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

Part 2 *Moulding reproducability and impact properties*

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Microstructure studies on injection-moulded plaques and the falling dart impact test method have been used to investigate the effect of incorporating recycled material as feedstock on the micromorphology and mechanical properties of polypropylene co-polymer injection-mouldings. It has been shown that marked and systematic changes in mould dimension reproducability and mechanical properties do occur with increasing percentage of recycled materials, and that the results can be explained in terms of changes in rheological properties and accompanying changes in micromorphology.

1. Introduction
The main aim of the work reported in this paper was to assess the effect of incorporating recycled material on the moulding reproducability, moulding microstructure and impact properties of a polypropylene co-polymer. The results of this investigation provide a basis for assessing the effect of selected pigments and additives, and the effect of external ageing on the properties of the polypropylene co-polymer compounds selected for investigation.

The moulds, moulding procedures and the characterization techniques used in the investigation have been described in Part 1 [1].

2. Experimental method
The set processing conditions used in the moulding of all test plaques prepared for the experiments reported in this paper are given in Table I.

Optimum mould filling was achieved for virgin feedstock with the aid of cavity pressure profile measurements, and these processing conditions were maintained for virgin plastics and for feedstocks containing recycled materials. Consequently, any reported differences in the moulding characteristics of the polymer melt and in the microstructure and mechanical properties of the mouldings were due to material changes and not due to changes in machine variables.

Prior to the moulding of the feedstock selected for investigation it was necessary to mould one hundred mouldings with virgin feedstock to achieve injection machine equilibrium. The full range of feedstocks studied is given in Table II (see also Part 1 [1]) where the following notation was adopted. In the notation used V represents virgin unpigmented polymer, and the second part of the notation represents the concentration and type of regound material present in the compound. 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 final number represents the number of times the material was recycled prior to the final injection-moulding cycle. (For example V-9G1-1G2 refers to a compound consisting of virgin unpigmented feedstock containing 9 wt% of material recycled once and 1 wt% of

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TABLE I Set processing conditions used in the moulding of all test plaques

	Moulding machine	
	Stubbe SKM 76 110	Bone Cravens Daniels 350-120
Nozzle heating (° C)	205	225
Heating 1 (° C)	190	215
Heating 2 (° C)	185	195
Heating 3 (° C)	180	180
Mould temperature (°C)	60	60
Cooling time (for the 3 mm thick plaque mould) (secs)	30	30
Cooling time (for the 6 mm thick plaque mould) (secs)	60	
Injection pressure Stage 1 (for the 3 mm thick plaque mould) ($MN\,m^{-2}$)	5.5	10
Injection pressure Stage 1 (for the 6 mm thick plaque mould) ($MN\,m^{-2}$)	8.9	
Injection pressure Stage 2 (for the 3 mm thick plaque mould) ($MN\,m^{-2}$)	5.2	3.5
Injection pressure Stage 2 (for the 3 mm thick plaque mould) ($MN\,m^{-2}$)	5.5	
Injection time (for the 3 mm thick plaque mould) (secs)	30	30
Injection time (for the 6 mm thick plaque mould) (secs)	50	

material recycled twice, prior to the final injection-moulding cycle. The impact testing procedure and the light microscopy technique used for the characterization of the mouldings has been described in Part 1.

3. Experimental results

The results presented below relate to a selection of injection-moulding runs which were designed to identify the extent of the change in the properties of mouldings which could be caused by introducing various formulations of recycled materials as part feedstock. The results are presented in the form of a plot of injection-moulding shot weight against shot number and the corresponding peak cavity pressure against shot number. The mean shot weight, mean peak cavity pressure and their standard deviations and coefficients of variation for specific formulations of virgin and recycled material are tabulated.

3.1. Run 1

These results related to the study of the effect of incorporating recycled material in the form of small irregularly shaped particles, as determined by a 10 mm screen, upon the variation in shot weight of mouldings and the corresponding peak cavity pressures. The experimental results are presented in Figs 1 and 2 and Table III. The feedstock in Run 1 consisted of virgin granules together with 0%, 10%, 25%, 50% and 100% of material which had been recycled once. These formulations and those used in Run 2 have been chosen to represent the range of formulations which could be used in commercial practice. It is clear from the figures and tables that accompanying an increase in the percentage of recycled material there is an increase in the peak cavity pressure and shot weight of the mouldings. The correlation between the shot weight and peak cavity pressures is also clearly demonstrated and confirmed by the rank in order

TABLE II Tabulation of the compounds from which the results presented in this work 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 pelletized material recycled once	Increasing percentage of material recycled twice	Material recycled three and four times	Continuously recycling at 10% level
Unpigmented, containing no additional	V V-10G1 V-25G1 V-50G1 V-100G1	V V-10P1 V-25P1 V-50P1 V-100P1	V V-10G2 V-25G2 V-50G2 V-100G2	V-100G3 V-100G4	V V-10G1 V-9G1-1G2 V-9G1-0.9G2-0.1G3 V-9G1-0.9G2-0.09G3-0.01G4

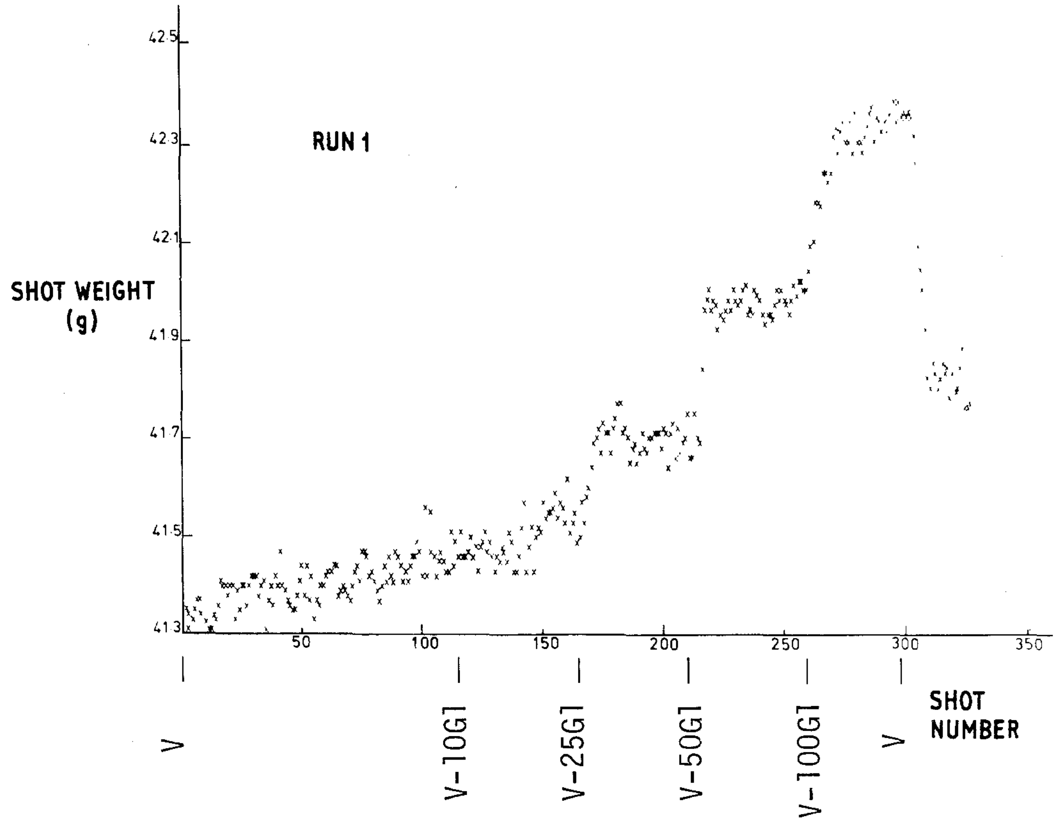

Figure 1 Measured shot weights plotted against shot number for mouldings produced in Run 1 for feedstocks containing proportions of material recycled once.

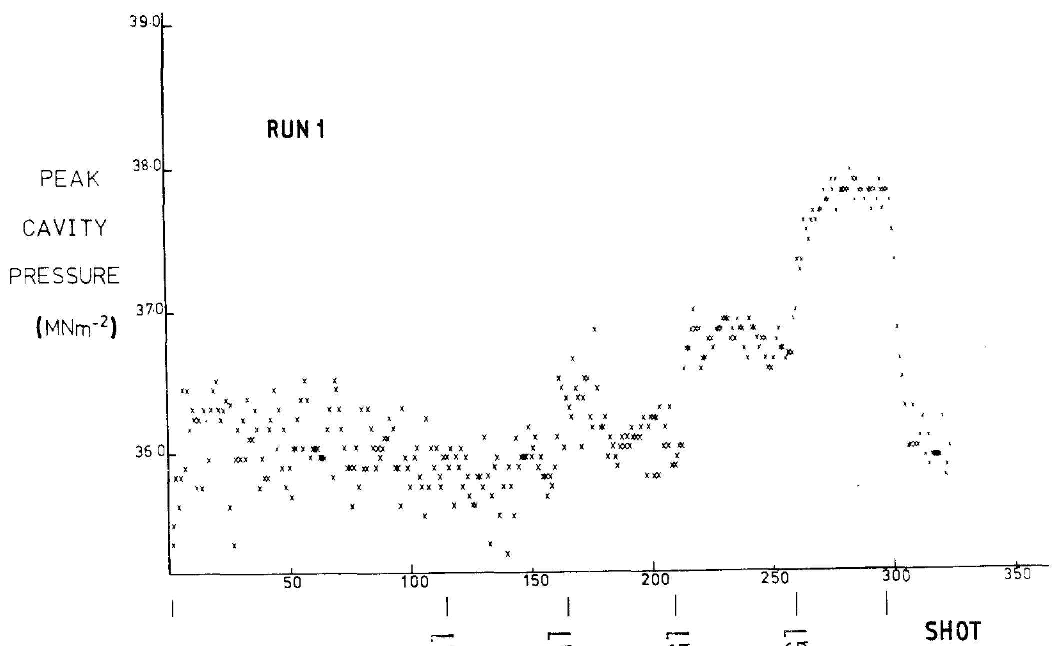

Figure 2 Measured peak cavity pressure plotted against shot number recorded during Run 1.

TABLE III Summary of shot weight and peak cavity pressure data recorded during Run 1

Type of feedstock	Mean shot weight (gms)	Standard deviation	Coefficient variation	Mean peak cavity pressure ($MN\,m^{-2}$)	Standard deviation	Coefficient variation
Virgin (V)	41.47	0.03	0.07	35.89	0.16	0.45
10% Regrind (V-10G1)	41.51	0.06	0.15	35.83	0.25	0.70
25% Regrind (V-25G1)	41.70	0.03	0.08	36.14	0.14	0.39
50% Regrind (V-50G1)	41.97	0.02	0.06	36.81	0.11	0.30
100% Regrind (V-100G1)	42.32	0.04	0.09	37.80	0.11	0.29
Virgin (V)	41.83	0.06	0.13	36.10	0.17	0.47

of increasing mean shot weight and peak cavity pressure. The use of between 10% and 30% of recycled material is regularly encountered in normal injection-moulding practice. The results presented in Figs 1 and 2 show that even at relatively low percentages of recycled material some quite marked effects on shot weight and cavity pressures can be identified. The correlation between the coefficient of variation of shot weight and peak cavity pressure is not so apparent as in the case of the mean values, although the largest variations encountered in shot weights, 0.15 and 0.13 for 10% regrind and virgin material, respectively, are reflected in the value of variations in peak cavity pressure of 0.70 and 0.47, respectively. The increasing trend in shot weight of virgin polypropylene mouldings observed from the beginning to the end of the run corresponded to an overall increase of 0.9% which was reflected by an increase in peak cavity pressure measurements of 0.6%. This phenomenon has been observed by other workers [2] and has been attributed to variations in injection pressure caused by hydraulic oil temperature rises during the course of a run. A slight increase in shot weight over a six hour moulding period was observed in all the runs reported. The trend in Run 1 was by far the most apparent but the cause was not identified.

3.2. Run 2

The experiments reported for Run 1 were repeated for material which had previously been injection-moulded, granulated and then pelletized using an extrusion compounding line into uniformly sized pellets, prior to use as part feedstock for the injection-moulding Run 2. A similar dependence to that reported for Run 1 was recorded for Run 2 and increasing the proportion of recycled material caused on increase in peak cavity pressure and shot weight of the mouldings. The results are summarized in Table IV.

3.3. Run 3

In an attempt to eliminate contamination of feedstock during storage, and to reduce storage costs, the procedure of continuous recycling during production is often employed in commercial practice. Run 3 shows the effect on the variation in shot weight and cavity pressure of continuously recycling at a level of 10%. The composition of the feedstock is shown in Table II which reveals that after only a relatively small number of cycles the composition becomes constant and this is reflected in the values of shot weight and peak cavity pressure shown in Figs 3 and 4 and Table V.

Continuous recycling at the 10% level has little effect upon the values of mean shot weight, mean cavity pressure or on the variation in shot weight with peak cavity pressure. Any increase may have been masked by the slight trend in the increase in shot weight from the beginning to the end of the run.

3.4. Run 4

The effects of incorporating recycled material, in the form of small irregularly shaped particles, which had been recycled two, three and four times prior to this final moulding run, on the shot weight and peak cavity pressure variations are shown in Figs 5 and 6 and in Table VI. The feedstock in this run consisted of virgin granules mixed with 0%, 10%, 25%, 50% and 100% of material which had been recycled twice. In addition 100% of material which had previously been recycled three and four times, respectively, were also used as feedstock. The stepped increases in shot weight and cavity pressures which correspond to increases in the percentage of recycled material in the feedstock were again apparent. More marked changes were recorded when the feedstock material consisted entirely of material which had been recycled two, three or four times, respecti-

TABLE IV Summary of shot weight and cavity pressure data recorded during Run 2 for feedstock containing proportions of pelletized recycled material

Type of feedstock	Mean shot weight (g)	Standard deviation	Coefficient of variation	Mean peak cavity pressure ($MN\,m^{-2}$)	Standard deviation	Coefficient of variation
Virgin (V)	40.98	0.03	0.07	33.27	0.27	0.81
10% Regrind (V-10P1)	41.01	0.03	0.06	33.42	0.30	0.90
25% Regrind (V-25P1)	41.06	0.02	0.05	33.89	0.20	0.59
50% Regrind (V-50P1)	41.21	0.03	0.07	34.84	0.22	0.63
100% Regrind (V-100P1)	41.41	0.02	0.06	37.72	0.36	0.95

vely. The rank in order of increasing shot weight and mean cavity pressure is one. It is evident that a narrowing in the variation of the peak cavity pressure with an increase in the percentage of recycled material of the number of times the feedstock had been recycled was not reflected in the variation of the shot weight distribution. This result was attributed to the occurrence of flashing on moulds prepared from compounds V-100G2; V-100G3; V-100G4 and the contribution of the flash to the measured shot weight. Flashing was unavoidable when the machine conditions previously set for moulding the virgin plastic were used for the moulding of compounds V-100G2, V-100G3 and V-100G4.

3.5. Run 5

In this run the feedstocks used were the same as those for Run 1. However, in Run 5 a 6 mm thick, fan-gated, plaque mould was used rather than the 3 mm thick mould used in Run 1. The results are shown in Figs 7 and 8 and Table VII.

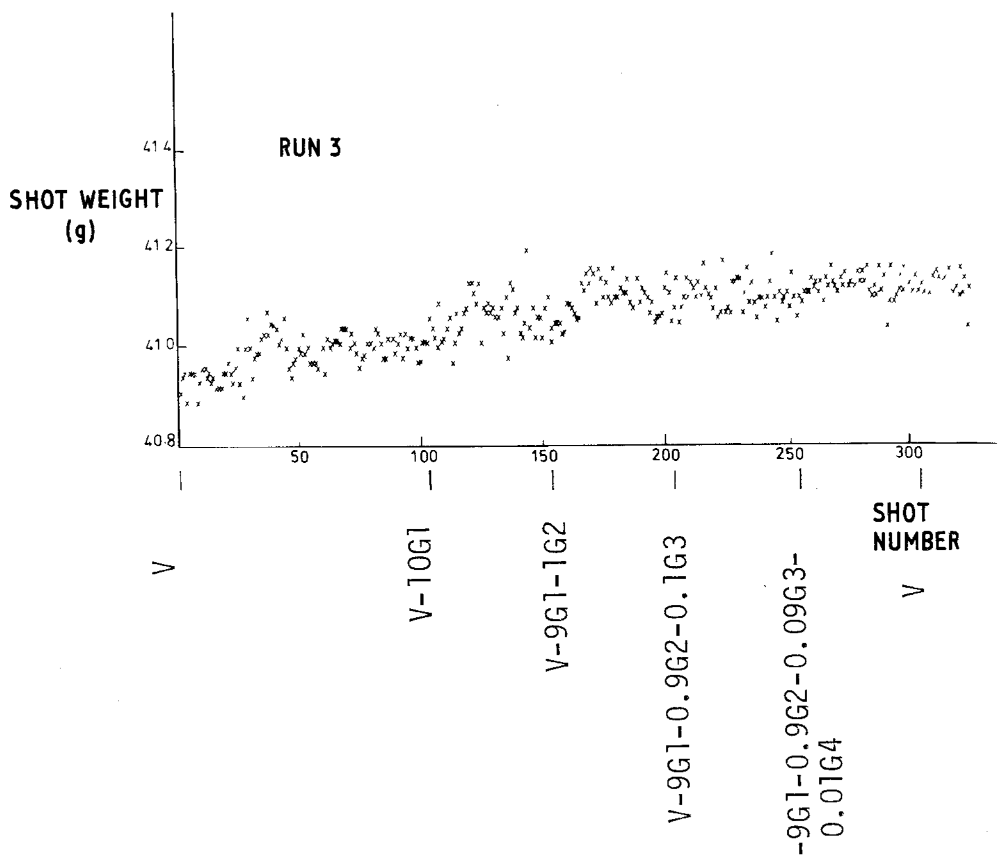

Figure 3 Measured shot weights plotted against shot number for mouldings produced during Run 3.

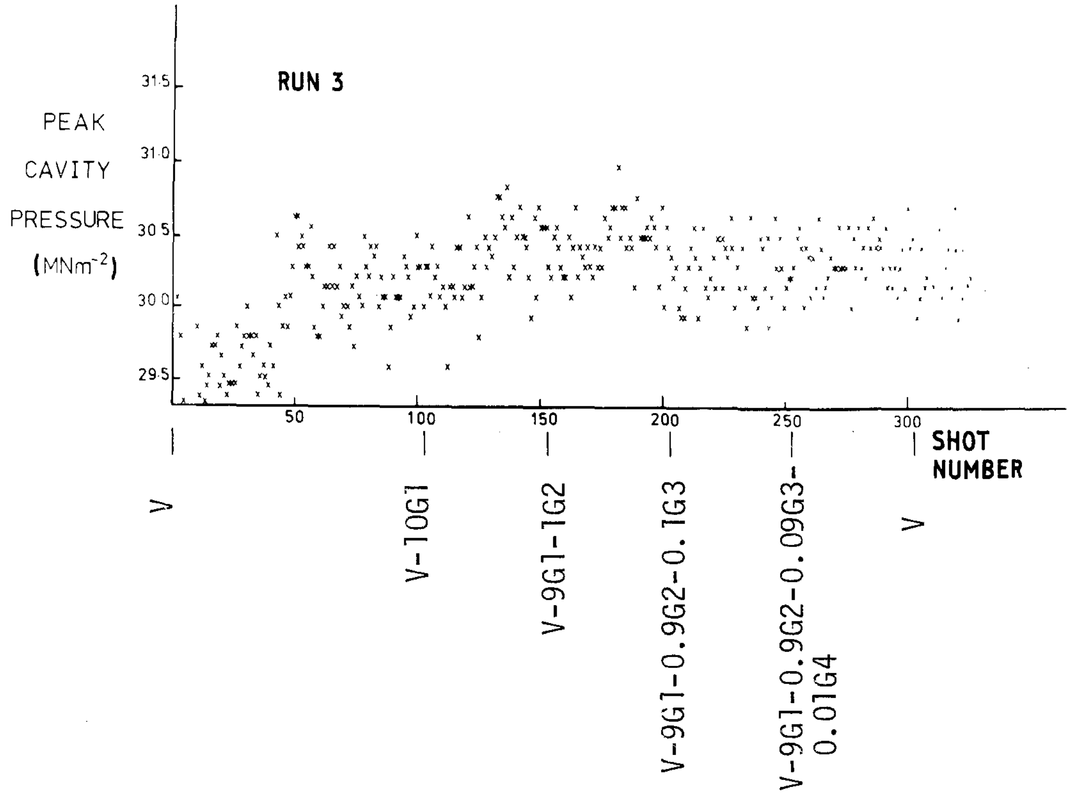

Figure 4 Measured peak cavity pressure plotted against shot number for mouldings produced during Run 3.

The values of the variation in shot weights measured in Run 5 were similar to those measured in Run 1. The dependence of both shot weight and peak cavity pressure on feedstock formulations containing recycled material were also similar for the two moulds although the values for the variation in peak cavity pressure are greater in Run 5 than those encountered in Run 1. This was attributed to the marked differences in mould geometry and the relative positions of the cavity pressure transducers in the two moulds.

3.6. Run 6

The observed changes in shot weight with increasing percentage of reground material could be explained on the basis that the corresponding increase in cavity pressure causes the two halves of the mould to separate during moulding. However the separation of the two halves of the mould during mould filling was not monitored in Runs 1 to 5, and it was therefore necessary to carry out the experiment using a Daniels 350-120 reciprocating screw injection moulding machine with the

TABLE V Summary of shot weight and peak cavity pressure data recorded during Run 3 for feedstocks containing proportions of recycled material which approximately relate to the method of continuous recycling at the 10% level.

Type of feedstock	Mean shot weight (g)	Standard deviation	Coefficient of variation	Mean peak cavity pressure ($MN\,m^{-2}$)	Standard deviation	Coefficient of variation
Virgin (V)	41.01	0.02	0.05	30.15	0.20	0.66
10% Regrind (V-10G1)	41.03	0.18	0.44	30.35	0.26	0.86
10% Regrind (V-9G1-1G2)	41.08	0.05	0.11	30.42	0.20	0.66
10% Regrind (V-9G1-0.9G2-0.1G3)	41.01	0.03	0.08	30.25	0.18	0.60
10% Regrind (V-9G1-0.9G2-0.09G3-0.01G4)	41.11	0.03	0.07	30.31	0.17	0.56
Virgin (V)	41.07	0.02	0.06	30.31	0.21	0.69

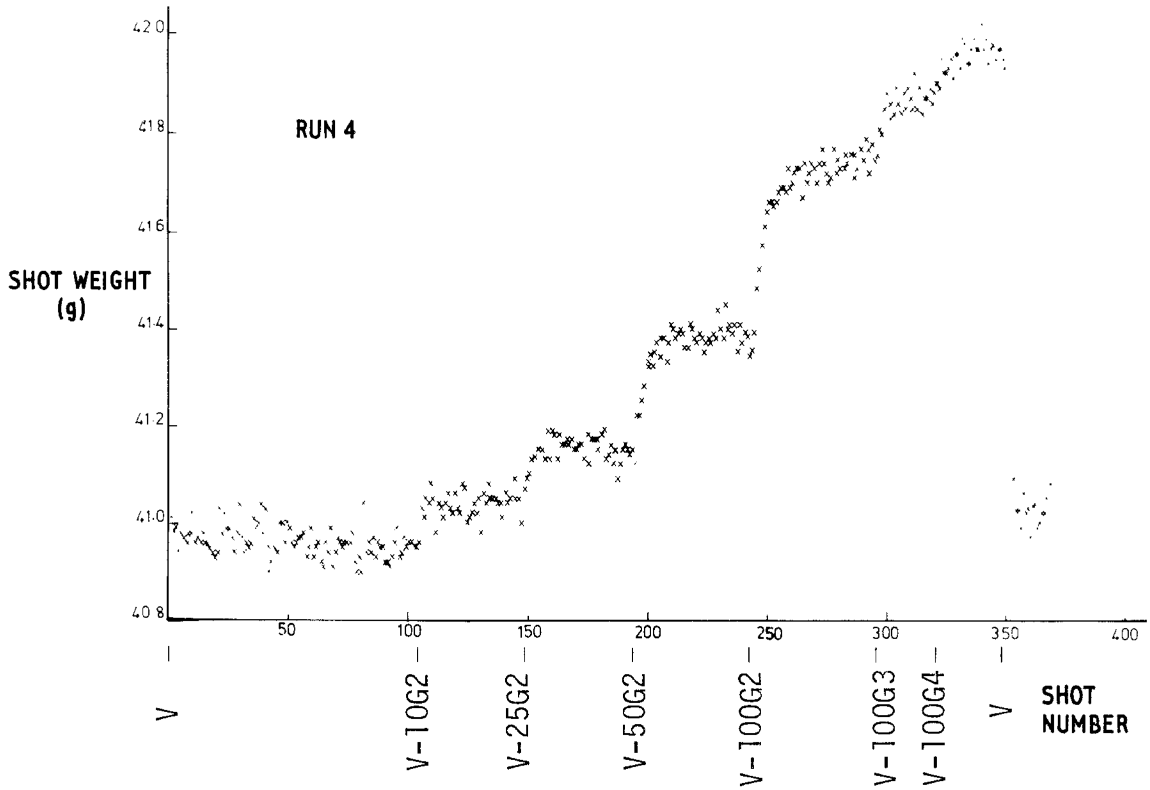

Figure 5 Shot weight plotted against shot number for mouldings produced during Run 4.

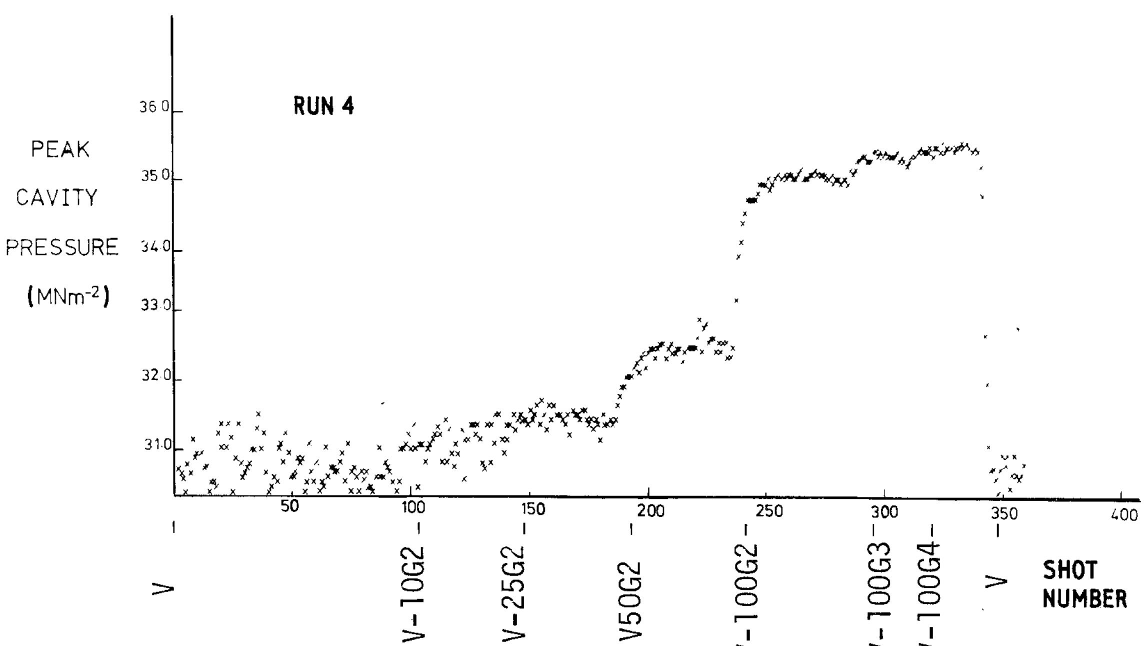

Figure 6 Peak cavity pressure plotted against shot number for mouldings produced during Run 4.

TABLE VI Summary of shot weight and peak cavity pressure data recorded during Run 4 for material recycled more than once

Type of feedstock	Mean shot weight (g)	Standard deviation	Coefficient variation	Mean peak cavity pressure (MN m^{-2})	Standard deviation	Coefficient variation
Virgin (V)	40.95	0.03	0.07	30.59	0.26	0.83
10% Regrind (V-10G2)	41.04	0.02	0.06	31.10	0.23	0.75
25% Regrind (V-25G2)	41.16	0.02	0.06	31.48	0.11	0.36
50% Regrind (V-50G2)	41.39	0.09	0.06	32.50	0.14	0.43
100% Regrind (V-100G2)	41.73	0.03	0.06	34.92	0.06	0.17
100% Regrind (V-100G3)	41.86	0.03	0.06	35.22	0.06	0.16
100% Regrind (V-100G4)	41.97	0.03	0.06	35.35	0.05	0.15
Virgin (V)	41.02	0.03	0.08	30.64	0.25	0.80

aim of identifying the main cause of the stepped increase in shot weight with increasing percentage of recycled material. Virgin plastic and plastic which had been repelletized three and six times were used as feedstock for preparing the 3 mm thick double-gated plaque mouldings required for the experiment and subsequently referred to as Run 6. The optimum moulding conditions identified for virgin feedstock were used for the three feedstock compositions V, V-100P3 and V-100P6. The relative displacement of the two mould halves was measured using a linear displacement transducer and simultaneously recorded together with the cavity pressure and mould temperature. One hundred plaques of virgin material were moulded and discarded prior to recording the relatively small number of measurements of shot weight, cavity pressure and mould thickness shown in Figs 9, 10 and 11, respectively. The measurements of the mould opening revealed that the two halves of the mould separated by less than 0.03 mm during mould filling and that this separation remained constant and was independant of the feedstock used and the accompanying changes in peak cavity pressure. The cause of the recorded steps in shot weight in Runs 1 to 6 when recycled material was introduced into feedstock was therefore found to be not due to mould opening.

Marked changes in the melt flow indices and molecular weights of the feedstock used in Runs 1 to 6 were identified and these are presented in Table VIII and IX. The results show that the incorporation of recycled material in the feedstock causes a reduction in molecular weight which results in more efficient transmission of pressure by the melt as well as a reduction in the melting

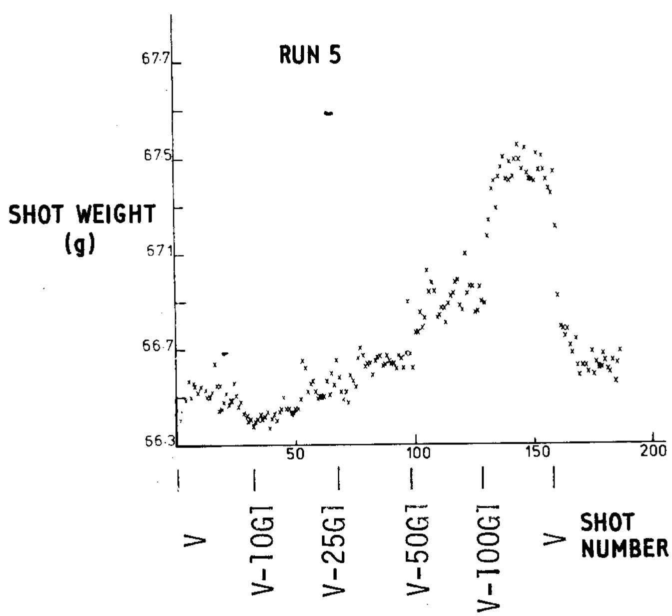

Figure 7 Shot weight plotted against shot number for 6 mm thick mouldings produced during Run 5.

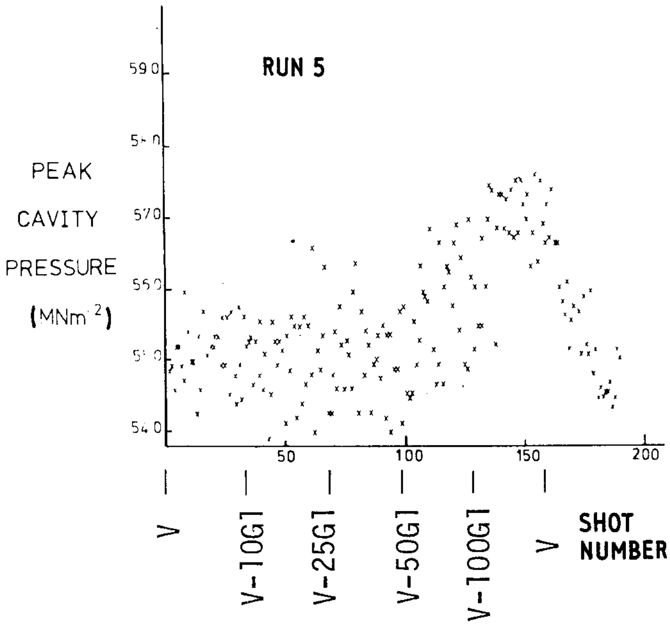

Figure 8 Measured peak cavity pressure plotted against shot number for mouldings produced during Run 5.

point of the polymer. The steps in shot weight recorded in Runs 1 to 6 may therefore be explained on the basis of better mould packing caused by an increase in the efficiency of the melt to transmit pressure from the screw to the mould, and a longer effective hold-on time which results from the longer gate-freezing times associated with the lower melting point recycled plastics compounds.

The effect of increasing the efficiency of pressure transmission and of increasing hold-on time is clearly shown by the cavity pressure profiles in Fig. 12a. When V-100G4 is used as feedstock rather than virgin material the peak cavity pressure increases from 30.59 to 35.35 MN m^{-2} and the hold-on time increases by 3.5 seconds, with resultant increases in shot weight from 40.95 to 41.97 gms. Similar increases were recorded for the extruded and pelletized feedstocks used in Run 6 as indicated by the cavity pressure profiles shown in Fig. 12b. The reported absence of mould opening indicates that the increase in shot weight is mainly a consequence of the longer gate-freezing time with a contribution from an increase in melt pressure transmission.

4. Physical testing

4.1. Impact testing

4.1.1. 3 mm thick mouldings

The plaques prepared during Runs 1 to 4 (the 3 mm thick double-gated mouldings) were impact tested two weeks after completion of moulding. Some of the plaques produced in Run 3 were also impact tested after storage for one year in the absence of any damaging ultraviolet radiation. A test temperature of $-10°$ C was used in all of the impact tests reported in Part 2.

TABLE VII Summary of shot weight and peak cavity pressure data recorded during Run 5 for the 6 mm thick plaque mouldings containing proportions of material recycled once

Type of feedstock	Mean shot weight (g)	Standard deviation	Coefficient variation	Mean peak cavity pressure (MN m^{-2})	Standard deviation	Coefficient variation
Virgin (V)	66.49	0.06	0.09	55.15	0.45	0.82
10% Regrind (V-10G1)	66.54	0.06	0.10	55.08	0.70	1.27
25% Regrind (V-25G1)	66.65	0.02	0.04	55.10	0.62	1.13
50% Regrind (V-50G1)	66.92	0.07	0.10	55.85	0.80	1.44
100% Regrind (V-100G1)	67.46	0.05	0.07	57.06	0.43	0.75
Virgin (V)	66.64	0.04	0.06	55.02	0.54	0.98

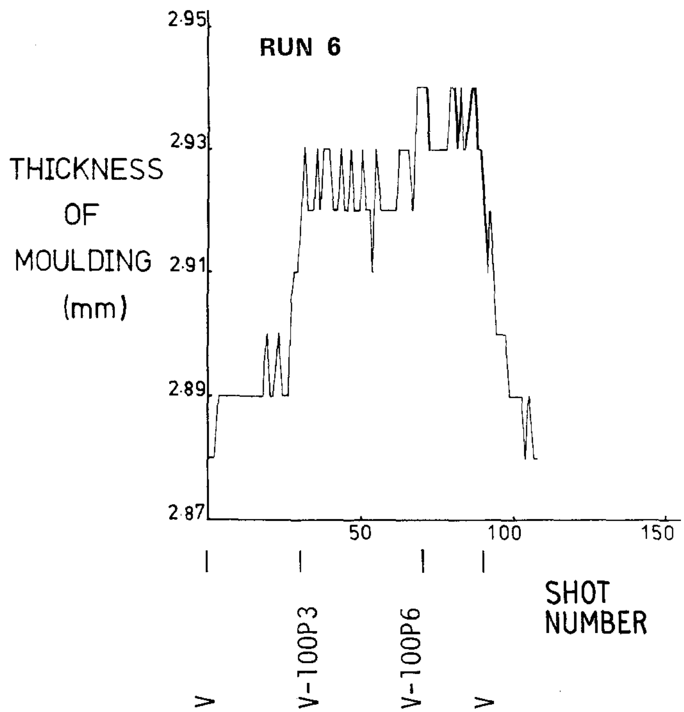

Figure 9 The effect of introducing material pelletized three and six times upon the thickness of 3 mm thick double-gated plaque mouldings.

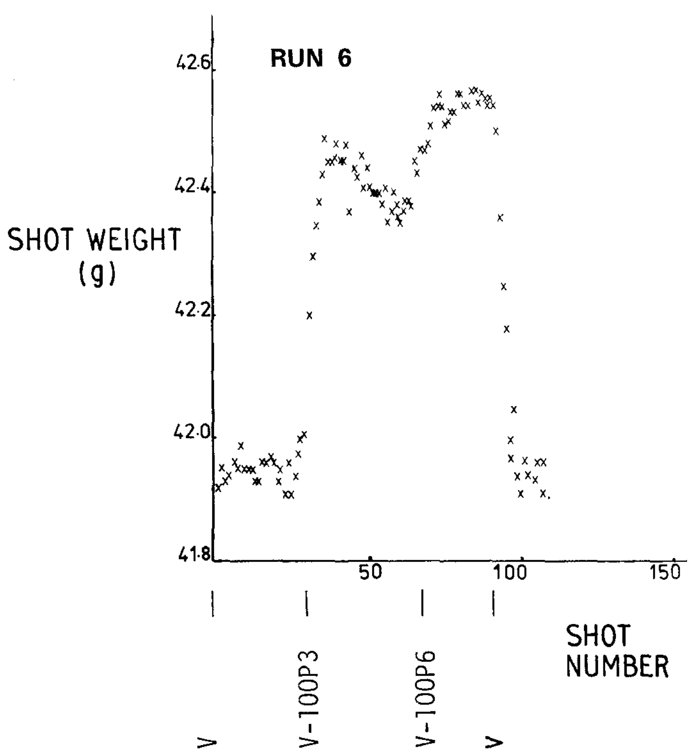

Figure 10 The effect of introducing material pelletized three and six times upon the shot weight of 3 mm thick double-gated plaque mouldings.

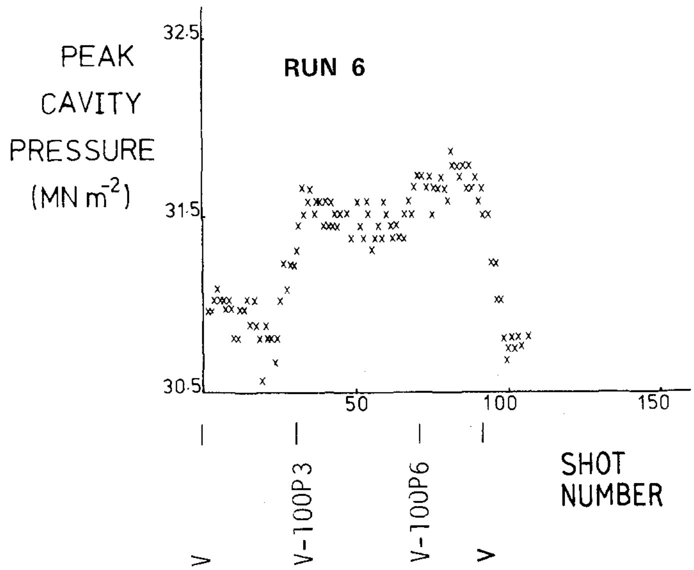

Figure 11 The effect of introducing material pelletized three and six times upon the peak cavity pressure of 3 mm thick double-gated plaque mouldings.

4.1.2. 6 mm thick mouldings

One half of the plaques prepared during Run 5 were impact tested two weeks after completion of moulding. The second half was exposed to ultraviolet radiation on a Marr weatherometer for 1162 hours under a one kilowatt carbon arc prior to impact testing. At least thirty plaques of each composition were tested to give the result shown in Figs 13 and 14. At the test temperature of $-10°$ C all the fractures were brittle and the values reported are accurate, unless stated to ± 1.5 Joules.

The impact test results given in Fig. 13 show that for Runs 1 and 2 the introduction of recycled material up to 100% in the form of granulate or uniformly shaped pellets has little effect on the impact performance of the injection-moulded plaques.

The results for Run 3 show that continuous recycling at the 10% level has little effect upon the impact properties of the plaques tested two weeks after moulding although the results presented in Table X show that a deterioriation in the impact properties of the mouldings is observed after storage for one year in the absence of ultraviolet light.

The impact test results on specimens produced in Run 4 show the effect of increasing the proportion of material recycled twice (V-100G2) as feedstock together with using 100% of material recycled three (V-100G3) and four times (V-100G4). The use of a feedstock consisting of 100% of material recycled twice (V-100G2) results in a marked deterioration in impact properties and a much more marked deterioration when 100% of material recycled three and four times is used as feedstock. The results presented in Fig. 14 show that there is little deterioration in the impact performance of the 6 mm plaque mouldings with increasing the percentage of material

TABLE VIII The melt flow indices of material after injection-moulding up to four times, together with the corresponding molecular weight

Number of passes	Melt flow index (g per 10 min)	Number average molecular weight, M_n (× 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

TABLE IX The melt flow indices of material after extruding and pelletizing up to six times

Number of passes	Melt flow index (g per 10 min)
0	5.6
1	6.8
2	7.9
3	8.9
4	9.6
5	10.0
6	10.9

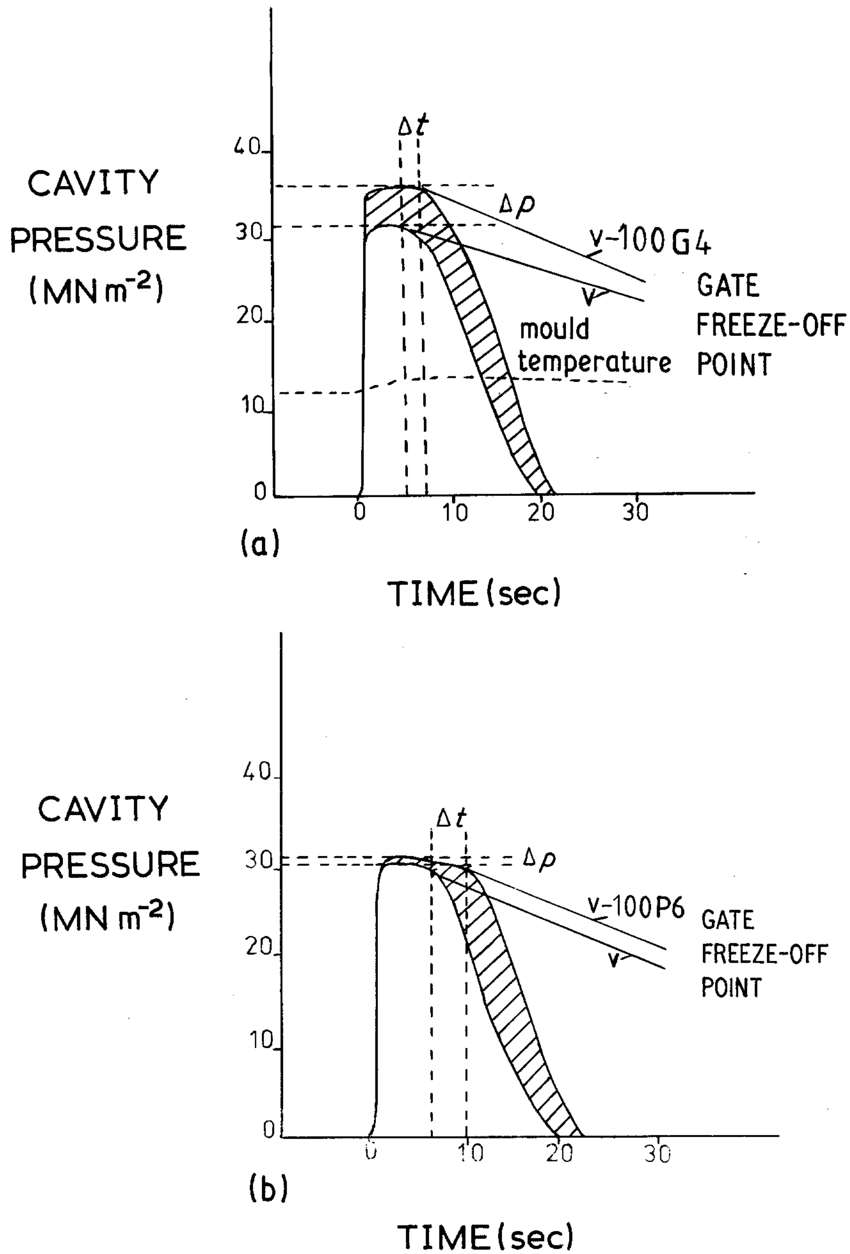

Figure 12 The cavity pressure profiles recorded (a) during Run 4 for virgin (V) polypropylene and 100% of material recycled four times (V-100G4) and (b) during Run 6 for virgin and 100% of material repelletized six times (V-100P6).

recycled once, as measured two weeks after moulding.

Impact tests on the plaques following 1162 hours of artificial weathering give rise to a different result. The impact strength of the weathered plaques increases with percentage regrind up to 50% recycled material but then drops down to the lowest recorded value for 100% recycled material. The impact strengths for all the weathered plaques were lower than for the corresponding unweathered plaques. The weathering resistance of the plaques containing 50% of recycled material was good, and represented a deterioration in impact strength of only a few Joules after the equivalent of one year's natural weathering. The virgin plaques containing 100% recycled material both showed similar weathering characteristics, where the impact strength fell by 14.3 Joules and 15.3

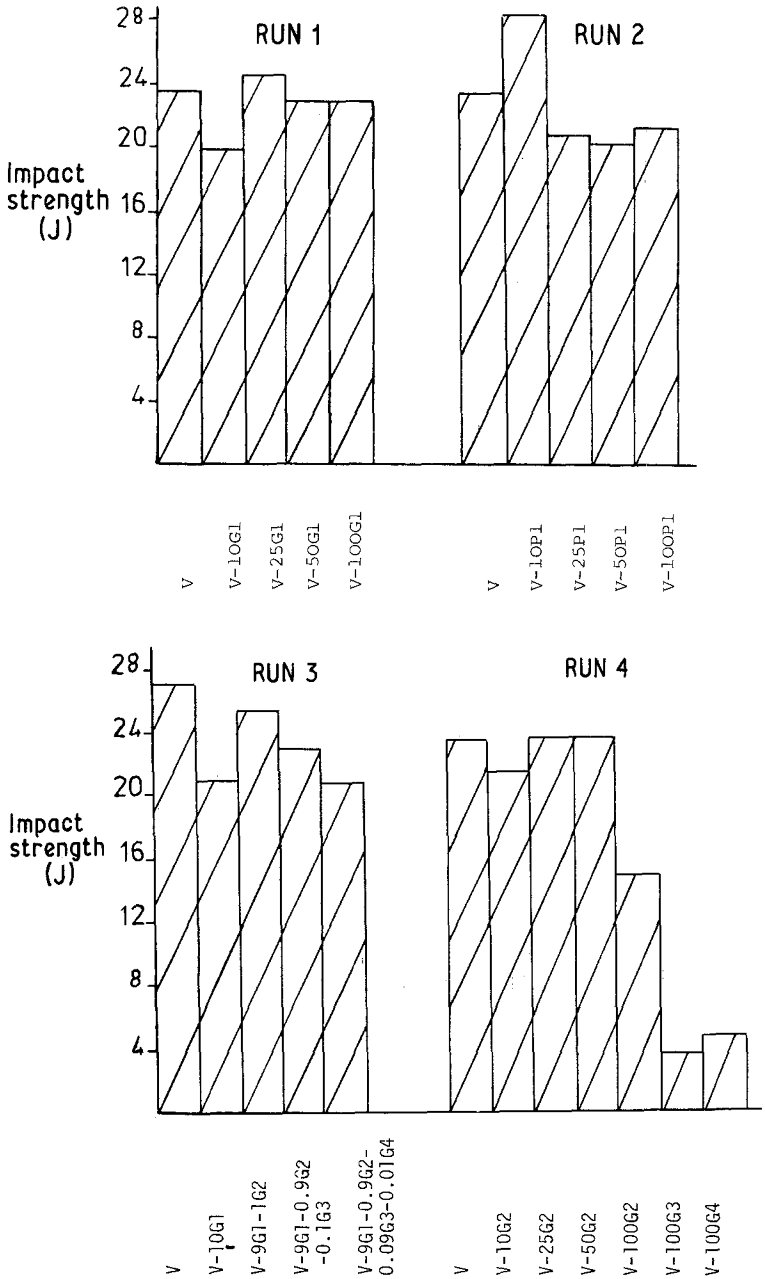

Figure 13 Impact test results for 3 mm thick, plaque mouldings produced during (a) Runs 1 and 2 and (b) during Runs 3 and 4. Test temperature = – 10° C.

Joules, respectively. The virgin plaques which were impact tested after storage for one year in the absence of ultraviolet light, showed an improvement in performance, the impact strength rising from 43.1 Joules two weeks after moulding to 49.0 Joules after storage for one year.

4.2. Tensile testing

Tensile tests according to ASTM D638 at A23° C and using a cross-head speed of 50 mm min^{-1} were carried out on specimens prepared from the 3 mm plaque moulds produced during Run 1 and Run 4. The tensile test specimens were cut from the plaques so that the tensile axis was perpendicular to the injection direction. The test results are given in Table XI and show that there is little change in the yield stress and only a small decrease in the failure stress with an increase in the level of recycling. There is, however, a substantial deterioration in the elongation-to-failure from 568% to 205% which is consistent with the results of the impact tests reported above.

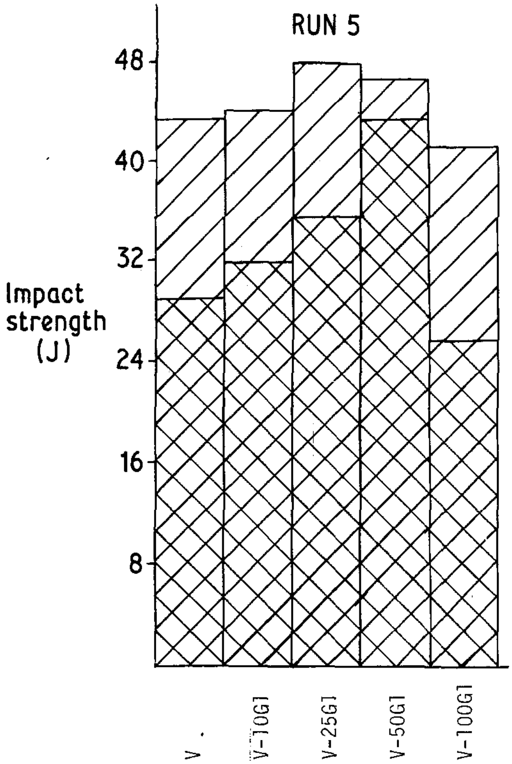

Figure 14 Impact test results for 6 mm thick plaque mouldings produced during Run 5. The cross-hatched areas relate to the impact measures carried out after artificial weathering of the test specimens. Test temperature = − 10° C.

4.3. Microstructure studies

The microstructures of the injection moulded plaques were examined with the aim of providing explanation for the marked changes in mechanical properties reported in Section 3.2., which occur with changes in the composition of the feedstock.

TABLE X Impact strength of mouldings produced during Run 2 as measured after two weeks and one year, respectively

Type of feedstock	Impact strength two weeks after moulding (J)	Impact strength after storage for one year in the absence of ultraviolet light (J)
Virgin (V)	27.2	19.2
10% Regrind (V-10G1)	21.2	16.1
10% Regrind (V-9G1-1G2)	25.6	19.2
10% Regrind (V-9G1-0.9G2-0.1G3)	23.0	19.2
10% Regrind (V-9G1-0.9G2-0.09G3-0.01G4)	21.0	17.5

TABLE XI Yield stress and elongation-to-failure of mouldings produced during Run 1 and Run 4

Type of feedstock	Yield stress ($MN\,m^{-2}$)	Elongation-to-failure (%)
Virgin (V)	27.1	568
100% Regrind (V-100G1)	26.9	447
100% Regrind (V-100G2)	26.9	352
100% Regrind (V-100G3)	26.8	334
100% Regrind (V-100G4)	26.8	205

The main technique used for the characterization of microstructure was microtomy combined with light microscopy. This technique was capable of revealing changes in spherulitic structure caused by changes in processing conditions and compound formulations. Fig. 15 shows a micrograph of a typical section taken from a plane parallel to the injection direction of a 3 mm thick plaque (as shown in Part 1, Fig. 2 [1]). The plaque was made from virgin feedstock. The skin-, shear- and core-zone microstructure as identified by Kantz *et al.* [3] and which is typical of polypropylene injection-mouldings is clearly revealed. The presence of preferred molecular orientation in the mouldings is also shown by the presence of rows of shear nucleated Type III spherulites in the skin-core boundary. The Type III spherulites are negatively bi-refringent and characteristic of the metastable hexagonal crystal form, and although they are found predominantly in the shear-zone they can also be found, especially when there is some degree of molecular orientation, in the core region of the mouldings.

The shear-zone is not readily observable in micrographs taken from a plane perpendicular to the injection direction, because the bi-refringence of the row nucleated spherulites when viewed at an angle perpendicular to the cylindrical axes of the rows differs little from that of the Type I core spherulites which predominate in the core. This is clearly demonstrated in the micrographs shown in Fig. 16, which were taken perpendicular to the injection direction and which reveal little additional information on the microstructure.

The core of the mouldings, which made up the bulk of the volume of the injection-moulded specimens predominantly contained the positively bi-refringent Type I spherulites, which are characteristic of the stable monoclinic form. The presence of preferred molecular orientation gives rise to the formation of rows of Type III spheru-

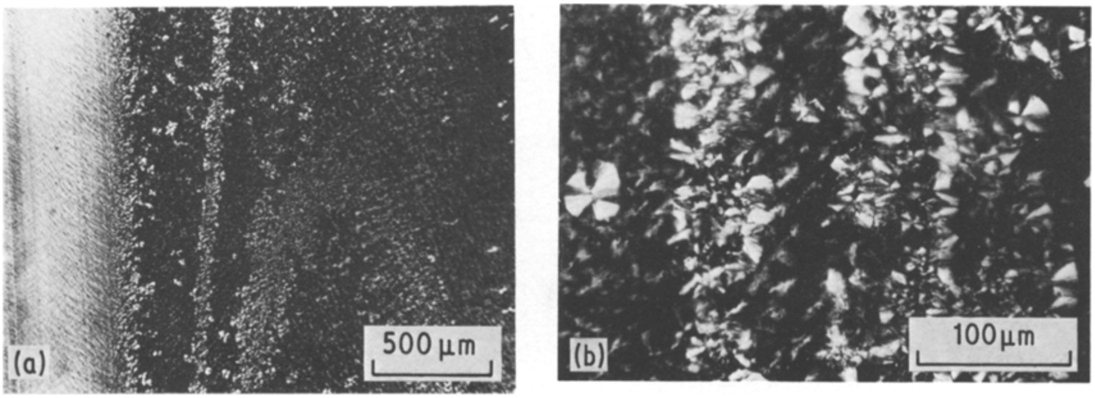

Figure 15 (a) Micrograph of typical section taken from a plane parallel to the injection direction of a 3 mm thick plaque moulding, revealing the skin-zone–shear-zone–core-zone microstructure as identified by Kantz *et al.* [3] and typical of polypropylene injection-moulding. The presence of molecular orientation in the mouldings is also indicated by rows of shear nucleated Type III spherulites; (b) Rows of shear nucleated Type III spherulites (shown in (a)) viewed in more detail.

lites that extend into the core region. The spherulites in each row are approximately the same size, as revealed in Fig. 15, and in general are larger than other negatively bi-refringent Type III spherulites which occur more randomly in the shear-zone.

Fig. 17 shows the microstructures of 3 mm thick plaques and the effects of incorporating material which was recycled one, twice, three and four times at the 100% level as feedstock. Introducing 100% of material recycled once as feedstock caused the thickness of the skin layer to be reduced whilst the character of the layer remained unchanged. One of the most marked changes in microstructure which occurred with changes in feedstock was connected with the presence of Type III spherulites. Row nucleated Type III spherulites were present in the core of plaques prepared from 100% virgin feedstock and in feedstock containing 10% and 25% of plastic recycled once. In the latter two cases the row-nucleated spherulites were present, as in the case of the virgin material, but to a lesser extent. Row-nucleated Type III spherulites were not observed in the plaques prepared from material recycled once at the 100% level or when the feedstock contained 50% virgin/50% recycled-once material. The decrease in the extent of the Type III spherulites can be explained on the basis of the decrease in shear which occurs in the melt, and accompanies the reduction in molecular weight of the co-polymer with recycling.

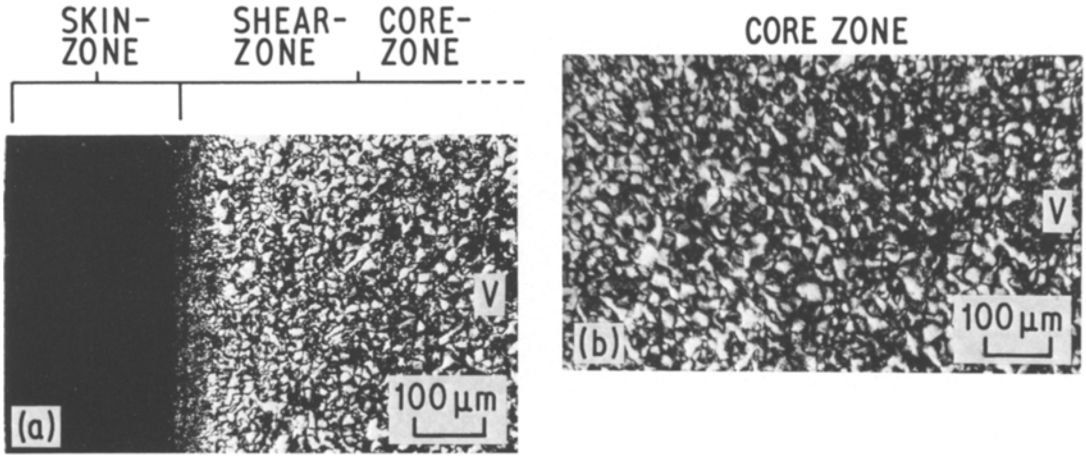

Figure 16 Micrographs taken from a plane perpendicular to the injection direction from 3 mm thick double-gated plaque moulding prepared from virgin feedstock (V), (a) showing the skin-core region and (b) the core region.

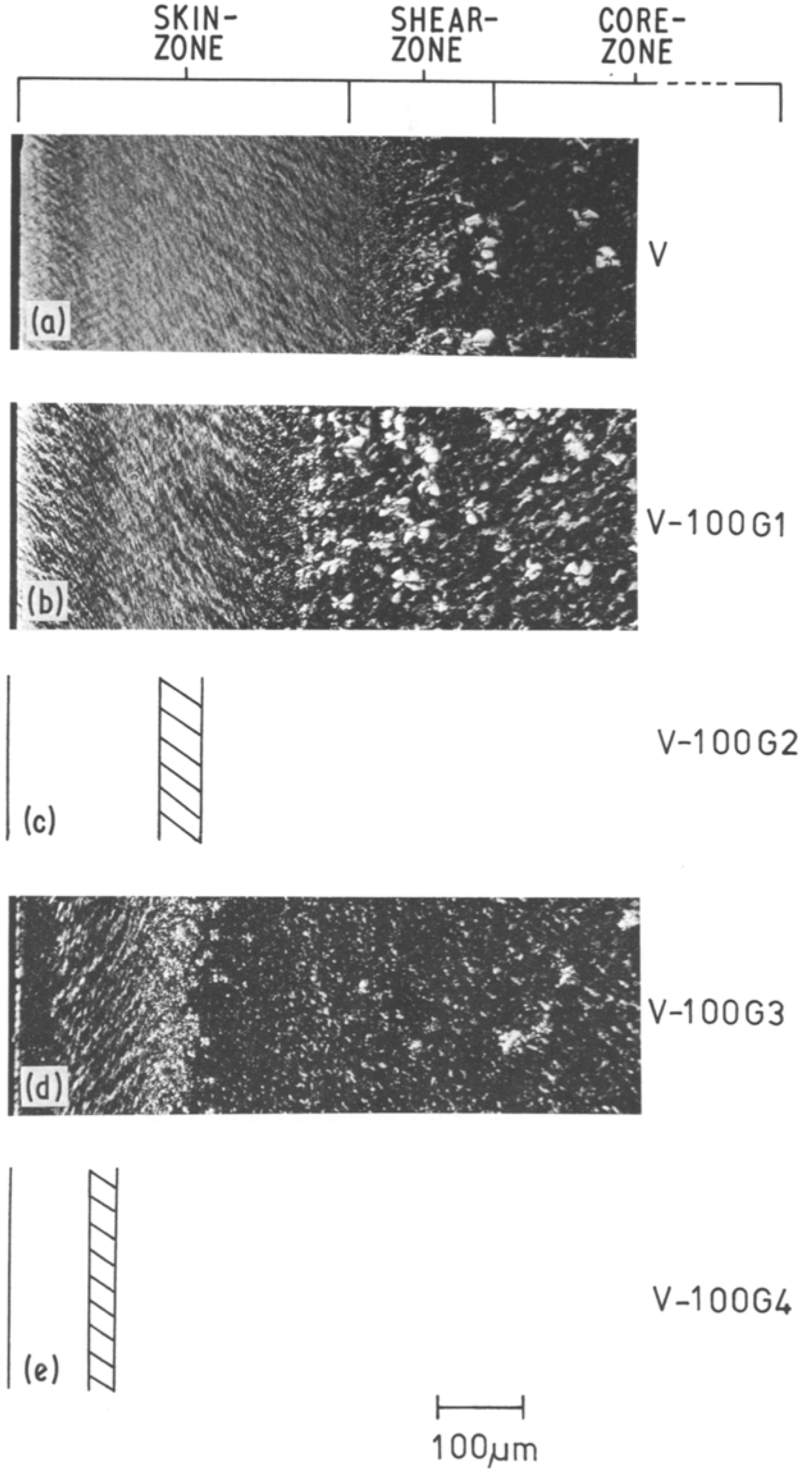

Figure 17 Micrographs taken from a plane parallel to the injection direction showing (a) virgin material and the effect of incorporating 100% of material recycled (b) once (V-100G1), (b) twice (V-100G2), (d) three times (V-100G3) and (e) four times (V-100G4) upon the skin-core microstructure of 3 mm thick plaque mouldings.

Thermal degradation of the co-polymer during recycling causes its molecular weight to be reduced by the mechanism of chain scission and consequently a reduction in the relaxation times of the polymer chains. In mouldings prepared from compounds containing recycled material the alignment of chains in the injection direction will be less pronounced, and the thickness of the skin layer will be reduced, as will the proportion of molecular chains which are aligned parallel to the injection direction and which tend to act as nucleii for row nucleation of the Type III spherulites.

The large reduction in molecular weight accompanying the second recycle is reflected in the morphology of the plaques prepared from feedstock that has been recycled twice. The character and thickness of the skin layer is substantially different from plaques prepared from virgin material. The difference in bi-refringence between the skin and core becomes much less apparent and there is some evidence for the development of a spherulitic structure in the skin. The thickness of the skin is reduced and the size of the negatively bi-refringent Type II spherulites is also reduced.

The microstructure of the plaques prepared from material recycled three times shows little change from that of plaques prepared from material recycled twice. This is also reflected in the values of the number average molecular weights, M_n, of the material in these plaques, both of which are approximately 2.5×10^4.

The core-zone of the moulding, which makes up the bulk of the volume in the injection-mouldings, contained randomly nucleated positively bi-refringent Type I spherulites identified by Padden and Keith [4]. The size of these spherulites was larger than the negatively bi-refringent Type III spherulites and in general the effect of recycling was to reduce the size of the core spherulites as well as that of the Type III spherulites. The decrease in size of the core spherulites may be due either to impurities introduced by recycling or to an increase in the proportion of low molecular weight species, which will cause a reduction in the growth rate of the spherulites. The small difference between the measured impact properties of the mouldings containing up to 100% of material recycled once and those prepared from virgin feedstock was unexpected. This result can be explained on the basis of the reported differences in microstructure of the mouldings. The adverse effect of the reduced skin thickness on mouldings containing recycled plastic is compensated for by the reduction in molecular orientation and the proportion of Type III spherulite which resulted from the presence of lower molecular weight polymer. It is unlikely that the reduction in M_n from 5.24×10^4 to 4.32×10^4 would be sufficient to cause a marked change in the impact properties of the equiaxed structure of the core.

In the subsequent recycle, however, the number average molecular weight is halved from 4.32×10^4 to 2.48×10^4, and this reduction does have marked effects upon the microstructure and impact properties of the mouldings. The character of the skin layer in the plaques prepared from virgin and 100% recycled-once material feedstock consisted of molecules that were highly orientated in the injection direction, as shown by the uniform extinction of the first-order white polarization colour when the specimens were rotated in the plane of the microscope stage. In plaques prepared from material recycled more than once at the 100% level there is some evidence for the development of a spherulitic structure as well as a marked reduction in the thickness of the skin layer. The reduction in molecular weight of the recycled polymer together with the accompanying change in thickness and character of the skin layer combine to reduce the impact strength of the mouldings. As the mouldings become more brittle the effect of any impurities present will become increasingly more important and would contribute to the sharp fall-off in the impact properties of mouldings prepared from material recycled four times.

5. Concluding remarks

The main results of the systematic investigation of the effect of incorporating recycled material into polypropylene feedstock for injection-moulding as reported above are as follows.

(a) The use of up to 100% of recycled polypropylene compound as feedstock material does not lead to a decrease in impact properties of injection-mouldings, but tends to cause an improvement in impact performance. These results were based on the use of compounds which were carefully prepared to avoid the inclusion of impurities, and set processing conditions were used throughout the reported work so that any changes in properties were caused by differences in compound formulation.

(b) Recycled material both in the form of pellets and in a granulated form was used as feedstock and it was shown that particle shape caused no change in the standard deviation of shot weights.

(c) The introduction of recycled material into the moulding compound caused substantial changes in microstructure of the injection-mouldings for a fixed set of processing conditions. The main microstructural features to change with the increasing proportion of recycled material were the skin thickness, which decreased and the size and proportion of Type III spherulites. The incidence of row-nucleated Type III spherulites decreased with increasing proportion of recycled material and this effect was proposed to account for maintenance of impact properties and compensate for the decrease in molecular weight and moulding skin thickness.

(d) The standard deviation of the injection-moulding shot weights was independant of feedstock material although the shot weight increased with increasing proportion of recycled material. The results indicated that for the work to be reported in Parts 3 and 4 there was little need for a detailed statistical analysis of moulding reproducability.

(e) The standard deviation of the peak cavity pressures decreased with increasing proportion of recycled material with a corresponding increase in shot weight.

(f) The increase in shot weight of the injection-mouldings with increasing proportion of recycled material was explained mainly on the basis of a longer effective hold-on time caused by the reduction in molecular weight of the polypropylene feedstock.

(g) The use of 100% of material which had been recycled two or more times prior to the final conversion process lead to a substantial decrease in impact strength.

(h) Successive recycling results in a marked change in molecular weight, moulding skin thickness and impact properties and would not normally be considered appropriate in commercal practice.

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