JOURNAL OF MATERIALS SCIENCE 15 (1980) 2834–2842

The effect of recycled plastic and compound additives on the properties of an injection-moulded polypropylene co-polymer

Part 1 *Study materials and experimental methods*

DERFEL WILLIAMS*, MICHAEL BEVIS*
Department of Non-Metallic Materials, Brunel University, Uxbridge, Middlesex, UK

A comprehensive analysis of the effects of recycled material, recommended pigments, pigment concentration, and selected stabilizers on the microstructure and impact properties of a polypropylene co-polymer has been undertaken. This paper reports on the study materials, which includes details of more than sixty compounds used in the investigation, and the experimental methods which were used to characterize the injection-moulded specimens to be reported in the following parts of this work.

1. Introduction

Scrap material inevitably arises during the manufacture of thermoplastic artefacts. In injection-moulding, for example, scrap can arise in the following ways:

(a) in the form of sprues and runners which can represent a large percentage of the shot weight, as determined by the design of the injection mould and the complexity of the finished artefact;

(b) during setting-up and colour changing operations;

(c) reject mouldings or trim.

Large quantities of thermoplastic can also be recovered after use and can, with the aid of various washing, filtering and densification methods, be used either solely or as a percentage of feedstock for the manufacture of artefacts. For example, boxes, twine and crates arising in the horticultural, agricultural, produce and brewing industries are examples of polypropylene produces which arise in large quantities and provide sources of materials for recovery and re-use. Crates and boxes can be recycled following communition by granulation, washing, separation of impurities (heavies) by sink–float methods, drying and melt-filtration. In Europe, a high proportion of recycled plastic has been successfully used in the manufacture of some types of crates. Pressures of an ecological or economic nature tend to encourage the practice of using reground or recovered material. However, apparent economic gains in this practice have to be balanced against deterioration in the properties of finished parts, compared with those prepared from virgin materials. These properties include impact strength, appearance, reproducability of dimensions and reject rate, all of which may arise from one or more of the following reasons:

(a) contamination by foreign particles such as barrel residues of other thermoplastics, dirt particles, grease etc.;

(b) decrease in thermal stability and irregularities in melt-flow characteristics arising from thermal degradation of the polymer;

(c) loss of additives such as plasticizer, lubricants and anti-oxidants, or changes in the nature of the filler particles such as a change in glass fibre length or dispersion with successive reprocessing;

(d) variable hopper flow characteristics as a consequence of using a non-uniform particle size.

Various studies of the influence of using re-

---

* The authors were formerly in the Department of Metallurgy and Materials Science at Liverpool University where the initial work reported in this paper was carried out.

0022–2461/80/112834–09$02.90/0

ground material have been carried out and the previously published papers report work on the effect of recycling on the properties of moulded polycarbonate [1], polystyrene and polyethylenes [2] and polyethylene melt rheology [3]. Some of the chemical problems which arise in the recycling of plastics have been reviewed by Scott [4] and aspects of quality control and quality maintenance in reprocessing thermoplastics have been extensively studied by Hermann [5] and Zoller [6] and Nightingale [7] has undertaken a study of these aspects, specifically for nylons. Plastics differ markedly from one another in the ways in which their properties change following successive processing operations and therefore it is necessary to consider the different plastics individually.

In general, the previously reported investigations have tended to concentrate on a single thermoplastic compound in each case, and have excluded a detailed consideration of the influence of processing conditions, pigment type, percentage of reground material, percentage of pigment and presence and percentage of other additives on the microstructure and consequent mechanical properties of mouldings. The aim of the investigation reported in this paper was to study, in a systematic manner, the influence of these parameters on the properties of polypropylene co-polymer injection-mouldings in which part, or all, of the feedstock consisted of recycled material.

2. Scope of the investigation

The complete survey will be presented in four papers and, in addition to providing a comprehensive analysis of the effects of reground material on the mechanical properties of a polypropylene co-polymer, it will provide a basis for comparing the relative effects of recommended pigments, pigment concentration, stabilizers and the percentage of reground material on the mechanical properties.

In this paper, the study materials and the experimental methods used in the work are described and the results of the survey will be presented in Parts 2, 3 and 4. In Part 2 the influences of percentage reground material used on the impact strength of moulded plaques and the moulding reproducibility and microstructure of mouldings in unpigmented polypropylene co-polymers are reported. The results obtained also provide a basis for the comparison of the properties of pigmented and ultraviolet light stabilized compounds. Part 3 concentrates on the effects of incorporating three different pigments, three pigment concentrations and the proportion of reground material on the microstructure and impact properties of the same co-polymer. The results of work on the influence of compound formulation on the weathering properties will also be reported. The influence of ageing as well as weathering time on the mechanical properties of the same range of injection moulded polypropylene co-polymer compounds will also be reported in this paper. Part 4 reports on the effects of incorporating a UV stabilizer, together with selected proportions of reground material, and of the type and concentration of pigment on the microstructure and material properties of co-polymer polypropylene.

TABLE I Tabulation of the compounds from which the results to be presented in Part 2 were obtained. All of the compounds were free from pigments and were based solely on a proportional content of recycled plastic. The notation used to identify the range of 19 compounds is described in the text

General description of feedstock	Increasing percentage of material recycled once	Increasing percentage of material recycled once	Increasing percentage of pelletized material recycled twice	Material recycled three and four times	Continuously recycling at 10% level
Unpigmented containing no additional stabilizer	V	V	V	V – 100G3	V
	V – 10 G1	V – 10 P1	V – 10 G2	V – 100G4	V – 10 G1
	V – 25 G1	V – 25 P1	V – 25 G2		V – 9 G1 1 G1
	V – 50 G1	V – 50 P1	V – 50 G2		V – 9 G1 – 0.9 G2 0.1 G3
	V – 100 G1	V – 100P1	V – 100 G2		V – 9 G1 – 0.9 G2 – 0.09 G3 – 0.01 G4

TABLE II Tabulation of the compounds from which the results to be presented in Part 3 were obtained. All of the compounds contained 0.1%, 0.5% or 2% phthalocyanine blue, ultramarine blue or carbon black pigments, and include a proportional content of recycled plastic of the same pigment composition

General description of feedstock	Virgin material with no pigment	Virgin material with 0.1% pigment; increasing percentage of recycled material	Virgin material with 0.5% pigment: increasing percentage of recycled material	Virgin material with 2.0% pigment; increasing percentage of recycled material
Phthalocyanine blue	V	0.1A – 0G1	0.5A – 0G1	2.0A – 0G1
		0.1A – 10G1	0.5A – 10G1	2.0A – 10G1
		0.1A – 25G1	0.5A – 25G1	2.0A – 25G1
		0.1A – 100G1	0.5A – 100G1	2.0A – 100G1
Ultramarine blue	V	0.1B – 0G1	0.5B – 0G1	2.0B – 0G1
		0.1B – 10G1	0.5B – 10G1	2.0B – 10G1
		0.1B – 25G1	0.5B – 25G1	2.0B – 25G1
		0.1B – 100G1	0.5B – 100G1	2.0B – 100G1
Carbon black	V	0.1C – 0G1	0.5C – 0G1	2.0C – 0G1
		0.1C – 10G1	0.5C – 10G1	2.0C – 10G1
		0.1C – 25G1	0.5C – 25G1	2.0C – 25G1
		0.1C – 100G1	0.5C – 100G1	2.0C – 100G1

3. Study materials and notation

The range of compounds reported on in Papers 2, 3 and 4 are identified in Tables I, II and III, respectively. The compounds will be identified using the following notation. The first letter will represent the pigment type. For the UV stabilized grades this letter will be followed by an S, and preceded in both cases by a number which will represent the weight per cent concentration of pigment. The type of pigmentation is represented by the letters V, A, B and C which correspond to virgin unpigmented polymer, phthalocyanine blue, ultramarine blue and carbon black, respectively.

The second half of the notation represents the concentration and type of reground material. The first number represents the weight per cent concentration of the reground material in the feedstock, the following letter represents the form of the reground material where P and G represent pelletized and granulated material, respectively, and the following number represents the number of times the material was recycled prior to the final injection-moulding cycle. GW is used to represent material in the form of plaques which had been weathered prior to granulation.

For example, VS–25G1 represents unpigmented UV stabilized polymer containing 25 wt% reground material in granulated form, which had been recycled once prior to the final injection-moulding cycle. To give another example compound 0.5B–25G1 represents polymer containing 0.5 wt% ultramarine blue pigment, and containing

TABLE III Tabulation of the compounds from which the results to be presented in Part 4 were obtained. In addition to pigment-free virgin polypropylene and compounds based on recycled virgin plastic these compounds contained weathered plastic, ultra-violet light stabilized grade of polypropylene which also contained 2% of phthalocyanine blue ultramarine blue or carbon black pigment

Unpigmented virgin material with increasing material recycled once	25% of material weathered and recycled	Unpigmented UV stabilised virgin material with increasing percentage of material recycled once	2.0% phthalocyanine blue pigmented UV stabilised virgin material with increasing percentage of material recycled once	2.0% ultramarine blue pigmented UV stabilized virgin material with increasing percentage of material recycled once	2.0% carbon black pigmented UV stabilized virgin material with increasing percentage of material recycled once
V	V	VS	2.0AS – 0G1	2.0BS – 0G1	2.0CS – 0G1
V – 10G1	V – 25GW1	VS – 10G1	2.0AS – 10G1	2.0BS – 10G1	2.0CS – 10G1
V – 25G1		VS – 25G1	2.0AS – 25G1	2.0BS – 25G1	2.0CS – 25G1
V – 100G1		VS – 100G1	2.0AS – 100G1	2.0BS – 100G1	2.0CS – 100G1

25 wt% reground material in granulated form which had been recycled once prior to the final injection-moulding cycle.

4. Preparation of feedstock

4.1. Natural compounds

The natural granules were moulded in the as-supplied condition. The co-polymer used throughout the work reported was a commercial injection-moulding grade, with a Melt Flow Index measured at 230° C, according to ASTM D1238, of between 3 and 4 grams in 10 minutes, and a density of 0.905 g cm^{-3}. The initial work, reported in Parts 2 and 3 was limited to a general purpose grade which was stabilized at a level sufficient to prevent excessive thermal degradation during normal injection-moulding, and which did not contain any additional ultraviolet stabilizer or anti-oxidant. The regrind was prepared under very clean conditons in order to reduce the possibility of contanimants affecting the results obtained. A Bone Cravens Daniels 350-120 and a Stubbe SKM 76-110 reciprocating screw injection-moulding machines were used to mould virgin polypropylene granules into plaques. The hopper, barrel and screw of the injection moulders were stripped and cleaned thoroughly, and the cleaning process was completed by purging the machine prior to moulding with a new compound. The mouldings, sprues and runners were then reduced to small particle size with a Leesona 1216 granulator. The stripping and thorough cleaning of the granulator prior to its use for preparing regrind was considered to be essential because this part of the process more than any other is likely to introduce contaminants into feedstock. The regrind so obtained was in the form of irregular shaped particles with dimensions determined by a 10 mm screen.

A proportion of the reground natural material was subsequently injection-moulded into plaques and granulated to give material which had been recycled twice. By repeating the procedure feedstock which had been recycled three or four times was produced. In order to study the effect of an additional heat cycle and of particle shape some of the regrind was extruded into strands with a Johnson Spartan 40 mm Extruder and subsequently pelletized to a uniform particle size. The different types of regrind were then mixed with virgin granules to give feedstocks containing 0, 10, 25, 50 and 100 % regrind.

The recycling of the polypropylene co-polymer, according to the thermal processing determined by the injection-moulded cycle described in Part 2, gave rise to a change in molecular weight characteristics. The melt flow index (MFI) values according to ASTM D1238 and the number- and weight-average molecular weights as determined by gel permeation chromotography* for virgin unpigmented polymer and 100% recycled material after successive recycles are given in Table IV. The natural polymer was also continuously recycled at the 10% level to give the substantial quantities of feedstock required for injection-moulding. The composition of the four feedstocks in terms of the percentages of G1, G2, G3 and G4 are given in the last column of Table I.

4.2. Pigmented compounds

Feedstocks consisting of pigmented polypropylene co-polymer were also prepared. The phthalocyanine and ultramarine blue pigments were separately tumble mixed with natural polymer and a small quantity of spindle oil for a set time to give 0.1, 0.5 and 2.0 wt% concentrations of pigments. Carbon black was introduced in master-batch form and blended with natural granules to give 0.1, 0.5 and 2.0 wt% concentrations of pigment.

The pigmented compounds containing recycled material were prepared in the same way as the natural compounds, with particular care being taken to minimize cross-contamination of the different pigments.

All three pigment types were recommended for use by the manufacturer of the co-polymer. The

TABLE IV The melt flow indices and number average molecular weights of pigment-free polypropylene after consecutive injection-moulding cycles

Number of passes	Melt Flow indices (g per 10 min)	Number average molecular weight ($\times 10^4$)
0	4.5	6.33
1	5.2	5.24
2	6.3	4.32
3	7.2	2.48
4	7.5	2.53

---

* These measurements were carried out by the Polymer Supply and Characterization Centre at the Rubber and Plastics Research Association.

carbon black was selected for the investigation because it is widely used as a pigment. The masterbatch contained 30 wt% of a medium size particle (17 mm) furnace black. The enhancement of weathering resistance by the incorporation of the carbon black has been substantiated. A good dispersion of 2 % carbon black is recommended by the polymer manufacturers for artefacts subject to outdoor applications. Carbon black has been reported as having an adverse effect on the efficiency of anti-oxidant systems [8], a factor which could prove to be important in recycling operations. The ultramarine and phthalocyanine blues were both supplied in powder form. Two pigments of the same colour were chosen because they absorb in approximately the same region of the visible spectrum yet have different chemical compositions. The use of copper phthalocyanine allowed the reported [9, 10] adverse effect of copper on the stability of polypropylene to be investigated.

4.3. Ultra-violet light stabilized compounds

The UV stabilized polymer and the unstabilized co-polymer, referred to in Section 4.1., were the same, except that the former contained an additional UV stabilizer. The stabilized co-polymer was purchased from the polymer manufacturer as a standard compound. Compounds based on this material were prepared using the procedures described above to give pigmented compounds and to give compounds containing several levels of recycled material. In this way a more complete range of compounds, and compounds which were more typical of those encountered in practice, were provided.

5. Injection-moulding

In all, more than 500 kg of material, representing more than 10 000 mouldings, were prepared on the Bone Cravens Daniels 350-120 and Stubbe SKM 76-11- injection-moulding machines. Great care was taken to ensure that no machine variations effectd the results obtained, and the first one hundred mouldings manufactured during start-up operations were not included in the study. Moulding was continuous during runs of up to ten hours for each condition being tested, using polymer from the same batch supplied by the manufacturers. All machine parameters were kept constant and the machines were kept scrupulously clean to avoid anomalies due to contamination.

Two moulds were used in the reported programme of work: a 3 mm thick, double film-gated plaque mould and a 6 mm thick, fan-gated plaque mould. The geometry, location of mould temperature sensor (Y) and cavity pressure sensor (X) are shown in Fig. 1.

The Dynisco FT444 strain-gauge transducer was located behind a dummy ejector pin, and the changes in cavity pressure and temperature during mould filling were continuously monitored.

The optimum conditions for injection-moulding virgin granules were identified, and included a consideration of first and second stage pressures, mould and melt temperatures, cooling and injection times and the geometry of the moulds and sprues.

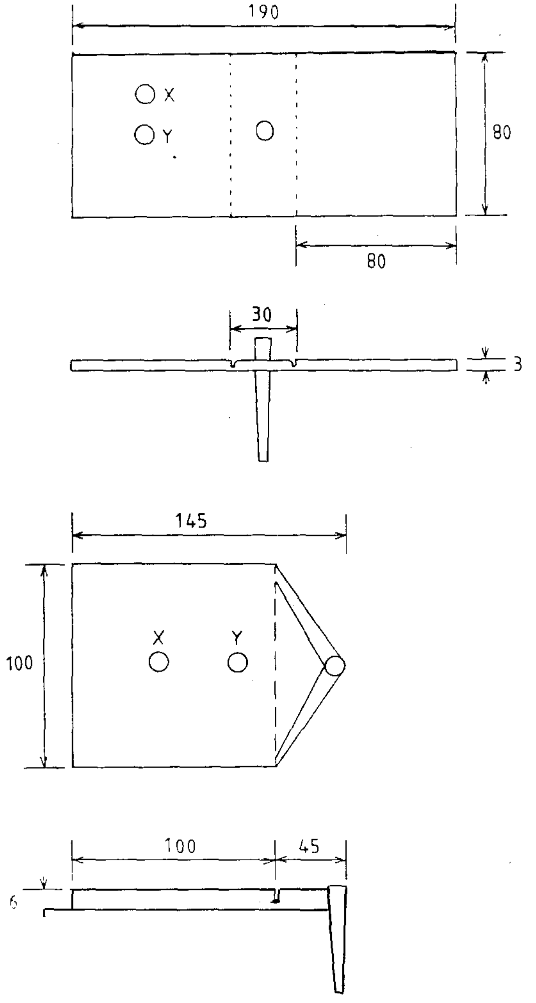

Figure 1 Schematic diagram of the mouldings used during the course of the reported investigation showing dimensions (in mm) and the location of pressure (X) and temperature (Y) sensors.

Optimization of the moulding conditions was realized with the aid of cavity pressure profiles, particularly in relation to the magnitude and timing of injection and hold-on pressures. Once the optimum conditions for the virgin granules had been established they were kept constant for the feedstocks containing recycled material, so that any differences in the moulding properties would be due to differences in the formulation of compounds and not to changes in the machine variables.

It is widely realized that satisfactory mouldings can be prepared under a wide, range of processing conditions, and it would generally be considered unwise to base an investigation on just one set of processing conditions. However, in a situation where a wide range of compounds is being investigated it soon becomes impractical to consider using a range of processing conditions, in view of the considerable time and cost involved in moulding and the critical assessment of the performance of the moulded parts. The authors were faced with a dilemma over which route to take, and eventually decided on the former. Work is now in progress, however, to assess the extent to which changes in processing conditions can influence the results presented in the following papers, particularly in relation to Parts 3 and 4 of this work.

The use of feedstock which consists of irregularly shaped particles can result in marked changes in the reproducability of shot weight and in the dimensions of finished parts [11–12]. Extrusion and pellet formation, whilst giving a more regularly shaped particle, cause further molecular degradation and changes in bulk density which effect the heat transfer characteristics during injection moulding. The use of recycled material and changes in particle size distribution will therefore influence pressure and temperature profiles in moulding which consequently effects shot weight, microstructure and the properties of the moulded parts. The reproducability of cavity pressure and shot weight was relatively easy to monitor with changes in compound formulation, and for this reason the variations in shot weight and cavity pressure were measured throughout the moulding runs reported in Paper 2. The variation in shot weight and peak cavity pressure measured for individual mouldings with changes in compound formulation will be presented, together with mean weight and mean peak cavity pressure, and the variance, standard deviation and coefficient of variation results, as calculated from data gained from the last thirty moulding produced from any one compound formulation.

6. Weathering

Molecular degradation and loss of additives may occur during processing and result in an increase in free radicals and active chain ends. Mouldings prepared from recycled material may therefore be more susceptible to weathering effects than mouldings prepared from virgin materials. The case for investigating the effects of weathering on the properties of the range of compounds identified in Tables I–III was further reinforced by reports [8, 13, 14] that certain pigments accelerate the degradation processes.

The 6 mm thick plaques containing no pigment but containing various percentages of recycled materials were artificially weathered using a Marr weathering machine. They were exposed for 1160 h under a kW carbon arc, which is approximately the equivalent of one year of outdoor exposure. One half of the 3 mm thick plaques made from the compounds identified in Tables II and III were tested soon after moulding, while the other half were exposed to natural weathering for 6 to 12 months.

The recommended standard practice for outdoor weatherings according to ASTM D1435 was adopted. The test plaques were placed on racks at 45° to the horizontal and so positioned that exposed surfaces of the samples faced the equator.

The test site for plaques discussed in Parts 3 and 4 was at Brunel University for test periods beginning in May 1978 and ending in October 1978, and beginning in January 1979 and ending in June 1979, respectively. The set of compounds consisting of increasing percentage of recycled-once material containing no additional stabilizer or pigment (compounds V, V-10G1, V-100G1 in Table I) was common to all sites and periods tested and provided a means of comparison between the three conditions studied.

7. Mechanical testing

7.1. Impact testing

The drop-dart impact test was selected as the main mechanical test method because of its sensitivity to molecular weight changes, and therefore to differences in which may arise when recycled material is used as feedstock material for injection-moulding. The advantages of this form of impact test for the characterization of complete moulded parts have recently been described by Cole [15], who also made clear both the need to reproducibly locate the site of the impact test, and the need for the tester to arrive at a suitable and practical criterion of failure which can be used quickly and routinely.

The ASTM D2444 and D3029 standard falling Tup test methods were adopted by the authors where practically possible. The C type Tup with a nose radius of 6.35 ± 0.05 mm and a weight range from 3 kg to 4.5 kg was used for all but the most brittle specimens. The point of impact for each specimen was the centre of the test piece. Because of the nature of the test, at least twenty-five specimens for each compound were tested, and the results presented are based on results taken from more than 10 000 specimens.

The ductile–brittle transition temperature for plaques prepared from 100% unpigmented virgin material was shown to be approximately 16° C. A ductile–brittle transition temperature of 23° C was measured for the plaques prepared from twice recycled natural material (V-100 G2). Impact tests were conducted at − 10° C and at 23° C, in order to simulate impact in the ductile and brittle temperature regimes, and in the former case under conditions which simulate unfavourable conditions likely to be encountered in practice.

All of the specimens that failed at − 10° C did so in a brittle manner and provided a straightforward criterion for failure. For the tests at 23° C, the failures were of a ductile nature and the criterion for failure was the visual detection of a crack which had propagated through the thickness of the test plaque.

7.2. Tensile testing

Tensile testing was carried out according to ASTM D638 at 23° C and 60% relative humidity (r.h.). The test specimens were cut from the 3 mm thick plaque mouldings so that the tensile axis was either parallel or perpendicular to the injection direction. The edges of the specimens were hand-ground using five grades of emery papers in the range 180 to 600, and finally polished by hand using α-alumina, distilled water and selvyt cloth.

8. Microstructure characterisation

The effects of processing variables upon the morphology of injection-moulded polypropylene homo-polymer and co-polymer have been reported by several investigators. Kantz, Newman and Stigale [16] used X-ray diffraction methods and transmitted light microscopy of microtomed sections to reveal the presence of three distinct zones of different microstructures through the thickness of the mouldings. These were identified as the skin-, shear- and core-zones. The skin-zone showed no perceptible crystalline development but the molecules were highly orientated with the flow direction as evidenced by pronounced bi-refringence effects. The shear-zone exhibited negatively bi-refringent rows of Type III spherulites of the metastable hexagonal form. The core-zone or region which usually makes up most of the volume in injection mouldings was shown mainly to contain the positively bi-refringent monoclinic Type I spherulites.

Marked changes in the area fraction of the different zones and the morphology of the spherulites were shown to occur with changes in the pro-

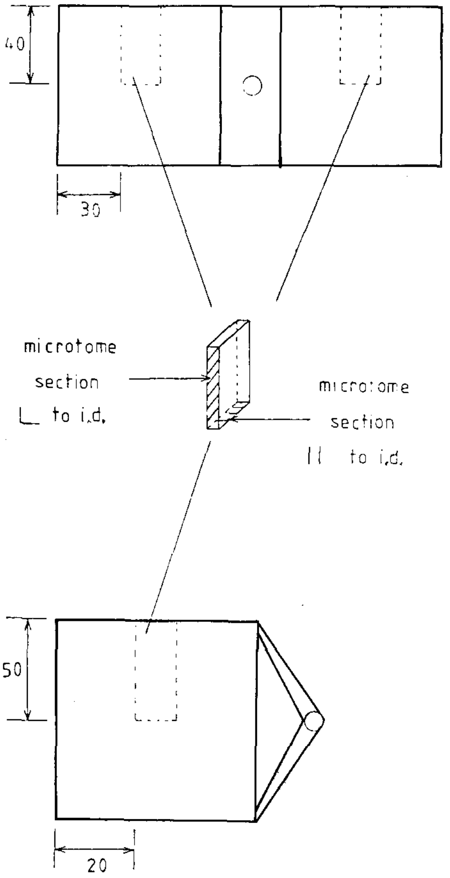

Figure 2 Schematic diagram showing the location and orientations of the thin-sections used for the micromorphological characterization of the injection-mouldings. Dimensions shown are in mm.

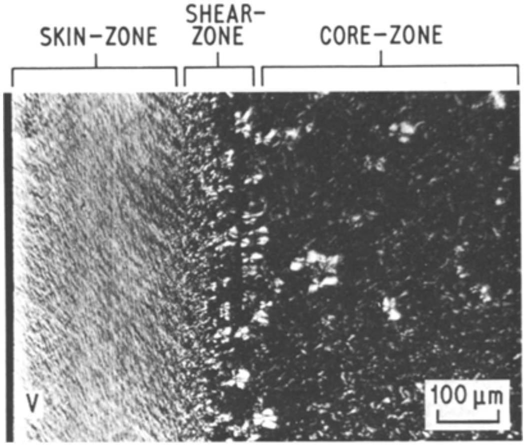

Figure 3 Light micrograph obtained using a thin section taken from a typical moulding prepared from the virgin polypropylene compound (V). The micrograph shows the characteristic skin-, shear- and core-zones which exhibit different micromorphologies.

cessing variables. These changes were readily identified by transmitted light microscopy examinations of thin sections prepared by microtomy from the bulk injection-moulded specimens.

The authors have adopted the transmitted light microscopy examination of thin sections as the primary method for the characterization of specimens prepared from the compounds listed in Tables I–III. Previously reported observations [16–20] indicate that the technique would be capable of identifying changes in micromorphology associated with either changes in molecular weight due to recycling or with changes caused by the addition of pigments with different nucleation characteristics.

In all cases the thin sections were prepared from the injection-mouldings by microtomy with a sledge microtome and in the orientations relative to the injection direction shown in Fig. 2. A typical section obtained from an injection-moulded plaque made from virgin material V in Table II is shown in Fig. 3, which also details the spherulite morphology.

Several investigators [16, 17, 21] have examined the micromorphology of polypropylene mouldings in more detail using etching and X-ray diffraction techniques, with the objective of explaining the occurrence of the different zones, which can be further sub-divided when the mouldings are produced under selected processing conditions or with particular additives [22]. An example of the influence of pigments on the microstructure of copolymer mouldings is illustrated in Fig. 4 which shows a transmission light micrograph of a section taken from an injection-moulding of 2.0A-0G1 (see Table II). The use of techniques other than transmission light microscopy of thin sections was considered to be unnecessary because of the effectiveness of the latter, and also because the volume of work that would have been involved in the examination of the large number of moulded compounds by several techniques could not be justified.

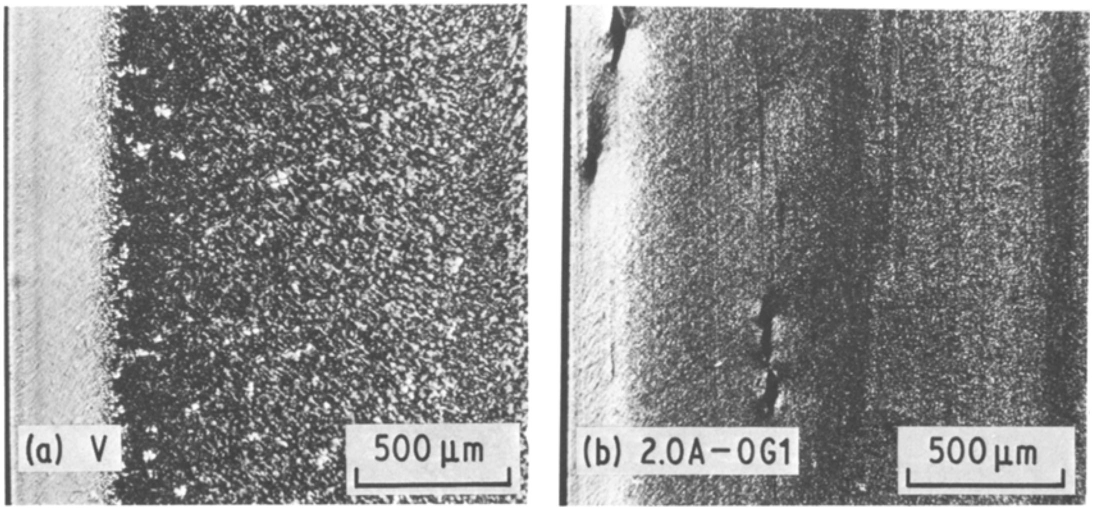

Figure 4 The two micrographs (a) and (b) illustrate the marked effect on the micromorphology of incorporating 2 wt% of recommended pigment into polypropylene injection mouldings. Showing (a) a virgin unpigmented polypropylene (V) moulding and (b) a 2 wt% phthalocyanine blue pigmented virgin polypropylene (2.0A–0G1) moulding, clearly showing the strong nucleating behaviour of the pigment.

9. Conclusion

Parts 2, 3 and 4 will describe the results of the application of the characterization and test methods described above to the range of compounds identified in Tables I, II and III, respectively. Additional variables in the form of compounding additives were introduced into the programme to be described in the following Parts 2, 3 and 4, and it was therefore necessary to successively reduce the extent of characterization work and the number of compounds studied. This could be justified from the earlier results which indicated that some variables caused little change in properties. For example, the results to be presented in Part 2 will show that the reproducibility of injection-mouldings for any one compound is not changed with increasing percentage of reground material, even though such an increase produces changes in the mechanical properties of mouldings. On the basis of this result it was considered that moulding reproducability studies with the pigmented compounds would be unnecessary. Work to be reported in Part 3 showed that changes in pigment type had far greater effect of structure and properties than the incorporation of reground material, so that in the studies on the effect of ultraviolet stabilizers in Part 4 it was possible without significant loss of useful results to reduce the number of study compounds containing reground material.

Acknowledgements

The authors are indebted to the Science Research Council for financial support, and wish to thank Mr S. Bentley for his valuable assistance in the injection-moulding of impact test specimens. Thanks are also due to Dr T. Owen and Mr J. Massey for their help in the physical testing and polymer characterization aspects of the work.

References

1. T. W. SHEA, E. D. NELSON and R. R. CAMMONS, Proceedings of the Society of Plastics Engineers 33rd Annual Technical Conference, Atlanta, Georgia, 1975.
2. H. VOGEL, *Kunstoffe-Plastics* **9(3)** (1975) 11.
3. M. BEIZAE and S. H. R. ZAIDI, *Kunstoffe* **64(4)** (1974) 171.
4. G. SCOTT and D. G. MELLOR, *Eur. Polymer J.* **9** (1973) 219.
5. C. HERMANN, *Kunstofftechnik* **12(8)** (1973) 225.
6. P. ZOLLER, *Kunstoffe-Plastics* **21(10)** (1974) 23.
7. J. E. NIGHTINGALE, Proceedings of the Plastics and Rubber Institute Conference, June, 1975 (Institute of Electrical Engineers, London, 1975).
8. C. W. UZELMEIER, *Soc. Plastics Eng. J.* **26(5)** (1970) 69.
9. J. SHIMADA, *Rev. Elec. Comm. Lab.* **17** (1969) 1146.
10. W. L. HAWKINS, M. G. CHAN and G. L. LINK, *Polymer Eng. Sci.* **11** (1971) 377.
11. K. A. SCOTT, R. T. HUMPIDGE, R. R. WHISSON and F. C. HARBERT, Rubber and Plastics Research Association Report 170, May, 1968.
12. R. R. WHISSON, Rubber and Plastics Research Association Report 188, September, 1970.
13. C. GOTTFRIED and M. J. DUTZER, *J. Appl. Polymer Sci.* **5** (1961) 612.
14. S. L. FITTON and W. TAYLOR, *Plastics* **31** (1966) 1139.
15. E. A. COLE, *Plastics and Polymers* **43(10)** (1975) 180.
16. M. R. KANTZ, H. D. NEWMAN and F. H. STIGALE, *J. Appl. Polymer Sci.* **16** (1972) 1249.
17. D. R. FITCHMUN *et al., ibid.* **14** (1970) 2441.
18. *Idem, ibid.* **14** (1970) 2457.
19. M. N. BECK and M. D. LEDBETTER, *ibid.* **9** (1965) 2131.
20. E. S. CLARK, *Soc. Plastics Eng. J.* **23(7)** (1967) 46.
21. L. BARTOSIEWICZ and Z. MENCIK, *J. Polymer Sci. Phys.* **12** (1974) 1163.
22. F. RYBNIKAR, *J. Appl. Polymer Sci.* **13** (1969) 827.
23. F. J. PADDEN and M. D. KEITH, *J. Appl. Phys.* **30** (1959) 1479.

Received 20 March and accepted 29 April 1980.