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Mixing of glass fibers with nylon 6,6

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Nylon 6,6 is one of the toughest of the engineering thermoplastic resins. It is resistant to corrosion and chemicals, but has a limited capability due to low rigidity, strength and moisture adsorption. Glass fibers are very strong and rigid but susceptible to environmental attack. Proper mixing of these two materials would form a fiber composite with high strength, toughness, rigidity and stability at elevated temperatures. The main purpose of this work was to study the effect of properties of a fiber composite containing 20% loading of glass fibers in Nylon 6,6. Two different types of twin screw extruders, one a co-rotating and the other a counter-rotating, were used. Two different screw designs, a high-shear and a low-shear design, were used on each of these extruders. A statistical process study was developed using ECHIP. RPM of the screw and the output rate of the extruder were the identified variables in the process. Molded samples were evaluated for tensile, flexural, impact and heat deflection characteristics. Scanning Electron Microscopy study was also performed to evaluate the fiber distribution, length and wetting characteristics. The results were analyzed for all of the above properties and it was concluded that there was a great improvement in the properties of the reinforced material. Also, it was found that low RPM and output rates on the co-rotating twin-screw extruder would result in the best properties.

1. Introduction

Reinforced plastics have achieved popularity in a short time due to design flexibility, corrosion resistance, high strength, dimensional stability, light weight, low tooling and finishing costs. Nylons that are toughened are known commercially as engineering polymers. Glass reinforced Nylons have become available in recent years and are used in gears, automotive industry and a variety of domestic appliances. Compared to unfilled Nylons, glass fiber reinforcement leads to a substantial increase in the tensile strength (160 MPa versus 80 MPa), flexural modulus (8000 MPa versus 3000 MPa), hardness, creep resistance and heat distortion temperature under load (245 °C versus 75 °C under 1820 MPa). The glass fiber filled reinforcement is obtained by passing chopped glass fibers through a polymer melt to produce Nylon strands that are chopped into pellets. In this study, Nylon 6,6 is loaded with 20 wt % glass fibers throughout all the runs. The pellets are then processed through an injection molder for the test bars that were tested for their properties.

This project presents a comprehensive study on the improvement of properties using high and low-shear screw designs on co-rotating and counter-rotating twin screw extruders. The techniques used were similar to those of previous studies [1]. Nylon 6,6 shows a marked improvement in its properties after reinforcement with chopped glass fibers.

2. Experimental design process

A statistical process design was developed using ECHIP™, taking into account the process variables. The identified variables in the above process were the RPM of the screw and the output rate of the extruder. ECHIP™ is a package that provides programs to perform statistically designed experiments. The effects of variables on the response were identified with a minimum number of experiments. The major benefits using this package are fewer experiments, predictable relationships and increased efficiency.

A matrix was developed using high and low values for RPM of the screw and the output rate of the extruder and the number of runs were optimized on each of the screw designs to 8 runs. This included repetition of the runs to minimize the experimental error.

3. Experimental details

Owings Corning® 492 AA glass fibers and Nylon 6,6 Zytel® 101 were compounded together at weight ratios of 20/80 in a Werner Pfleiderer® ZSK-30 co-rotating twin-screw extruder and in an American Leistritz® 30 mm counter-rotating twin-screw extruder. Before processing Nylon 6,6 was dried for better mixing with the glass fibers.

Two different screw designs, a low-shear and a high-shear design, were used on each of the extruders to get the extruded pellets. The screws consisted of kneading blocks, mixing elements and conveying elements. These screws were enclosed in barrels and the whole unit was utilized for transportation of the polymer melt to the outlet. The configuration of the elements could be altered to produce the desired effect.

The low-shear screw on the ZSK-30 was 878 mm long. It had the main feed at the barrel 1 (42 mm), the glass feed was at barrel 6 (504 mm) and the vacuum port was at barrel 8 (704 mm). But the high-shear screw design on the ZSK-30, 1332 mm long, had more kneading blocks than the low-shear screw. The main feed was at barrel 1 (42 mm), the glass feed was at barrel 10 (869 mm) and the vacuum port was at barrel 12 (1044 mm).

The low-shear screw on the Leistritz® 30 mm counter-rotating extruder was 840 mm long, the main feed was at the zone 1 (30 mm), the glass feed was at zone 3 (367 mm) and the vacuum vent was at zone 5 (622 mm). The high-shear screw was also 840 mm, the main feed was before zone 1 (0 mm), the glass feed was at zone 3 (480 mm) and the vacuum vent was between zone 5 and zone 6. The difference between the high and the low-shear screws lies in the configuration of the elements of the screw.

The extruded pellets were dried for 6 h before injection molding. They were molded into ASTM D638 Type 1 bars and 1/8 in. bars. The barrel temperature in the injection molder was maintained at around 266 to 276 °C for improved dispersion of glass fibers. The injection-hold-pressure was also maintained at 4137 MPa to allow the material to completely fill the mold. Tensile strength and flexural modulus were obtained using an Instron Model 6302 Hydraulic Testing machine according to ASTM D638 and ASTM D790 respectively. Five specimens of each of the different compositions were tested. The Heat Distortion Test (HDT) was performed on the two specimens each and the temperatures were noted. The 1/8 in. bars were notched with a 45° 'V' notch that has an apex with a curvature of 0.25 mm and the Notched Izod Impact test was performed on 5 specimens each according to ASTM D256, Method A. The extrudate was collected as a 200 g blob and fractured using liquid nitrogen. The fractured samples were mounted on an Amray 1000A Scanning Electron Microscope (SEM). Fiber wetting and adhesion characteristics were analyzed using three different magnification ranges. Quantitative microscopy techniques using the Quantimet 570 Image Analyzer were used to determine the length distribution of the fibers in a composite. The blob was burnt in an oven and the resulting ash was used for the above purpose.

As in many designed experiments, some of the variables had to be modified for smooth running of the study as the equipment could not be operated with the theoretically intended values. However, no appreciable error was generated as a result of the modifications. For example, the RPM had to be raised by 25% and the Feed/Output rate had to be reduced by 25% on the runs 6 and 7 with the low-shear screw design on the ZSK-30 as the extruder was over torquing at the stipulated rates.

4. Results

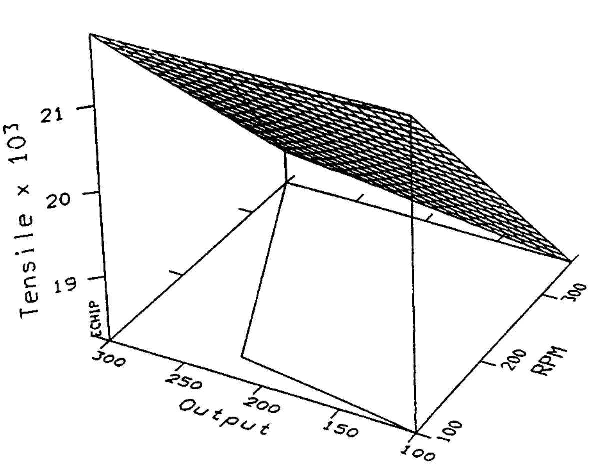
The samples were tested for physical properties and the SEM photographs and Image Analyzer data were inputted into the ECHIP™ program. A statistical table was generated which was then graphically analyzed. The results were generated for the following combinations: first, the best properties attained within a particular screw design on an extruder, second, the comparison of properties between different screw designs on the same extruder. Finally, the comparison of the properties on both the ZSK-30 and Leistritz® extruders with different screw designs were evaluated.

Table I shows the results of all the runs on a low-shear screw design on the ZSK-30 co-rotating extruder. High and low values of 350 and 100 for the RPM and 300 g/min and 100 g/min for the output rate were maintained throughout the experiment in order to comply with the requirements of the ECHIP™ program. The bars made by molding the reinforced material were tested for tensile, flexural, notched izod strength and heat deflection properties.

From Table I it can be seen that for low RPM, high values of tensile and flexural strength are observed. The HDT and the notched izod test also give very high values for low RPM when compared to high RPM rates. Output rate does not seem to affect the strength of material appreciably as for both low and high output rates; high values of tensile, flexural and notched izod properties are seen, though for low RPM and output rates, the best properties are clearly observed.

TABLE I Results from the low-shear-screw design on the ZSK-30

Run no.	RPM	Output (g/min)	Tensile (psi)	Flexural (psi)	HDT (°C)	Notched izod (ft lb/in)	MFL (μm)
1.	350	100	15730	25150	245.5	—	220
2.	100	100	21270	35780	251.5	1.393	218.8
3.	350	300	19600	31710	243	1.05	244.15
4.	100	100	22390	35440	250	1.531	260.8
5.	350	300	17610	29710	228.5	0.861	195.8
6.	125	225	21090	34290	244.5	1.353	251.5
7.	125	225	21890	35400	245.5	1.365	291.44
8.	350	100	20690	34780	246	1.36	227.17



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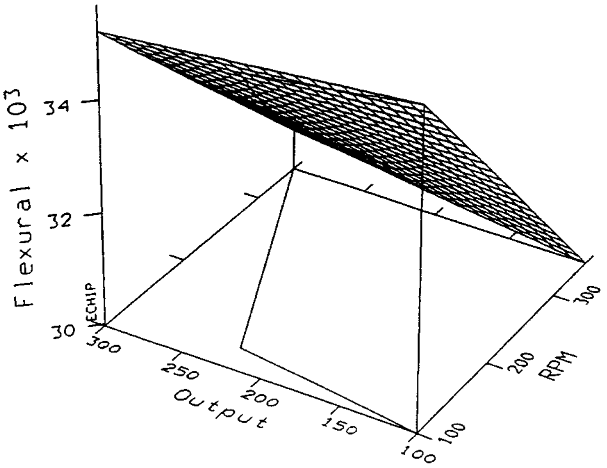

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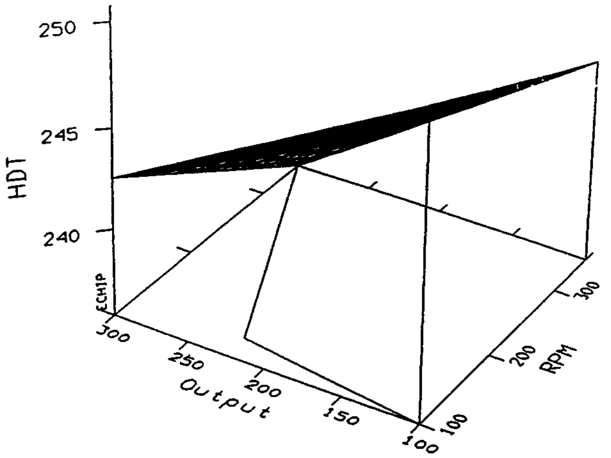

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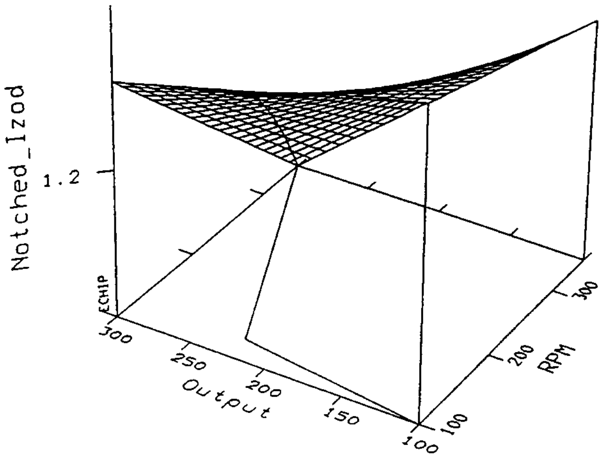

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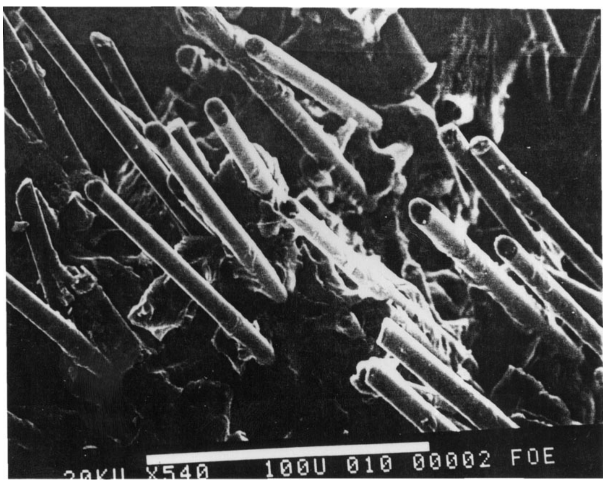

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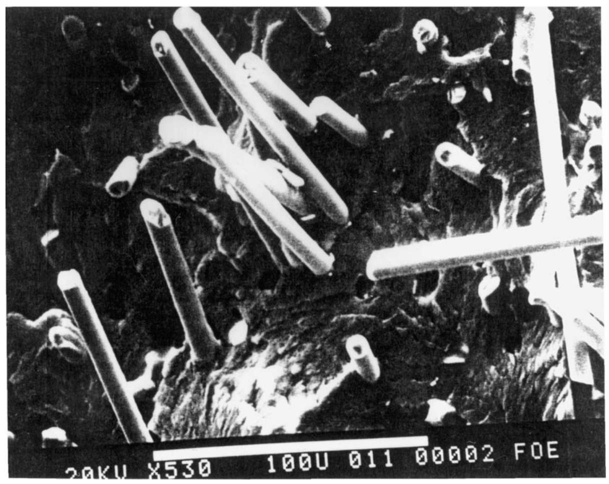


(f)

Figure 1 (a) Tensile properties on the ZSK-30 with low shear screw. (b) Flexural properties on the ZSK-30 with low shear screw. (c) HDT variation on the ZSK-30 with low shear screw. (d) Notched izod variations on the ZSK-30 with low shear screw. (e) SEM picture at low RPM and low output rate-good wetting. (f) SEM picture at high RPM and high output rate-bad wetting.

Three-dimensional graphs were generated for the above results. Fig. 1a shows the variation of tensile strength with the RPM and output rates of the extruder. It can be inferred from the graph that at low RPM rates and for both low and high output rates, values of over 21,000 psi (144,795 MPa) are observed and as the RPM increases, the tensile strength drops below 20,000 psi (137,900 MPa).

Fig. 1b shows the flexural strength variation with RPM and output rate. It can be seen that at low RPMs of the screw flexural strengths of over 234,422 MPa are obtained and as the RPM increases the flexural strength decreases much below the values obtained with low RPM rates. Even here output rates do not seem to affect the strength of the material. Fig. 1c shows the Heat Deflection Test temperature variations with RPM

TABLE II Results from the high-shear-screw design on the ZSK-30

Run no.	RPM	Output (g/min)	Tensile (psi)	Flexural (psi)	HDT (°C)	Notched izod (ft lb/in)	MFL (μm)
1.	350	100	16980	30200	240	1.27	300.2
2.	100	100	17110	33210	249	1.464	307.4
3.	350	300	20220	33210	244.5	1.317	331.5
4.	100	100	21130	35820	247	1.532	301.1
5.	350	300	20700	34720	251.5	1.283	276
6.	100	300	22120	36310	253	1.568	255.2
7.	125	225	21380	35550	234	1.406	265.5
8.	350	100	19600	34360	236	1.268	288

and output rates. It can be seen that high temperatures of heat distortion are obtained at low RPM rates of the extruder. But at low RPM and high output rates the HDT temperatures obtained were comparatively lesser than those with high RPM and low output rates. Fig. 1d shows the notched izod impact variations with RPM and output rates. It can be inferred from the graph that at low RPMs the best impact properties are obtained and output rate does not seem to affect the izod impact strength of the material.

It can also be seen from Table I that the mean fiber length (MFL) does not seem to appreciably affect the properties with the low-shear screw design.

The scanning electron microscope pictures taken from the blobs provided further insight into the fiber wetting characteristics. The scanning electron micrographs show the wetting and fiber distribution characteristics.

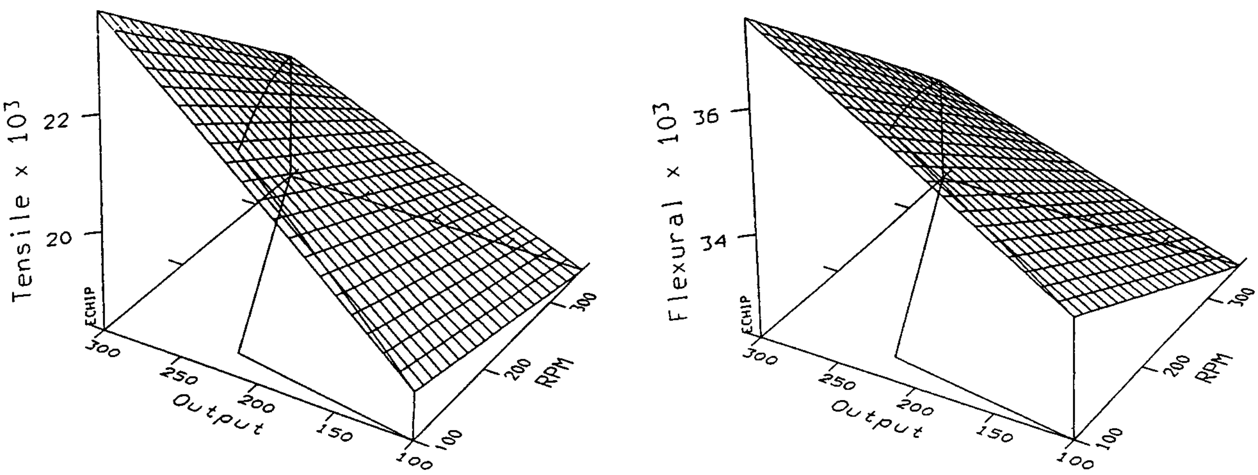
Fig. 1e and f show the wetting characteristics where the glass fibers can be seen in the Nylon polymer melt. The results suggest good wetting between the polymer and the glass fibers. Fig. 1e shows good wetting characteristics and uniform distribution of glass fibers in the extruded material at low RPM and low output rates. Fig. 1f shows undesirable wetting in the glass fibers at high RPM and high output rates. Both of these micrographs are for the low-shear screw design on the ZSK-30. Good wetting and distribution suggests improved toughness for the material and the same fact is illustrated through the properties in Table I.

Table II tabulates the results of all the runs on a high-shear screw design on the ZSK-30 co-rotating extruder.

Fig. 2a shows the variation of tensile strength with RPM and output rates of the extruder with the high-shear screw. It can be observed from the graph that the material with the maximum tensile strength is produced at low RPMs of the screw and at high output rates. Low RPMs and low output rates did not result in producing acceptable tensile properties. Fig. 2b shows the flexural strength variation with RPM and output rates of the extruder. It can be seen from this graph that low RPM and low output rates can produce the best flexural strengths. At low RPMs and low output rates, flexural strengths of over 248,220 MPa were obtained where as low RPMs and low output rates produced flexural strengths of less than 234,422 MPa.

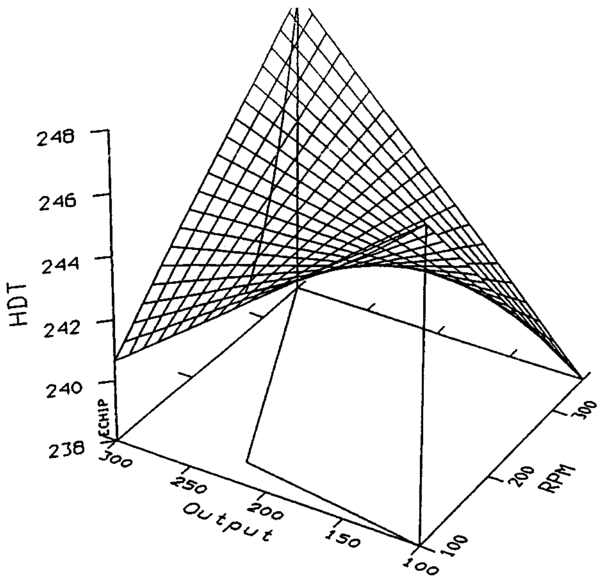
Fig. 2c shows that at low RPMs and low output rates, one can obtain a material with high Heat Deflection Temperatures. In Fig. 2d, low RPMs and output rates as well as low RPMs and high output rates give the best notched izod impact properties. Fig. 2e, f and g, the scanning electron pictures, illustrate the glass fiber wetting and uniform distribution characteristics.

Fig. 2e shows good wetting and uniform distribution of the glass fibers on the ZSK-30 with a high-shear screw design. This run was at a low RPM and output



(a) (b)

Figure 2 (a) Tensile properties on the ZSK-30 with high shear screw. (b) Flexural properties on the ZSK-30 with high shear screw. (c) HDT variation on the ZSK-30 with high shear screw. (d) Notched izod variations on the ZSK-30 with high shear screw. (e) SEM picture at low RPM and low output rate-good wetting. (f) SEM picture at low RPM and high output rate-good wetting. (g) SEM picture at high RPM and high output rate-bad wetting. (*Continued*)



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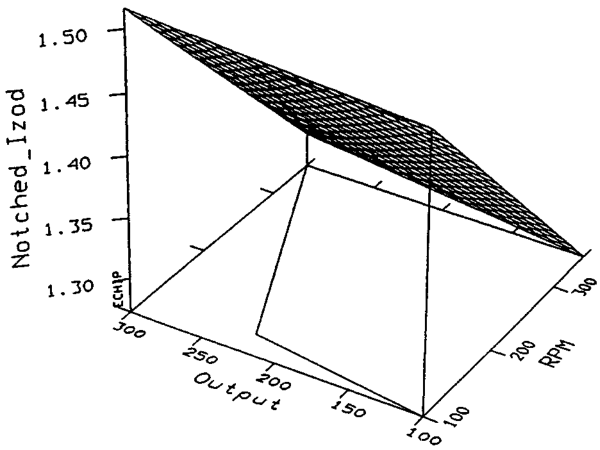

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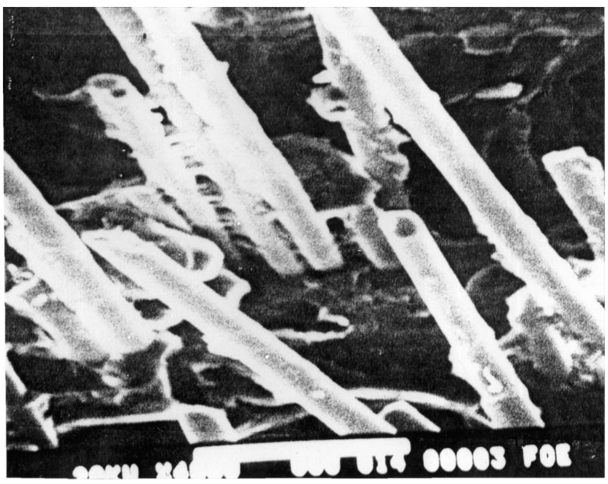

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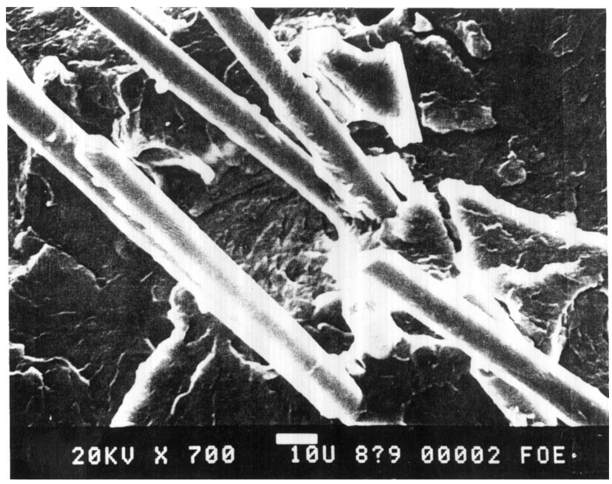

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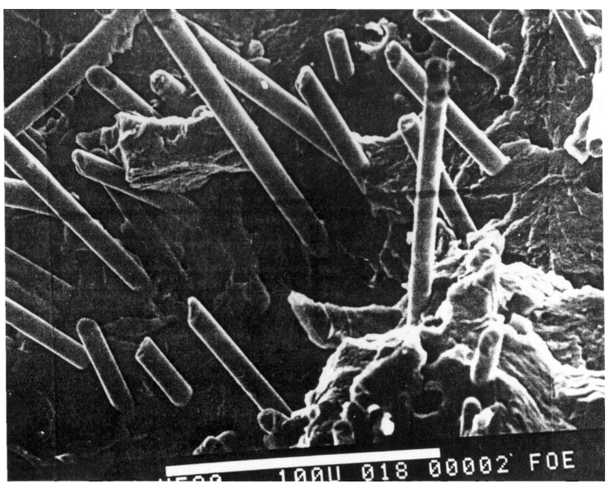

(g)

Figure 2 (*Continued*).

rate. At low RPMs and high output rates, good wetting is observed as shown in Fig. 2f. Observations from Fig. 2e and f might suggest that the output rate of the extruder does not affect the properties of the reinforced material as significantly as the RPM of the screw of the extruder. Fig. 2g shows undesirable wetting at high RPM of the screw and high output rates.

The properties obtained by low-shear and high-shear screw designs have been evaluated separately and a comparative study was performed for the ZSK-30 co-rotating extruder on both these designs using the results shown in Tables I and II.

From the data shown in these two tables, the best properties can be seen at low RPM rates of the extruder. The output rates however do not seem to affect the properties. There seems to be a consistency in the properties obtained for low RPM and output rates. Comparing the values of tensile and flexural strengths, heat deflection temperatures and notched izod impact properties, one can conclude that the low-shear screw design gives higher values for the above stated conditions. This indicates that on the co-rotating twin-screw extruder, e.g. the run with 20% glass loading of Nylon 6,6; a low-shear screw design would be preferred to the high-shear screw design. Processing on the ZSK-30 was very smooth and much of the run variables conformed to the actual design. However slight alterations had to be made for low RPMs and high output rates as the barrels were clogged due to overflooding which could cause system shutdown.

The Leistritz® is a counter-rotating twin-screw extruder where the screws rotate in the opposite direction and the polymer melt is carried across the screw in the middle of the screws. The Leistritz® extruders along with the Werner Pfleiderer® ZSK's are commonly used in the plastics industry for processing a variety of polymers. Certain problems were encountered while operating the Leistritz® counter-rotating extruder when trying to conform the operation to the statistical process design values. As such, some slight changes had to be made in the RPM and output rates to allow smooth running of the extruder, e.g. the run with the RPM of 100 and output rate of 300 g/min could not be done because of clogging of the material in the extruder line and as a result either the RPM had to be increased or the output rate had to be decreased. Table III summarizes the results with a low-shear screw design on the Leistritz® 30 mm twin-screw extruder.

From Fig. 3a, it can be observed that the best tensile properties were obtained when the extruder was operated at low RPMs. The extruder could not be operated with high output rates as certain modifications were required. But with low RPMs, the tensile properties were around 124,110 MPa.

Flexural properties do not show very high values when compared to the values obtained from ZSK-30, but the low RPM of the screw gives the best material, i.e., a strong material with high tensile and flexural strengths. The values of low RPMs and low output rates are around 213,745 MPa and it can be clearly seen that low RPMs and low output rates produce the toughest material. Run No. 4 in Table IV had to be repeated a second time as the extruder could not handle RPM of 150 and a high output rate of 300 g/min. Therefore, this run was repeated with lower values of RPM and output rate.

Fig. 3c shows the HDT variations with a low-shear screw on the Leistritz® extruder. There was no sizeable change in the HDT temperatures of the test bars at low RPMs, both at high and low output rates. Temperatures of around 245 °C were obtained for the runs with low RPMs and output rates.

Fig. 3d illustrates the notched izod strengths of the reinforced material with a low-shear screw on the Leistritz® extruder. The notched izod values were comparatively lower than the values obtained with ZSK-30. Low RPM and output rates gave the best values of around 1.1 ft lb/in.

Table IV summarizes the results of a high-shear screw design on the Leistritz® extruder counter-rotating twin-screw extruder.

With the high-shear screw design on the Leistritz®, procedural difficulties were encountered such as clogging with low RPMs and very high output rates and two runs had to be cancelled as the machine was over torquing. In general the output rates on this extruder

TABLE III Results from the low-shear-screw design on the Leistritz®

Run no.	RPM	Output (g/min)	Tensile (psi)	Flexural (psi)	HDT (°C)	Notched izod (ft lb/in)	MFL (μm)
1.	150	110	15810	26840	237	0.784	246.5
2.	150	300	19380	32980	243	1.09	261.1
3.	100	100	18140	31870	242.5	1.125	303.5
4.	150	300	18620	31990	242	1.097	242
4A.	90	200	18060	30970	244.5	1.004	243
5.	120	300	18570	31860	242	1.085	246.6
6	200	300	18160	32830	238.5	1.195	237.5
7.	70	110	18430	32190	247	1.159	320.8
8.	70	110	19730	33520	248	1.291	257

TABLE IV Results from the high-shear-screw design on the Leistritz®

Run no.	RPM	Output (g/min)	Tensile (psi)	Flexural (psi)	HDT (°C)	Notched izod (ft lb/in)	MFL (μm)
1.	150	100	18430	31550	241.5	1.22	421.5
2.	150	200	19210	32480	246	1.202	318
3.	150	100	17980	30890	242	1.22	436.5
4.	90	200	19220	32690	244.5	1.201	342.5
5.	—	—	—	—	—	—	—
6.	—	—	—	—	—	—	—
7.	70	100	20200	33630	244	1.294	420
8.	70	100	20840	34590	245.5	1.359	553.25

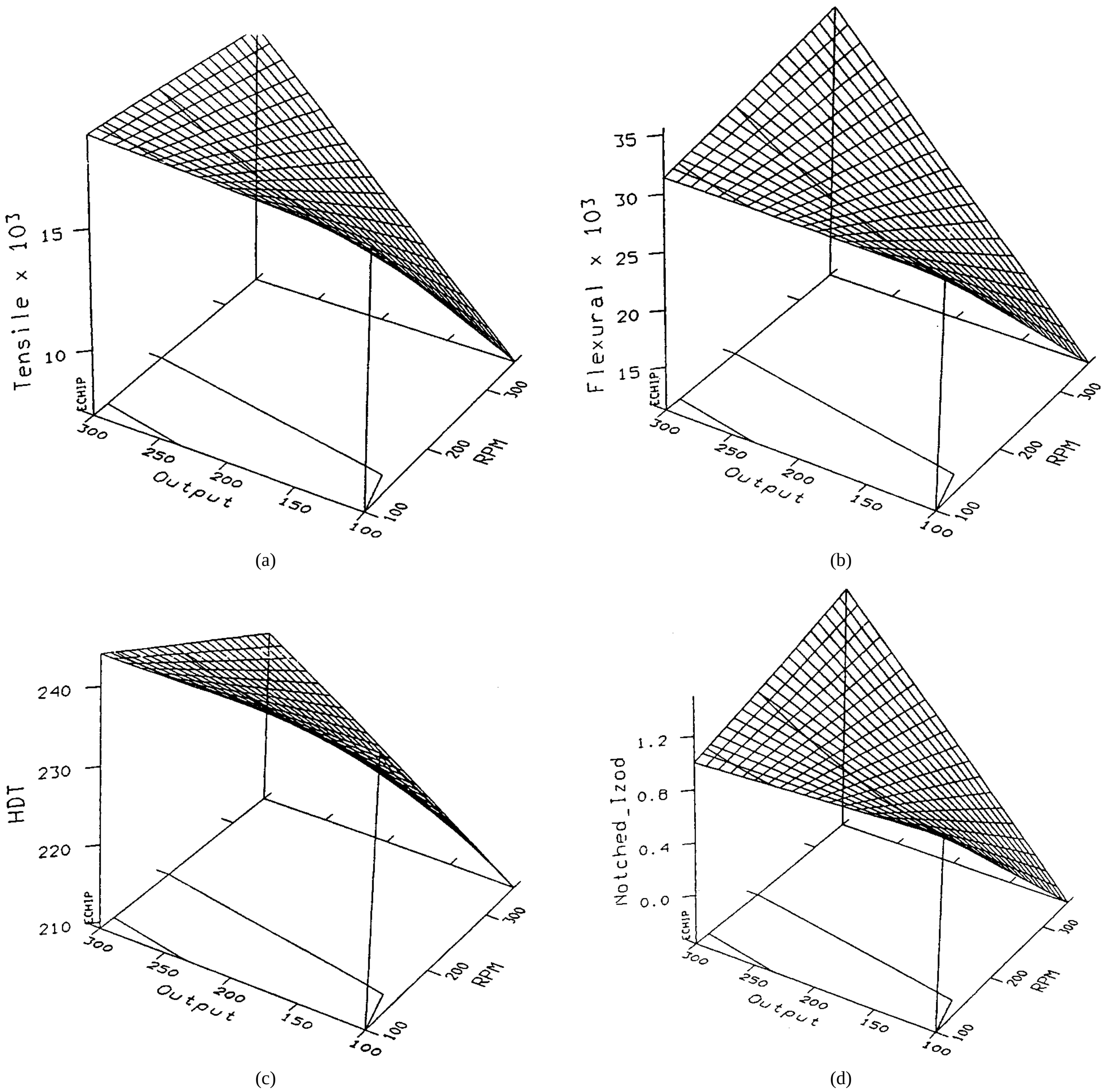

Figure 3 (a) Tensile variation with low shear screw on the Leistritz. (b) Flexural variation with low shear screw on the Leistritz. (c) HDT variation with low shear screw on the Leistritz. (d) Notched izod variation with low shear screw on the Leistritz.

with the high-shear screw could not be increased more that 200 g/min for the designed RPMs.

Fig. 4a shows the variation of tensile strength with RPM and output rates for the high-shear screw design on the Leistritz®. High values of tensile strength are obtained for very low RPMs (70) and low output rates (100 g/min).

Fig. 4b shows the flexural strengths for the high-shear screw design on the Leistritz®. Comparing these values with the tensile strengths, similar pattern of values are repeated. Even here for high RPMs and output rates, less flexural strength values were observed whereas for low RPM and low output rates, higher values are obtained.

Fig. 4c shows the HDT temperature variation with RPM and output rates under a pressure of 1,820 MPa (264 psi). It can be clearly seen from the graph that as the RPMs increase, the DDT values and high output rates give better values for HDT. Fig. 4d reveals the notched izod variation with RPM and output rates. Clear indications from the graph reiterate the high strength of the material at low RPMs and low output rates. The findings of the notched izod values further support the results obtained for tensile, flexural and HDT properties for the high-shear screw design on the Leistritz® 30 mm twin-screw extruder.

The results with both the low and high-shear screw designs on the Leistritz® 30 mm extruder can be compared for the best properties. Upon examination of the various properties in Tables III and IV, the strength of the material from the Leistritz® extruder seems to be unaffected by the design of the screw. Both low-shear screw and high-shear screw seem to give similar results.

5. Discussion

The results on low and high-shear screws were evaluated for both ZSK-30 co-rotating twin-screw extruder

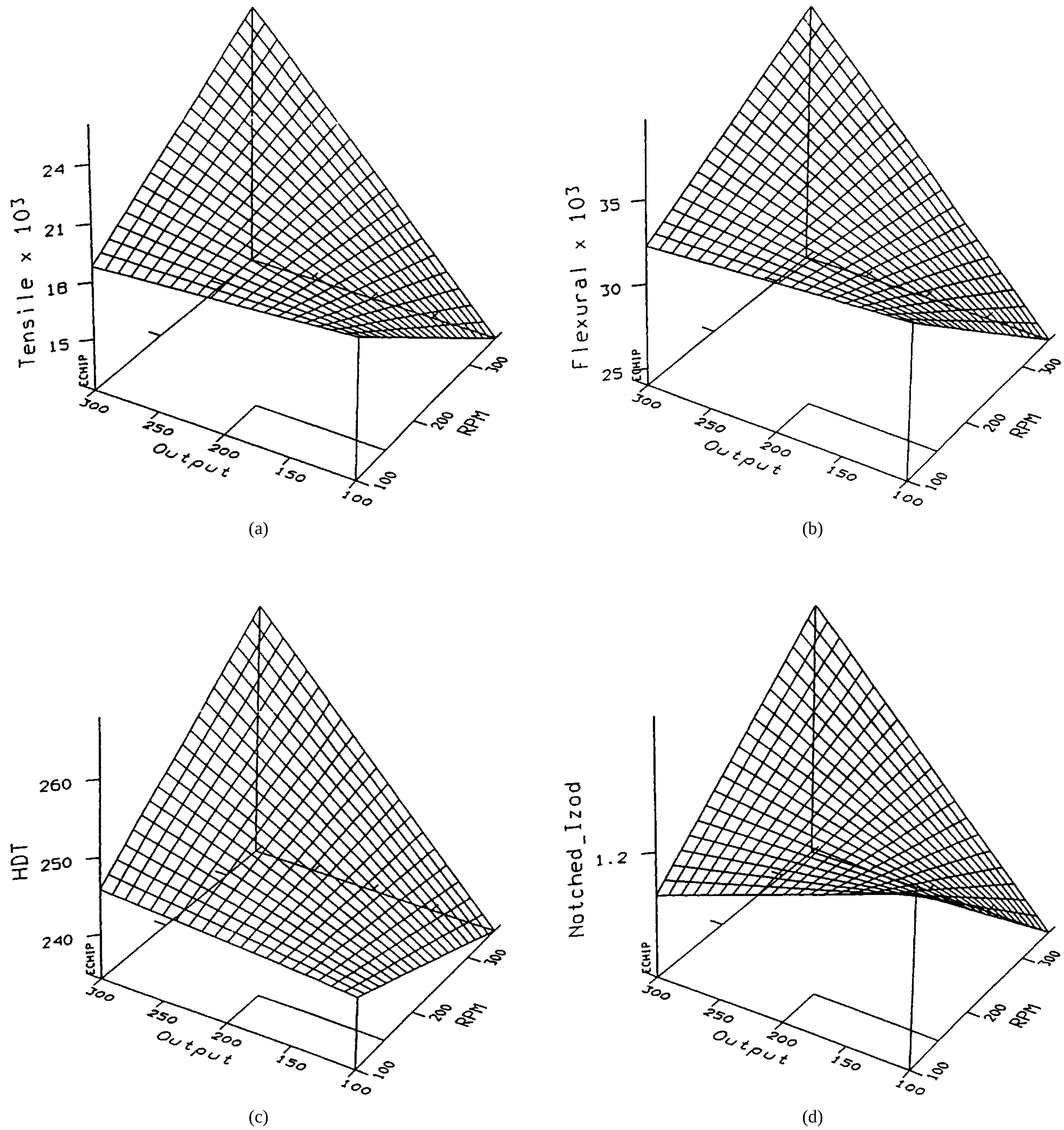

Figure 4 (a) Tensile variation with high shear screw on the Leistritz. (b) Flexural variation with high shear screw on the Leistritz. (c) HDT variation with high shear screw on the Leistritz. (d) Notched izod variation with high shear screw on the Leistritz.

and Leistritz® 30 mm counter-rotating twin-screw extruder. Examining the results, Tables I and II, one notes that more desirable properties were obtained with low-shear screw design on the ZSK-30.

Similarly, from Tables III and IV, it can be concluded that the screw design on the counter-rotating twin-screw extruder does not affect the properties of the material. The fiber reinforced material with similar strength was observed with both high and low-shear screw designs.

On a wide comparison of the various results obtained from Tables I–IV, higher values of tensile strength, flexural strength, HDT and notched izod were obtained using both low and high-shear screw designs for the ZSK-30 when compared to the Leistriitz®. For example, a run processed with the low-shear screw design on ZSK-30 extruder with RPM of the screw at 100 and output rate of 100 g/min; resulted in tensile and flexural strengths of 146,652 and 246,694 MPa respectively. But for the same RPM and output rates on the Leistritz® 30 mm extruder, the tensile and flexural strengths were 125,071 and 219,736 MPa respectively. HDT and Notched Izod values were also very high with the ZSK-30 when compared to the Leistritz®.

The scanning electron microscope photographs suggest better distribution of the glass fibers in the polymer for the material for the ZSK-30 extruder. Superior wetting of the glass fibers was also revealed through these photographs for the reinforced material obtained through ZSK-30.

6. Conclusions

The results of this study clearly show that the best properties, tensile, flexural, HDT and notched izod were obtained at low RPM of the screw on the co-rotating ZSK-30 Werner Pfleiderer® extruder. The output rate of the extruder was found to have only moderate effect on improvement of the properties. Low RPMs and output rates of the screw with both counter and co-rotating twin screw extruders showed good wetting and adhesion characteristics. It was also observed that the fiber length did not affect the properties of the material.

This study could extend further into the microstructure of the reinforced material. Glass fiber alignment in the polymer melt could be an important factor in determining the strength of the material. Further work on extruders of higher magnitude and varying glass loading rates is required to establish all the factors responsible for improving the properties of the fiber reinforced plastics.

Acknowledgements

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