydrolysis were investigated. The mechanical performance was characterized by tensile tests, and process-induced fiber shortening was determined by fiber length measurements. The thermal stability of the polyamide matrix was characterized by DSC.

EXPERIMENTAL

The test samples were injection-molded from commercial heat-stabilized pigmented polyamide 66 (DuPont Zytel EFE 7185 BK99) reinforced with 30 wt % of short glass fibers. To study the influence of recycling, samples were obtained through grinding of moldings and repeated injection molding. Figure 1 gives a description of the different operations and samples examined. The samples were remolded up to seven times. Standard injection-molded test bars, with dimensions according to ISO 527 specifications, were supplied by Du-Pont.

Injection-molded test samples were aged at elevated temperatures as an accelerated test condition. The effects of both thermal-oxidative aging and coolant aging were investigated. The thermal aging was induced using a conventional air circulating oven. The samples were aged at

Table I Samples Examined

Sample Designation	Number of Injection Molding Operations	Thermal Aging Time (h)	Coolant Aging Time (h)
M1	1	0–3000	48–1000
M2	2	0–3000	48–1000
M3	3	—	48–1000
M4	4	0–3000	48–1000
M5	5	—	48–1000
M6	6	0–3000	48–1000
M7	7	—	48–1000
M8	8	0–3000	48–1000

a temperature of 140°C for up to 3000 h. The coolant aging was performed at 110°C from 48 h up to 1000 h in a 1 : 1 mixture of distilled water and the commercial glycol Glysantin G48-94 produced by BASF. The reference samples were aged 48 h. All examined samples are listed in Table I.

Tensile testing according to ISO 527 was carried out in air at 23°C and 50% relative humidity (RH). Before testing, the test bars were held in 50% RH at 80°C until equilibrium was reached. Five bars were tested in all cases.

Fiber lengths were determined using a microscope connected to an image analyzer system (Seescan Ltd.). A video camera was used to capture images of the samples. Fiber samples were prepared by burning off the matrix polymer. A preweighed sample of recovered fibers was dispersed in an aqueous solution of sodium diphosphate (0.2 g/l). The fiber suspension was then transferred to a Petri dish, which was placed in the microscope for measurement. The captured images were digitized by the image analyzer (256 gray levels) and displayed on a monochrome monitor with a resolution of 512×512 pixels. Fibers longer than 25 μm were studied and, for each sample, about 600 fibers were evaluated. The weight-average fiber length was used to characterize the samples.

Calorimetric analysis was carried out in a TA Instruments thermal analyzer equipped with a 910 DSC module. Starting at 50°C, dynamic measurements were carried out at a heating rate of 10°C/min to a temperature above the oxidation peak. The sample weight was 10 ± 0.2 mg, and the samples were cut from injection-molded test bars. During operation, the instrument was purged with a constant flow of oxygen of 4 l/h.

RESULTS AND DISCUSSION

Fiber Length

Extrusion compounding of chopped strands of fibers and polymer into pellets and subsequent injection molding result in fiber attrition.[6–9] The mechanical properties of items manufactured from glass-fiber-reinforced thermoplastics are strongly influenced by the length distribution of the fibers.[2,10,11] Fibers longer than a certain critical value, the critical fiber length, offer a higher contribution to the reinforcement.[12] The greatest improvements in strength and stiffness are obtained with high-volume fractions of long fibers aligned in the load direction.[13]

The weight-average fiber length was used to characterize the samples and is given by

$$l_w = \frac{\sum\limits_{i}^{n} n_i l_i^2}{\sum\limits_{i}^{n} n_i l_i} \tag{1}$$

where n_i is the number of fibers within a certain fiber-length range (in this case 0.025 mm) centered at l_i.

The weight-average fiber length for all studied samples is reported in Table II. The average fiber length decreases as the quantity of moldings is increased. This decrease is attributed to the cumulative breakage of fibers caused by mechanical working during the granulation process and subsequent molding operation. It can also be seen that fiber shortening dominates during the first injection molding cycles using 100% regrind.

Matrix Stability

The durability of a polymer product is often related to its resistance to oxidative degradation.

Calorimetric analysis has been widely used to study the oxidation of polymers. The oxidative stability can be evaluated in an oxygen atmosphere by dynamic measurements in which the temperature is increased at a constant rate.[3,14–19] The attraction of this dynamic method is its simplicity. During a DSC-run, the initial breakdown of stabilization protection and oxidation of the polymer are accompanied by an exothermic reaction, causing a deflection of the heat flow curve. This change in heat flow is a signature of a severe degradation of the polymer. The difficulty in studying oxidation by DSC of heat-stabilized polyamide 66 is that the exothermic reaction starts more or less immediately above the melting region. In unstabilized polyamide 66, the oxidation process already starts in the solid phase.[14]

Typical DSC dynamic scans of heat-stabilized polyamide 66 are shown in Figure 2. The heat flow of samples injection molded one (M1) and eight (M8) times is illustrated as a function of temperature. The endotherm that peaks at about 260°C for both samples is due to the crystalline melting. The onset of the oxidation, where the exothermic reaction causes an upward deflection of the heat flow curve, already commences in the melting region. The peak temperature of the exothermic reaction (oxidation peak temperature) was used to characterize the samples. When polyamide is tested in oxygen in a DSC analyzer, the oxidation peak temperature (OPT) decreases with lower matrix stability. The OPT was determined to be about 318 and 312°C for samples M1 and M8, respectively. The recycled sample M8 clearly exhibits a lower degree of oxidative stability, as shown by a lower oxidation peak value.

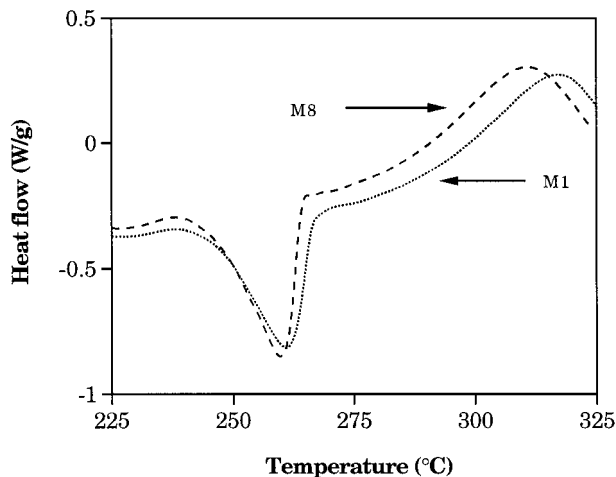The OPT for all samples is also reported in Ta-

Figure 2 Heat flow of samples M1 and M8 in an oxygen atmosphere as a function of temperature (heating rate 10°C/min).

ble II. As expected, those samples that have been molded from recycled material exhibit a lower degree of oxidative stability, since the OPT decreases with increasing numbers of repeated injection molding operations. Furthermore, the OPT shows a linear decrease with increasing numbers of molding operations for up to eight times.

To summarize, the thermal stability data indicate a decreased matrix stability as a result of recycling. This may be explained as being due to process-induced decomposition of heat stabilizers. If this is the case, these data may serve as reliable indicators regarding the durability of reprocessed items, since the results suggest that, unless restabilized, recycled material will experience higher degradation rates during subsequent service or during accelerated aging to simulate service conditions.

Thermal-Oxidative Aging

It has been shown by several studies that the exposure of polyamides to both heat and air causes changes in physical, chemical, and mechanical properties.[4,5,20–25] The influence of thermal aging at 140°C on tensile properties of samples M1, M2, M4, M6, and M8 is illustrated in Figure 3(a–c). The tensile strength [Fig. 3(a)] and modulus [Fig. 3(b)] of unaged samples decrease with increasing numbers of recycling operations. These results can be explained by a lower degree of reinforcement imposed by shorter fibers (Table II). The tensile strength
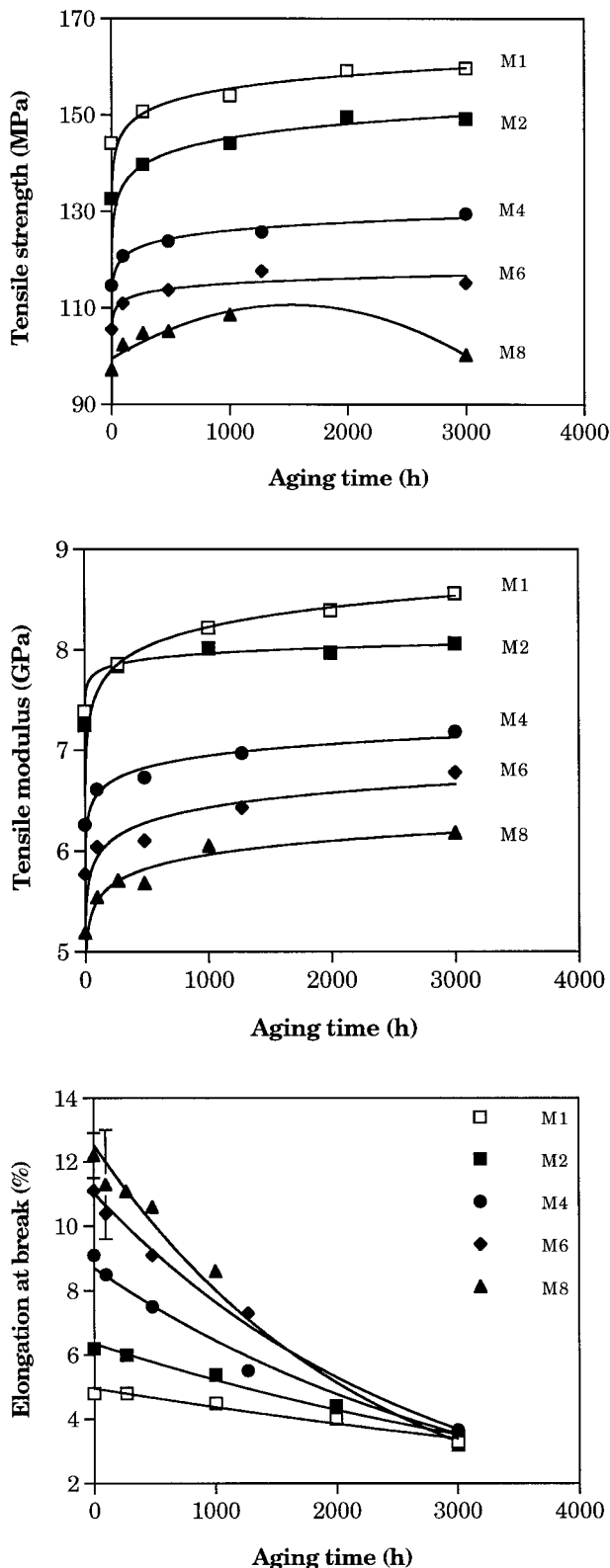
Table II   Weight Average Fiber Length and Oxidation Peak Temperature for Samples M1–M8

Samples	Weight Average Fiber Length (mm)	Oxidation Peak Temperature (°C)
M1	0.341	317.7
M2	0.276	316.1
M3	0.240	315.8[a]
M4	0.220	315.2
M5	0.204	314.3[a]
M6	0.187	313.9
M7	0.168	312.7[a]
M8	0.150	311.7

[a] Calculated using linear interpolation.

and modulus increase as a function of aging time. Harding and MacNulty[5] showed that the tensile strength of unreinforced material starts to fall drastically when the molecular weight of the material in the surface of the samples drops below a critical value. In our study, a slight drop was only observed for sample M8 after 3000 h of aging. The initial stages of the aging had a more pronounced influence on the rate of change of the properties. Note also that the relative difference between the samples seems to be more or less constant.

The elongation at break [Fig. 3(c)] of unaged samples shows that recycling results in increased values. This can be explained by a lessening of the restriction imposed by the shortened fibers on the deformation of the matrix during mechanical loading. During aging, the elongation at break decreases as a function of aging time. The tensile elongation shows a faster drop with increasing numbers of recycling operations.

The increased tensile strength and modulus during thermal aging may be explained by an increased molecular weight induced through polycondensation in the bulk of the samples.[5,25] The decreased elongation at break confirms that exposure to heat and air induces embrittlement. The overall embrittlement of thermally aged polyamide 66 has been shown to be related to oxidative degradation in the surface region of the samples.[4,5] Harding and MacNulty[5] showed that the molecular weight of samples from the surface region of aged specimens decreases considerably through oxidative chain scission. Interestingly, after 3000 h of thermal aging all samples exhibited similar values of elongation at break. This observation indicates that the degraded matrix in the surface region of the samples completely controls elongation at break regardless of recycling and fiber length.

In summary, the relative differences in tensile strength and modulus between recycled and virgin samples were similar within the whole period of aging. These results indicate, therefore, that process-induced fiber breakage controls the differences in these properties. The onset of embrittlement is revealed in a significant drop in tensile elongation at break. Reprocessed samples deteriorate at a higher rate compared with samples containing virgin material, and their degradation is more intense. However, within examined times of aging, the recycled samples were never worse than the virgin reference samples.

Figure 3   Influence of thermal aging at 140°C on (a) tensile strength, (b) tensile modulus, and (c) tensile elongation at break.

Coolant Aging

The performance in a hydrolytic environment was investigated by coolant aging at 110°C for 1000 h in a 1 : 1 mixture of distilled water and glycol. Durability tests on polyamide grades for radiator end-cap purposes often include these aging conditions. The automobile industry requirements mostly call for less than a 50% drop in mechanical properties after accelerated coolant aging. The aging in our study was performed for 48, 210, 500, 700, and 1000 h.

It is known that polyamides hydrolyze in the presence of water at high temperatures, which is manifested by a decrease in molecular weight.[5,26] The coolant usually consists of a 1 : 1 mixture of water and ethylene glycol. The latter may have some plasticizing action on polyamide 66. In general, polyamides are more resistant to bases than to acids. It has been shown that injection-molded bars of polyamide 66 suffer a loss in performance after exposure for 1 year at 70°C to a pH 4 or pH 10, but not pH 7, solution.[27] This suggests that both acid and base catalysis can cause degradation. The pH of the coolant in the present work was measured at all investigated aging times and was found to be 8.0 ± 0.1 during the whole period of aging. Before tensile tests, the coolant-saturated test bars were conditioned in 50% RH at 80°C until equilibrium was reached.

The tensile strength, modulus, and elongation at break of samples M1–M8 as a function of aging time are illustrated in Figure 4(a–c). As can be seen, fiber length characteristics (Table II) control the initial properties. The tensile strength [Fig. 4(a)] and modulus [Fig. 4(b)] were essentially unchanged after 1000 h, relative to 48 h of aging. The strength exhibited a small decrease of 4–5%. This decrease may be explained by hydrolysis of the matrix as well as of the fiber–matrix interface. The tensile modulus increased about 3% during aging. This influence on modulus suggests that the performance of the fiber–matrix interface is not affected in a negative way by hydrolysis under these aging conditions. This is not surprising because the fiber-reinforced polyamide 66 grades that are used for radiator end-caps have an interface that is stabilized against hydrolysis.[28] The hydrolysis resistance is mostly obtained by the strength of the interfacial bonding between the glass fibers and the polyamide matrix. Furthermore, the relative differences in tensile strength and modulus between the samples re-
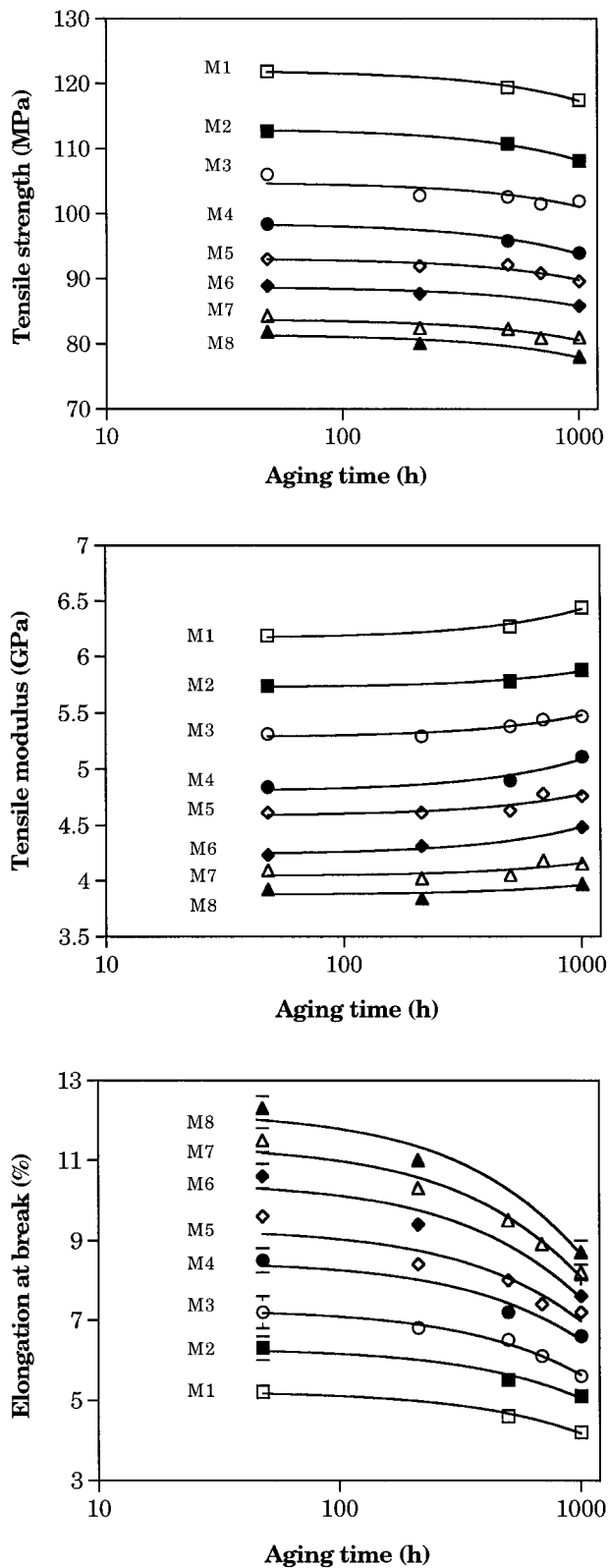

Figure 4 Influence of coolant aging at 110°C on (a) tensile strength, (b) tensile modulus, and (c) tensile elongation at break.

main more or less constant during the examined period of aging.

The elongation at break [Fig. 4(c)] is negatively influenced by coolant aging. The elongation showed a 20 to 30% decrease for samples M1 to M8. These results are similar to those found for aging in water at 71 and 120°C for 1000 h or exposure to steam at 120°C.[29] There was essentially no change in tensile strength but about a 50% drop in elongation at break.

During coolant aging, the results were similar to those of aging in air at an elevated temperature in that the elongation at break decreased as a function of aging time. The tensile strength displayed a small decrease, but this is significantly less pronounced compared with that of elongation at break. The induced embrittlement indicates that the coolant does hydrolyze the polyamide. Even though the recycled samples were always at least as high, considering elongation at break, as the virgin reference samples, increasing the number of molding operations resulted in a more intense drop in elongation at break. This indicates a lower matrix stability of reprocessed samples towards hydrolysis. The relative differences in tensile strength and modulus between recycled and virgin samples show the same behavior as during thermal aging in air, indicating that fiber length controls these differences.

Durability of Recycled Glass-Fiber-Reinforced Polyamide 66

The purpose of our study was to investigate durability in a service-related environment of recycled glass-fiber-reinforced polyamide 66 compared with that of virgin material. The derived results indicate the following:

1. Tensile properties of unaged samples are strongly related to the fiber length characteristics. This is also illustrated in Figure 5(a,b).
2. Thermal stability, as measured by DSC analysis of the matrix polyamide, decreases as a result of increasing numbers of reprocessing and molding operations. These measurements suggest that recycled material will experience higher degradation rates during subsequent service or during accelerated test conditions.
3. Since thermal aging in air at 140°C for up to 3000 h and coolant aging at 110°C for up to 1000 h showed no significant differences
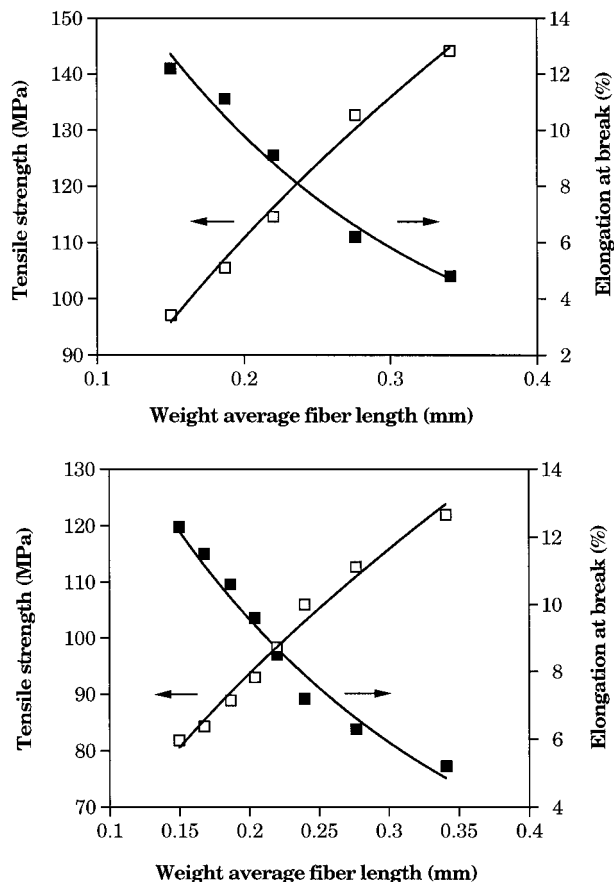

Figure 5 Tensile strength and elongation at break of conditioned reference samples as a function of weight-average fiber length: (a) unaged samples conditioned at 50% RH; (b) samples aged in coolant for 48 h prior to conditioning at 50% RH.

in behavior pattern and the relative differences in tensile strength and modulus between recycled and virgin samples were similar within the examined times of aging, it appears that process-induced fiber shortening controls the differences in these properties.
4. The onset of embrittlement during thermal aging and coolant aging is revealed in a drastic drop in tensile elongation at break.

Changes in elongation at break with aging time (i.e., the degradation rate) are expected to be influenced by the initial stability of the matrix. Furthermore, recycled materials are expected to experience higher deterioration rates during subsequent service or accelerated aging. What follows is an attempt to correlate the changes in elongation at break during aging with the OPT mea-

sured by DSC. The induced embrittlement during both thermal-oxidative aging and coolant aging was investigated. The time to a certain drop in elongation at break relative to unaged samples was used to characterize the deterioration rate during aging. Figure 6 shows the times to a 30% drop (thermal aging) and a 20% drop (coolant aging) as a function of numbers of molding operations. As can be seen, the time to a 30% drop during thermal aging at 140°C is strongly influenced by the numbers of moldings for up to about 4 operations. As expected, the degradation rate increases as a result of recycling. However, further processing had less effect on the deterioration rate, which more or less is unchanged after the fifth molding. The time to a 20% drop during coolant aging shows almost a constant value for up to two molding operations. Between two and five moldings, however, the results indicate a significantly increased degradation rate as a function of recycling. As for thermally aged samples, further processing after five molding operation has only a slight effect on degradation rate.

As mentioned earlier, the deterioration rate, or the time to a certain drop in mechanical properties, is expected to be related to the OPT, as measured by DSC. The times to a 30% drop (thermal aging) and a 20% drop (coolant aging) in elongation at break relative to unaged samples are shown in Figure 7 as a function of the OPT values reported in Table II. As can be seen, the deterioration rate under these aging conditions is only related to the initial stability of the matrix polyamide of samples molded for up to 4–5 times, or for samples with OPT values above 314–315°C. The
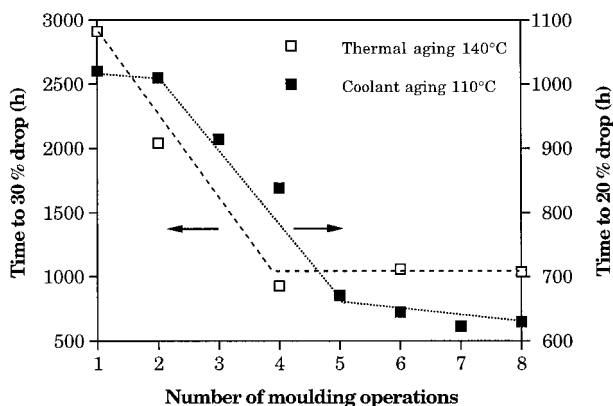

Figure 7 Times to a 30% drop (thermal aging) and a 20% drop (coolant aging) in elongation at break relative reference samples as a function of OPT measured by DSC.

results also suggest that the matrix stability has a maximum efficiency, concerning the drop in elongation at break, for samples molded one and two times prior to coolant aging at 110°C. The steady-state region occurring for samples with an OPT below 314°C suggests that the matrix stability is of an insufficient efficiency or that it has no influence on the deterioration rates during aging.

Our study has demonstrated that the behavior of glass-fiber-reinforced polyamide 66 is strongly related to the fiber length during both thermal aging and coolant aging. The fiber length controls not only all initial tensile properties but the relative differences in tensile strength and modulus between recycled and virgin samples during accelerated aging as well. The OPT determined by DSC showed a correlation with the rate of decline in elongation at break of samples injection molded for up to about 5 times.

It should be stressed that degradation that takes place during service of radiator end-caps may not simply be related to hydrolysis or thermal oxidative degradation but rather to the combination of hot coolant and air. Furthermore, fresh water is a more harmful environment than stagnant water because of the combined hydrolytic and oxidative degradation effects. It has been reported that the estimated service life in hot water rich in oxygen is 50–70% of that in stagnant water.[26]

Figure 6 Times to a 30% drop (thermal aging) and a 20% drop (coolant aging) in elongation at break relative reference samples as a function of numbers of injection molding operations.
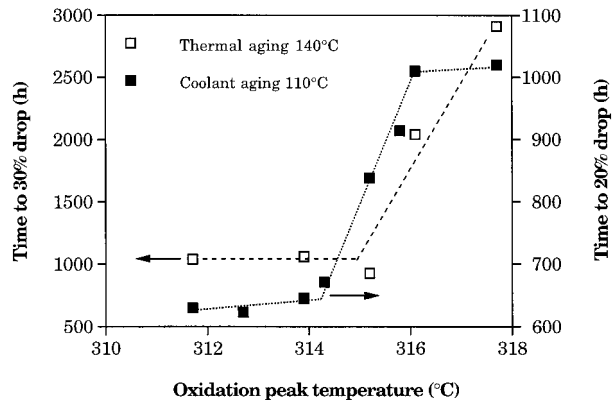
Durability as a Function of Regrind Ratio

Table III shows the composition of material injection molded different numbers of times using dif-

Table III Composition of Material Molded Different Numbers of Times for Different Levels of Regrind in a Continuous Recycling Operation

Regrind Ratio	Number of Times Molded										
	1	2	3	4	5	6	7	8	9	10	> 10
None	100	—	—	—	—	—	—	—	—	—	—
0.10	90	9	1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
0.20	80	16	3	1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
0.30	70	21	6	2	1	< 1	< 1	< 1	< 1	< 1	< 1
0.40	60	24	10	4	2	1	< 1	< 1	< 1	< 1	< 1
0.50	50	25	13	6	3	2	1	< 1	< 1	< 1	< 1
0.60	40	24	14	9	5	3	2	1	1	< 1	1
0.70	30	21	15	10	7	5	4	2	2	1	3
0.80	20	16	13	10	8	7	5	4	3	3	11
0.90	10	9	8	7	7	6	5	5	4	4	35
1.00	—	—	—	—	—	—	—	—	—	—	100

ferent regrind ratios in a continuous recycling operation. To study steady-state property values when materials are continuously reprocessed at levels lower than 100% recycled material, algorithms can be used. In the case of a 10% regrind level, for example, 90% of the material has been injection molded once, 9% twice, 1% three times, and < 1% more than three times.

The measured tensile strength and elongation at break shown for samples M1–M8 in Figure 3(a,c) and Figure 4(a,c) were used to calculate values for materials continuously recycled at different regrind ratios. This was done by using the proportions of material molded different numbers of times reported in Table III. Thus, for example, the expected tensile strength at a regrind ratio of 0.1 for samples aged in coolant for 1000 h will be $(0.9 \times 117.5) + (0.09 \times 108.9) + (0.01 \times 101.9) = 116.6$ MPa. Similar calculations were performed for samples thermally aged at 140°C for 3000 h and for conditioned reference samples. All calculations were based on the assumption that the values of different levels of recycled material vary linearly according to the "rule of mixtures". Linear interpolation has been shown to be valid for blends of virgin 30 wt % glass-fiber-reinforced polyamide 66 and recyclate made from the same material.[30] The calculated values are shown in Figure 8(a,b) during thermal-oxidative aging at 140°C [Fig. 8(a)] and coolant aging at 110°C [Fig. 8(b)] for regrind ratios up to 0.5.

The results of conditioned reference samples (unfilled symbols) and aged samples (filled symbols) indicate that during both aging conditions, the elongation at break of samples based on differ-

ent regrind ratios up to 0.5 never drops below the values of a sample containing virgin material (regrind ratio = 0). Thus, only considering elongation at break, the results suggest that samples based on regrind fulfill the requirements of virgin material.

Because of a lower degree of fiber reinforcement, the tensile strength decreases for increasing levels of recycled material. At a regrind ratio of 0.3, the tensile strength is generally decreased by 3% for both unaged and aged samples. This drop is within design limits. As the regrind level increases, the influence of recycling becomes more pronounced and, at a regrind ratio of 0.5, the decay in tensile strength is about 7%.

CONCLUSIONS

This article has discussed studies conducted in service-related environments to investigate durability of recycled glass-fiber-reinforced polyamide 66 compared with that of virgin material. Material reinforced with 30 wt % of short glass fibers was reground and remolded up to seven times. To study the interaction between process-induced and service-related degradation, injection-molded test samples were exposed to accelerated thermal aging and coolant aging. The results indicate the following:

1. Tensile properties of unaged samples are strongly related to the fiber length.
2. Thermal stability as measured by DSC analysis of the matrix polyamide decreases as
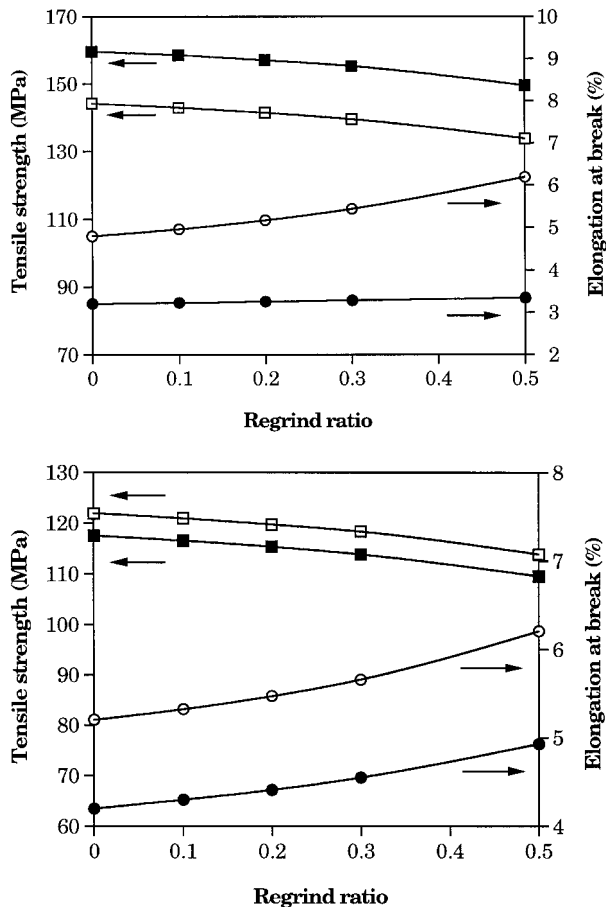
Figure 8 The tensile strength and elongation at break of unaged reference samples (unfilled symbols) and aged samples (filled symbols) as a function of regrind level: (a) thermal aging at 140°C; (b) coolant aging at 110°C.

a result of increasing numbers of repeating molding operations.

3. Thermal aging in air at 140°C for up to 3000 h and coolant aging at 110°C for up to 1000 h will result in no significant differences in behavior pattern, since the relative differences in tensile strength and modulus between recycled and virgin samples were similar within the examined times of aging. These results indicate that process-induced fiber shortening controls the differences in these properties.

4. The onset of embrittlement during thermal aging and coolant aging is revealed in a drastic drop in tensile elongation at break. Even though the recycled samples were always as high as the virgin reference samples, increasing the number of molding oper-

ations resulted in a more intense deterioration in elongation at break.

The oxidation peak temperature measured by DSC was used to correlate the influence of process-induced degradation on the properties obtained during accelerated aging. DSC data showed a correlation with the rate of decline of elongation at break for samples remolded up to about 4 times. Further injection molding had less effect on the rate of decline.

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