RECENT DEVELOPMENTS IN HOT COMPACTED FIBRE COMPOSITES

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SUMMARY
In this paper recent research on the hot compaction process developed at Leeds University is presented. In this process, thick section homogeneous sheets of fibre composites are produced by melting a thin skin on the surface of each fibre, which on cooling recrystallises to bind the structure together to produce a consolidated, single material, composite

For automotive applications, hot compacted sheet based on polypropylene (PP geotextile fabrics) shows an excellent combination of mechanical properties, especially low temperature impact strength. A range of weave styles and cloth weights have been compared and properties such as thermal expansion and high temperature creep studied. A comparison with isotropic random mat short glass fibre reinforced PP and Twintex unidirectional glass fibre will be presented.

KEYWORDS
Fibre composites, hot compaction, polypropylene, polyethylene

INTRODUCTION
The essential feature of the hot compaction process developed at Leeds University (British Patent No. GB 2253420) is that the matrix phase of the composite is produced by melting a thin skin on the surface of each fibre, which on cooling recrystallises to bind the structure together to produce a consolidated composite. The compacted material is therefore composed of a single material type, with molecular continuity (i.e. excellent bonding) between the fibre and matrix phases. The properties of the compacted sheets rely on retention of the preferred molecular orientation of the original starting materials, which are normally in the form of highly oriented fibres or tapes. This aspect of enhanced properties through controlled molecular orientation is often termed 'self -reinforcement'.

Previous work has concentrated on the hot compaction of highly oriented thermoplastic fibres, and materials studied have included melt spun polyethylene [1-3], gel spun polyethylene [4], polyethylene terephthalate [5] and cross-linked melt spun polyethylene [6]. In the last couple of years the work has concentrated on the compaction of woven oriented polypropylene tapes [7-8].
The most recent research has focused on several major issues. First, there is the requirement to define protocols for the hot compaction process, both in terms of the temperature/pressure profile required and the most satisfactory starting fibres and fabrics. This involves a basic

understanding of the melting and recrystallisation and possibly the annealing (changes in molecular structure) which occurs as a result of the compaction process. Secondly, there is the requirement to define the physical properties of the compacted sheets wherever possible on the basis of ASTM standards but also including other tests, such as high temperature creep, to assess the practical engineering possibilities for these new materials. Finally, there is the issue of postforming and developing tests to assess this aspect of the sheet behaviour which is vital for many applications. These three aspects of the research on hot compacted polypropylene sheets will be discussed in this paper.

EXPERIMENTAL

Our research has shown that hot compacted sheet can be made by either a batch process using matched metal moulds in a hot press or using an autoclave, or continuously using a double belt press. For the majority of the investigations detailed here, material made by the autoclave route was used: there were three main reasons for this choice. First, it is possible to make substantial sized sheets (2m x 5m) so that realistic parts can be made for test purposes and even initial production purposes. Secondly, the amounts of material required in preliminary tests to establish satisfactory production procedures (temperature, pressure, times) is not excessive. Finally, it was desirable to explore the viability of a single step compaction procedure at a single compaction pressure, in contrast to the initial research studies, where a more complicated procedure was used with a low pressure compaction stage followed by a second high pressure compaction stage. When thicker or thinner samples were required for testing purposes, most notably for the notched Izod impact tests (thicker) or for creep tests (thinner), they were made in a hot press using matched metal moulds.

For both the autoclave route and the hot press route the processing conditions were very similar. First the required number of layers of the woven PP cloth are cut and stacked in position. At this point a pressure, termed the compaction pressure, is applied to the assembly: the assembly is then heated to the compaction temperature, which is nominally 184°C. Once the material reaches the compaction temperature it is left to soak to allow even temperature and melting to be achieved throughout the stacked layers: For the autoclave route the soak time is one and a half hours while for the hot press route a soak time of 10minutes is used. Finally the assembly is cooled to 110°C and the compacted sample can then be removed. In this study samples were made in the autoclave using a compaction pressure of 100psi and in the hot press using either 100psi or 400psi. The two starting materials chosen for work were both taken from the AMOCO Geotextile (PROPEX) range. The first, code 6082, was a plain weave and had a fabric weight of 210 g/m2: the second, code 6088, was a twill weave and had a fabric weight of 500g/m2.

RESULTS

EFFECT OF WEAVE STYLE ON INTERLAYER ADHESION AND MODULUS

In the compaction process there is always a trade-off between the development of inter-fibre or inter-tape bonding due to selective surface melting, and a fall in the mechanical properties of the final sheet due to a loss of molecular orientation associated with this melting. In the previous compaction studies, where unidirectionally arranged fibres were used, this 'optimum compaction temperature' was established by measuring the longitudinal modulus of the compacted sheets in the fibre direction and the transverse strength of the sheets perpendicular to the fibre direction. For the current study, where woven tapes were used, the optimum temperature was found by measuring the in-plane tensile modulus of the sheets and the interlayer adhesion of the sheets. The interlayer adhesion was measured using a T peel test (ASTM D1876) and employed samples compacted from two layers of the woven PP material.

During lay-up, a piece of 10μm thick aluminium foil was placed between the two layers of cloth, at one end of the sample: after compaction this foil acts as a starter crack for the peel test.
The compacted samples (autoclave process - 100psi compaction pressure) showed a more or less constant peel load, with an average value of 12.5 ± 2.5N for the 6088 weave style and 25 ± 4N for the 6072 weave style. The 6088 weave style has a much rougher surface and it is possible that a compaction pressure of 100psi is not sufficient to flatten the weave and produce the best bonding. In order to test this proposition, samples were made in the hot press, using the 6088 weave style, at a higher compaction pressure of 400psi. The value of the average peel load for this higher pressure was 20 ± 3N, an increase on the autoclaved material. These experiments showed that the peel force depends on the weave style and for the 6088 style it depends on the compaction pressure. The results suggest that optimum interlayer adhesion will require a compaction pressure greater than the 100psi that can currently be achieved in the autoclave.
The tensile modulus of the 6082 material was measured to be significantly higher than that of the 6088 material: 3.5 GPa compared with 2.5GPa The difference in values is a consequence of the difference in the tensile modulus of the starting tapes which make up the two weave styles: the 6082 contains stiffer (i.e. more highly oriented) tapes.

NOTCH IMPACT TESTS

Notched Izod impact tests were undertaken, following the protocol of ASTM D256, using a Rosand Instrumented Falling Weight machine: the tests were carried out at a speed of 3.46m/s using a 25kg falling weight. As 6mm thick samples were required for this test, they were produced, as described above, using the hot press route: a compaction pressure of 100psi was used. Twelve samples were tested for each weave style and each testing temperature (+20°C and -40°C). The results should be seen as minimum values as the samples did not fail completely.
The results of the impact tests are shown below in Table 1.

Fabric style	Compaction conditions	Impact Energy (J/m) +20°C	Impact Energy (J/m) -40°C
Plain weave (6082) 210 g/m2	184°C/100psi	2726 ± 240	5225 ± 1228
Twill weave (6088) 500 g/m2	184°C/100psi	2848 ± 610	5428 ± 378

Table 1: Notched Izod impact results.

Very high values of the impact energy were obtained and it is particularly notable that these values are substantially greater at -40°C than at +20°C i.e. there is no ductile-brittle transition in these sheets in contrast to the behaviour of isotropic PP sheet.

THERMAL EXPANSION MEASUREMENTS

The thermal expansion behaviour of the hot compacted PP sheets was determined over the temperature range -40 to +80°C, using a custom built facility [9]. For these tests only the 6082 weave style was used: the results are shown in Figure.1.

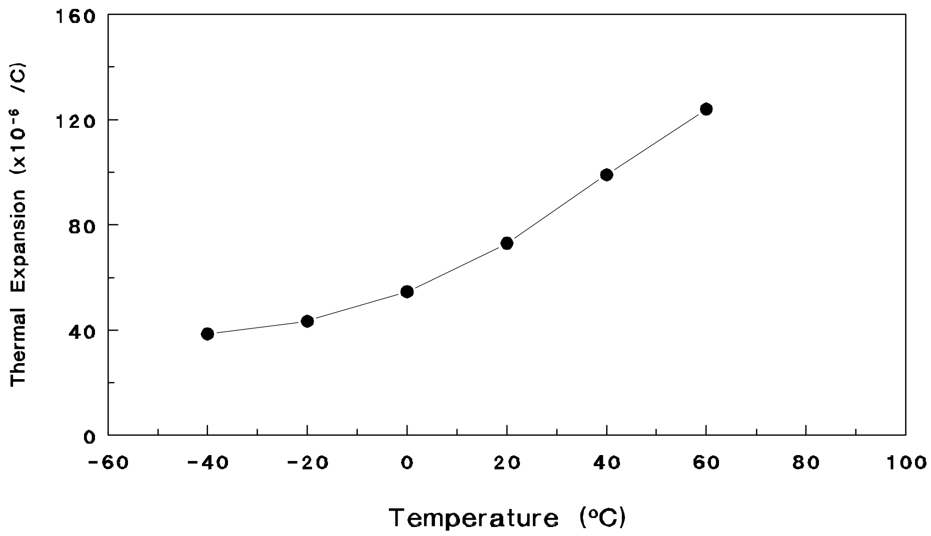

Figure 2: Thermal expansion of compacted PP (6082 style) vs temperature

For comparative purposes, measurements according to ASTM D690, which require data over the temperature range -30 to +30°C, are shown in Table 2, together with reported results for other materials. As would be anticipated, the thermal expansion coefficient for the compacted PP sheet lies between that for isotropic PP and glass-fibre reinforced PP (Twintex).

Material	Compacted PP (6082)	Isotropic PP	Twintex Glass/PP	Stainless steel	Mild steel
α ($x10^{-6}$ /°C)	73	96	21	15	12

Table 2: Thermal expansion of compacted PP sheet and other materials.

CREEP EXPERIMENTS

Tensile creep measurements have been undertaken on hot compacted PP sheets (1 ply thick) in the two directions corresponding to the weft and warp directions in the original fabric: 6082 style. The creep tests were performed using a dead loading creep apparatus which was designed and built in the Leeds laboratory. Specimens of dimensions 80x2mm were placed in an environmental chamber where the temperature can be maintained within ±0.5°C between +20 and +100°C. Typical results for loading at a nominal stress of 10MPa are shown in Figure 3: results are shown for test temperatures of +20, +70 and +100°C.

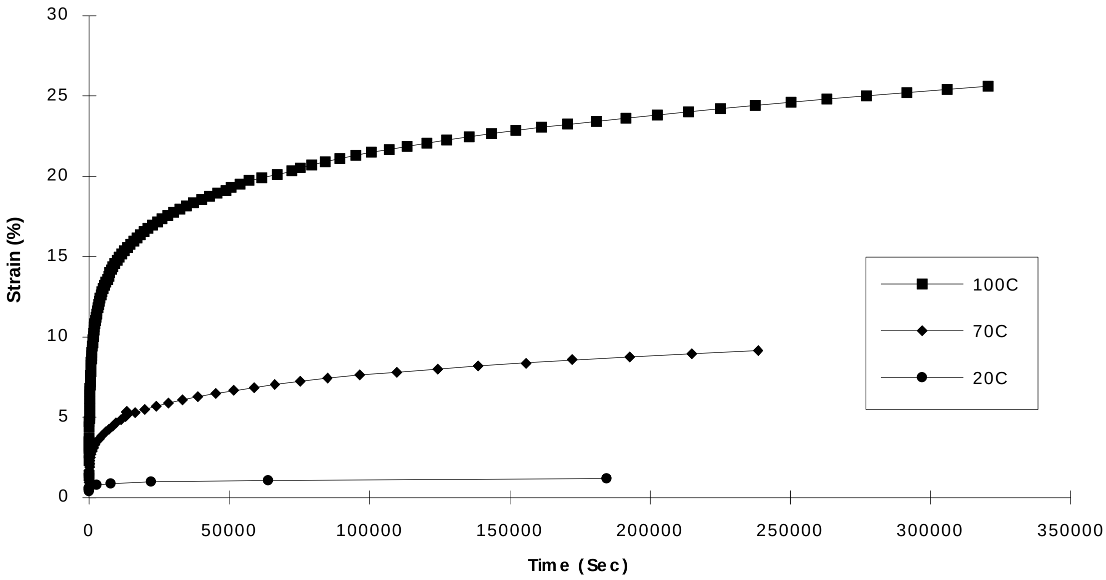

Figure 3: Tensile creep curves for compacted PP sheets at 20, 70 and 100°C

These results show that creep at room temperature is very low but suggest that 70°C would represent a practical limit for the application of the hot compacted PP sheet.

POSTFORMING EXPERIMENTS

A major advantage of the hot compacted sheets is that they can be postformed into the final shape for specific applications. This is most simply undertaken by compressing in a matched metal mould and preliminary trials showed that the thermoforming temperature should be in the range 10-40°C below the compaction temperature. A study has been undertaken to determine the key parameters for successful postforming and to explore the relationships between the postforming behaviour and the mechanical properties of the hot compacted sheet.
For this study a hemispherical matched mould was constructed which fitted into the oven of an Instron tensile testing machine. A spring loaded outer holding plate was employed which was designed to make contact with the sheet before the male part of the mould, as shown below in the schematic diagram. By varying the spring pressure the holding plate was able to either clamp the compacted sheet completely, or with less pressure, could allow the sheet to move into the mould in a controlled fashion. This stamp forming rig was based on the design of Hou [10]. Practical thermoforming operation was simulated by lowering a stainless steel hemisphere into the mould under controlled conditions.

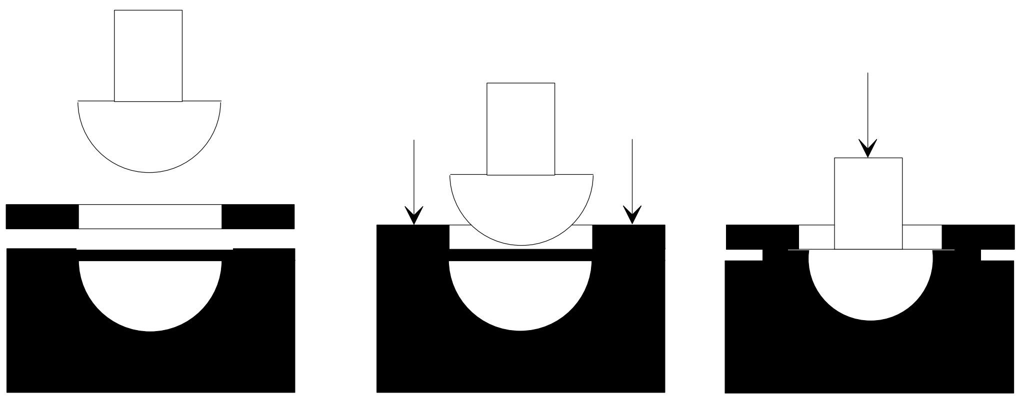

Figure 3: Thermoforming scheme for compacted PP sheets

So far, these experiments have been confined to hot compacted PP sheet only. The following parameters were varied:

(1) Temperature: the hot compacted sheet was placed in the oven, left for 5 minutes to come to temperature, prior to conducting tests at temperatures in the range 140°C - 175°C.

(2) Testing rate: closing speeds between 2mm/min and 500 mm/min were investigated: a typical industrial closing speed for loudspeaker cones is 250 mm/min.

(3) The sheets were allowed either to flow into the mould or gripped completely around their circumference.

(4) Different weave styles were tested with the AMOCO Geotextile fabrics range.

The tests showed that if the compacted sheet is unclamped i.e. free to flow into the mould in a controlled fashion, successful components can be made under all conditions within a wide range of temperatures and rates, although it is best that the temperature is in the range between the melting of the recrystallised skin at ~162°C and the melting point of the fibres at ~176°C. This postforming window is clearly identifiable in the DSC trace shown in Figure 4

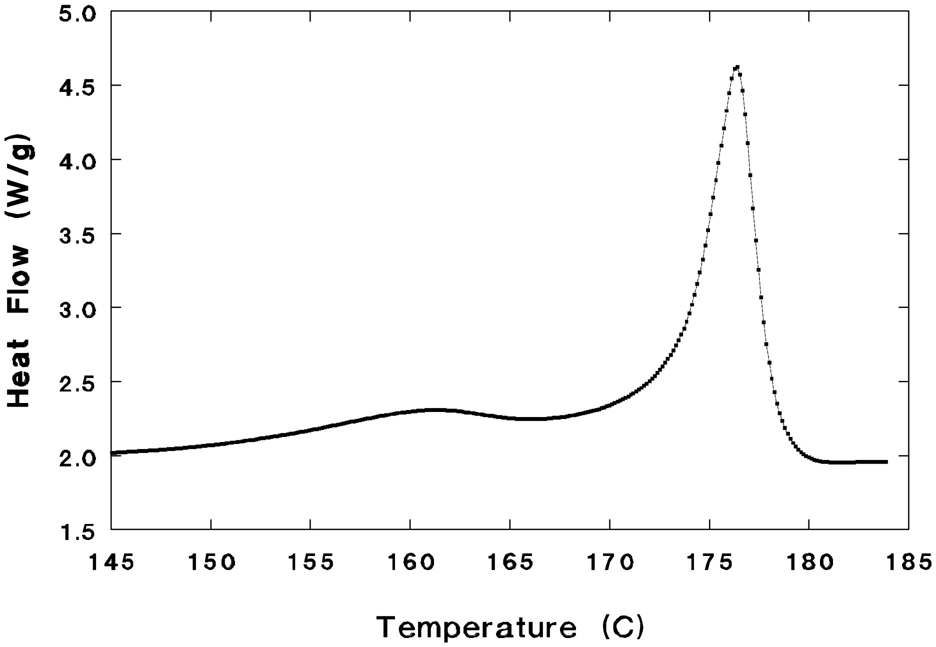

Figure 4: Melting behaviour of compacted polypropylene (6082 Style)

Heating above 162°C allows the second phase to become molten, thereby permitting interply and intraply shear to occur. The worst temperature for postforming is just below the melting temperature of the second phase (i.e. around 160°C) when this phase is very weak and breaks easily, but is not molten and so will not rebond on cooling. Samples thermoformed around this temperature showed significant interply delamination. Thermoforming above this temperature allows the second phase to melt and to form a strong bond on cooling. If only small strains are required (~10%) then a temperature of 150°C is optimum.

Successful postforming depends on all the variables (temperature, closing rate, weave style etc.) to some degree. It was considered of interest to attempt to relate the postforming process to the deformation of the compacted sheet, in the first instance, by determining the tensile stress-strain curves for the compacted woven sheets at high temperature. As shown in Figure 5, these stress-strain curves show several notable features, compared to a standard isotropic material which would be expected to show a clear yield point, followed by a load drop associated with thinning of the material.

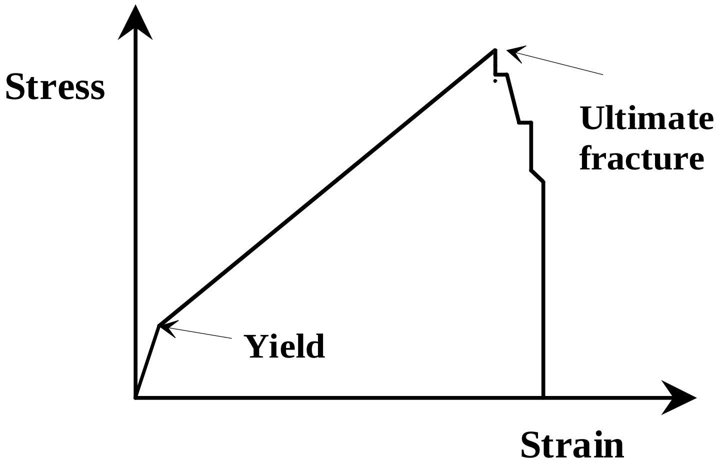

Figure 5: A schematic of the tensile stress/strain curve for compacted PP.

First, there is steep initial slope, relating to the initial modulus of the sheets, followed by the appearance of a yield point at a low strain (~5%). After this yield point the stress/strain curve continues to rise sharply, but with a lower slope until ultimate fracture occurs, which is at a much higher strain of around 60%. The slope after yield (strain hardening region) reduces as the temperature is increased and the maximum strain range for postforming increases with temperature, as shown in Figure 6 below.

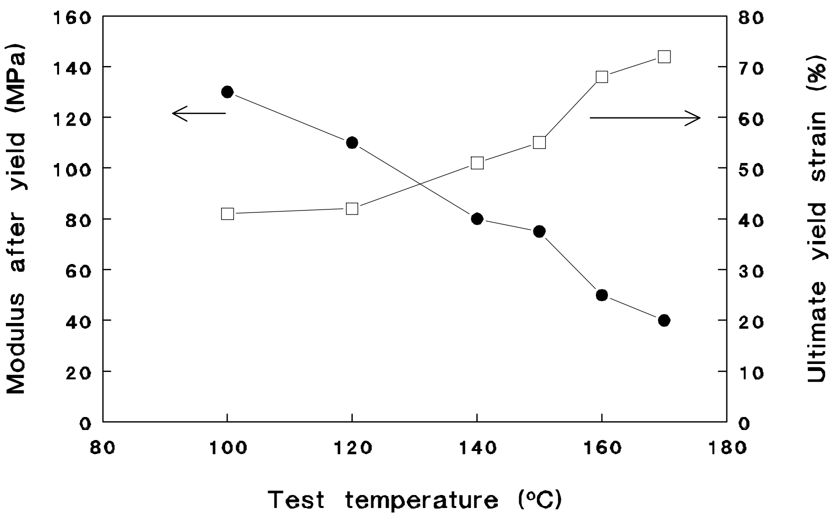

Figure 6: Effect of test temperature on modulus after yield and ultimate yield strain

The forming force is not very sensitive to closing speed but is very sensitive to temperature as are the tensile stress-strain curves. As would be expected, the force required to form the sheet is directly related to the tensile stress-strain behaviour and Figure 7 shows that there is a direct correlation between the forming force and the tensile force at 10% strain.

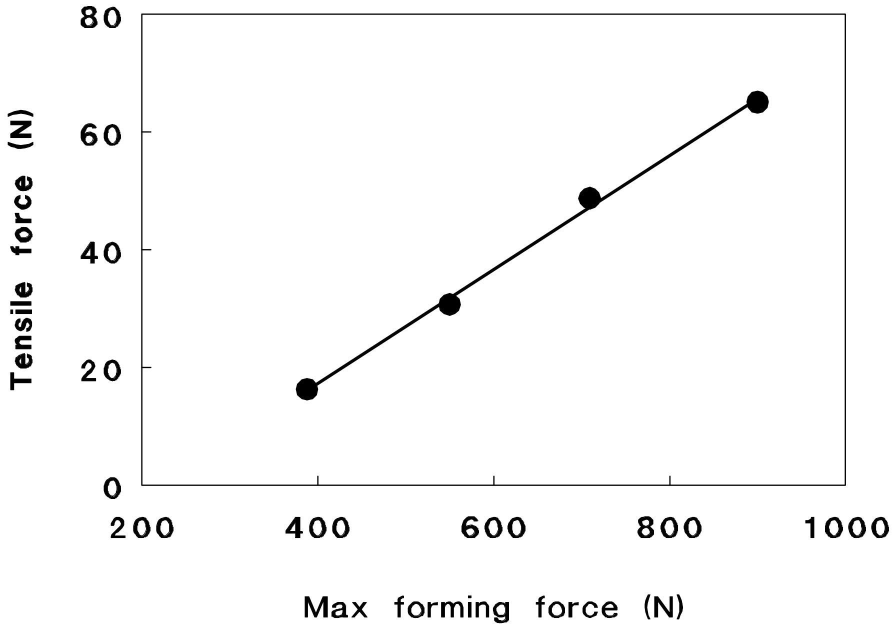

Figure 7: Force in tensile test vs the maximum forming force (10% strain)

It does appear, however, that tensile deformation of the sheet is not the only factor which influences the postforming behaviour. Postforming is also sensitive to the interlayer bonding and this means that the weave style is important. Simple calculations show that, especially if the sheets are clamped, very large shear strains (~100%) can occur. Interlayer failure is affected by the closing speed; at lower speeds delamination only occurs at higher strains.

COMPARISON WITH OTHER MATERIALS

Table 4 below shows a comparison of the properties of compacted PP sheet (6082 - plain weave) with typical properties of other likely competitive materials.

	Compacted PP sheet (6082)	Isotropic PP (Homopolymer)	Random mat Short glass/PP 40wt% fibre	Twintex Unidirectional 60 wt% fibre
Density (kg/m3)	920	900	1190	1500
Tensile Modulus (GPa)	3.5	1.12	8.28	15
Tensile Strength (MPa)	87	27	101	350
Heat deflection temperature (°C)	143	110	154	159
Notched Izod impact strength (J/m)	>3000	200	752	2000
Thermal expansion ($x10^{-6}$/°C)	73	96	27	21
Recyclability	EASY	EASY	HARD	HARD
Thermoforming	MEDIUM	EASY	MEDIUM	HARD
Surface style and colouring	MEDIUM	EASY	HARD	HARD

Table 4: A comparison of the mechanical properties of compacted PP sheet with other materials

As would be expected the majority of the mechanical properties of the compacted PP sheet lie somewhere between isotropic PP sheet and random mat short glass fibre filled PP: the impact strength, however, is the best of all the materials. In addition the compacted PP sheets are easier to recycle and thermoform than the glass filled grades, and offer the potential for a range of surface styles and colours.

FUTURE PROSPECTS

Future work in this area will be split into two main themes: continuation of ongoing development programmes and more fundamental compaction studies. Current ongoing development programmes include the use of compacted PP for loudspeaker cones (for a report see Gramophone July 1998), a joint study with Ford Motor Company into the possible uses of compacted PP sheet for automobile applications, and the development of hot compacted polyethylene fibre sheets for radome covers because of the very low absorption of electromagnetic radiation. Fundamental studies will include an investigation into how the morphology of the starting oriented tapes/fibres translates through into the properties of the compacted sheets, and the development of a hot compaction route for higher performance materials.

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