# ENHANCED BONDING OF POLYPROPYLENE TO POLYPROPYLENE AND OTHER MATERIALS WITH NOVEL THERMOPLASTIC HEAT-ACTIVATED ADHESIVES

S-W Tsui, H M White and A F Johnson GLUCO Ltd., School of Chemistry, University of Leeds, Leeds LS2 9JT, West Yorkshire, United Kingdom.

#### Abstract

It is commonly accepted that bonding polypropylene to itself or other adherends is difficult and the options available for cost-effective bonding using adhesives are very limited. The aim of the presentation will be to describe a new range of heat-activated adhesives, recently developed in our research laboratories, and their applications. These adhesives, which are now commercially available, offer numerous advantages for the rapid manufacture of composite materials, in addition to promoting new or improved assembly methods in a wide range of market sectors. e.g. automobile, aerospace, construction, textiles, footwear and packaging, to mention but a few. The main focus of the presentation will be to outline various ways in which the film, strand or pellet forms of the new adhesives may be used to solve a variety of industrial problems. The range of materials to which polypropylene can be successfully bonded, (e.g. to itself, to many metals, and to a range of other materials, notably, cellulosics), will be outlined, together with their associated manufacturing methodologies, such as hot compression, lasers and induction heating. An indication of the mechanical bond strengths, which can be achieved at various temperatures, will also be outlined.

### **1. Introduction**

Polypropylene (PP) is a very high tonnage polymer which has found use as a raw material in numerous industrial sectors such as textile, packaging, automotive and a wide range of consumer goods. The low cost, processability and chemical inertness of PP is largely responsible for its application to a variety of manufacturing processes. However, the chemical inertness has also curtailed the use of PP for some industrial applications because it is difficult to print on PP or to bond PP to itself or to other materials such as aluminium or steel. To overcome this problem, many commercial adhesives rely on subjecting the PP substrate to either a chemical or physical pre-treatment to modify the polymer's surface, e.g. flaming, chemical etching, plasma and corona treatments. Such adhesives and processes often involve the use of chemicals and are so expensive and slow in operation that they are only used as the last resort, and are often rejected by large manufacturing operations, such as car assembly, on grounds of both cost and hazards to health.

The new materials which have been developed now present a solution to many PP bonding problems – a one-part, no-curing, user-friendly, heat-activated adhesive which can routinely bond PP both to itself and to other more polar substrates, e.g. aluminium, steel, copper, cellulosic materials, leather, etc., <u>without</u> any substrate pre-treatment.

## 2. Bonding of plastics

Successful joining of plastics relies on significant interactions between the adhesives and the surfaces being joined. Aubrey (1) has classified these interactions under the following headings:

#### 2.1. Mechanical interlocking

The macro- and microscopic roughness present at the bonding surface potentially promotes adhesive interactions via the formation of 'mechanical anchors' at the plastics' interfaces. This is generally achieved by roughening the bonding surfaces with abrasive materials.

#### 2.2. Physical adsorption (wetting)

The adhesive wets the surface of the plastic enabling attractive forces to occur between the liquid (adhesive) and the two solids.

#### 2.3. Molecular diffusion and inter-penetration

This is the physical mixing of the adhesive and plastic at the interfacial layers owing to the absence of a well-defined boundary between two materials. This is not always easy to achieve due to reasons of thermodynamics, especially when bonding non-polar polymers such as polyolefins.

It is believed that all these mechanisms are involved in bonding PP with the novel heat-activated adhesives. However, it is not within the scope of this paper to discuss the bonding mechanism.

### 3. Novel thermoplastic heat-activated adhesives

The technology discussed here was developed by GLUCO Ltd., a spin-off company of the University of Leeds, United Kingdom, that builds on many years of fundamental research by the Polymerisation Reaction Engineering Group, based in the Interdisciplinary Research Centre (IRC) in Polymer Science & Technology, Department of Chemistry. The company retains strong links with its academic base. The technology is based on chemically modified PP prepared by reactive extrusion (REX). Reactive extrusion refers to the deliberate performance of chemical reactions during extrusion, whereby speciality polymers are made either by modification of existing polymers, or in certain cases, polymerization of suitable monomers. The use of REX for modifying polyolefins is not new. Many relatively cheap polymers have been modified successfully by REX to produce value-added speciality polymers, e.g. polypropylene grafted with maleic anhydride, by some multinational companies.

Early work on REX by Brown, Johnson and Tsui has led to a number of patents (2-4). The same basic chemistry and processing technique allows a wide range of chemically modified PP to be produced and it is some of these that are showing considerable promise as heat-activated adhesives (5). A route has been found to produce such adhesives in a manner which is both cost-effective and compatible with existing methods employed in the PP industry. REX has been used to chemically modify PP, yielding products with excellent adhesive properties. The advantages of these adhesives include the following: (i) when used to bond polypropylene, the final product can be recycled, (ii) when bonding metals, the bonds can be reverse-engineered to aid repair or dismantling of products for recycling or other purposes, (iii) no curing time is required and continuous bonding processes are possible, and (iv) there are no wet chemicals involved in the bonding processes i.e. the system is environmentally friendly.

### 4. Processing and applications

As the name implied, bonding with these novel heat-activated adhesives is achieved by the application of heat. Conventional heating processes such as hot lamination (batch and continuous), hot calendaring, hot air welding, dielectric or induction heating (with the use of appropriate additives) can be employed. With the advent of comparatively compact and 'affordable' diode lasers, it has been shown successfully that high strength joints between thermoplastics and mild steel, of 'engineering standard', can be produced using these novel heat-activated adhesives (6). A cost-effective, environmentally friendly method for bonding PP to other plastics and to metals has been developed. The combination of diode lasers and the heat-activated adhesives offers the opportunities for automatic bonding systems for, e.g., the automotive and aerospace industry.

Bonding PP to PP with these new adhesives is usually carried out at temperatures around 155°C which is below the melting point of PP (approximately 165°C). The heat-activated adhesives can be used in the form of pellets (e.g. co-extrusion and injection moulding), strands or rods (e.g. hot air and induction welding), and so on. However, it has been found that the most effective way to use these adhesives is in the film form for laminating large flat panels in a continuous manner. For laminating curved and shaped panels, a semi-continuous thermoforming process can be adopted. Successful applications, to name a few, include the following:

- Compacted PP fabric/Compacted PP fabric for engineering applications.
- PP/PP foam composite panels for engineering and insulation purposes.
- PP/PP honeycomb composites for lightweight automobile structures.
- PP/aluminium composite structures for aerospace and other industries.

- Aluminium/Formica® panels for bus and coach construction.
- PP/steel for pipeline lagging.
- Cotton/cotton for textile applications.
- Cotton/aluminium laminated fabrics for safety and other applications.
- PP/leather for the footwear industry.
- PP/PP packaging materials.

## 5. Experimental

### 5.1. Preparation of dry film adhesive

Approximately 4 kg of adhesive pellets were dried in an oven at  $60^{\circ}$ C overnight. The pre-dried pellets were then used to extrude into rolls of film of approximately 250 mm in width and 60-70  $\mu$  in thickness. The extrusion temperature was approximately 180°C and the wind-up speed was 1.5 m min<sup>-1</sup>. The film so produced had excellent optical clarity and was non-tacky at ambient temperatures. This process was later adopted to manufacture dry film adhesives on a commercial scale.

#### 5.2. Preparation of adhesive joints

The bond strength of the dry film adhesive, as described in Section 5.1, was examined by lap-shear test of single overlap joints (with reference to BS 5350 : Part C5 : 1990, ASTM D 1002-94 and D3164-97) and T-peel test (with reference to BS 5350 : Part C12 : 1994 and ASTM D 1876-95). Sheets of hot-compacted PP (CURV<sup>®</sup>, BP Chemicals), glass-filled PP, aluminium and mild steel were used to prepare all the test samples. The sample surface was degreased with acetone prior to bonding. No further surface treatment was carried out. The bonding temperature and contact time were 155°C and 1 minute respectively. A gentle pressure of approximately 0.02 MPa (3 psi) was applied to ensure a good contact between the adhesive and the bonding surfaces. A bond was formed as soon as the molten thermoplastic adhesive had solidified. No further curing was required. The joints were then tested at temperatures between -40°C to 130°C.

## 6. Results and discussion

### 6.1. Effect of temperature on the performance of dry film adhesive

#### 6.1.1. Shear strength

Figure 1 shows the results of the shear test of single overlap joints of various adherends as described in Section 5.2. The bond strength increases with decreasing temperature as the molecular chains of the adhesive become more rigid. Since the adhesive is PP based, one would expect the Tg of the adhesive and the sigmoidal shape of the temperature-dependent performance curves in Figure 1 to resemble that of PP. The bond strength for hot-compacted PP was somewhat lower compared to that of aluminium and mild steel. This was due to the fact that the bonded surfaces of the various hot-compacted PP samples delaminated during the test, i.e., the bond was stronger than the adhered. The results thus only represent the inter-laminar shear strength of the hot-compacted PP. The performance of the adhesive on PP was examined by using a glass-filled PP as adherend. It was found that the bond strength (shear) was 11.4 MPa at  $18^{\circ}$ C, comparable to the corresponding values for aluminium and mild steel as adherends (11.5 and 14.3 MPa respectively).

## 6.1.2. Peel strength

It can be seen in Figure 2 that the maximum peel strength for each adherend is at temperatures in the region of  $20-40^{\circ}$ C. When the test was carried out at higher temperatures, the thermoplastic adhesive was softened, resulting in a lower bond strength. On the other hand, when the test was carried out at temperatures below  $20^{\circ}$ C, the PP based adhesive went through its glass transition and became brittle. The high stress concentration at the separating front could have cracked the brittle adhesive just before the two adherend surfaces were separated, again resulting in a lower bond strength. Similar to that discussed in Section 6.1.1, the bond strength for hot-compacted PP was somewhat lower compared to that of aluminium and mild steel.

Again, the performance of the adhesive on PP was examined by using a glass-filled PP as adherend. It was found that the peel strength was  $3.7 \text{ Nmm}^{-1}$  at  $18^{\circ}$ C.

#### 6.2. Effect of contact pressure on bond strength

The results described in Sections 6.1.1 and 6.1.2 show the general trend of performance of the adhesive with temperature. The bond strength can also be affected by the preparation conditions such as contact pressure, glue-line thickness and so on. The bonds for the tests in Sections 6.1.1 and 6.1.2 were prepared with one layer of adhesive film of 60-70 $\mu$  in thickness, and the contact pressure was approximately 0.02 MPa (see Section 5.2). The pressure applied to prepare bonds must not be so great that the PP which is being bonded becomes deformed. The risk of deforming a metal adherend is lower compared to that of PP, and the effect of contact pressure on peel strength can be examined using untreated aluminium as adhered. All peel tests reported in Table 1 were carried out at 18°C.

It can be seen in Table 1 that, by increasing the pressure from 0.02 MPa to 0.2 MPa, the peel strength of these samples was increased by 100%. It is important to apply sufficient pressure during the bonding process in order to ensure an intimate contact, hence interaction, between the adhesive and the adherend surfaces. However, pressure has to be applied with discretion so as not to deform the articles which are being bonded, especially when joining PP at temperatures close to its melting point.

## 6.3. Effect of adhesive thickness on bond strength

The effect of adhesive thickness on peel strength was examined using untreated aluminium as adhered. All the tests were carried out at  $18^{\circ}$ C. A contact pressure of 0.2 MPa was applied to all the samples in order to give an intimate contact between the adhesive and the adherend surfaces (see Section 6.2). Results are shown in Table 2.

It can be seen in Table 2 that, in addition to applying sufficient pressure to ensure an intimate contact between the adhesive and the adherend surfaces, the quantity of applied adhesive also affects the bond strength. This is particularly important when dealing with porous or absorbent adherends. Sufficient quantity of adhesive has to be used in order to wet the entire surface being bonded. The advantage of using a dry film adhesive is that the quantity of applied adhesive is well-controlled and defined by the film by virtue of its area and thickness.

Results shown in Sections 6.1 - 6.3 serve as guidelines for using these novel heat-activated adhesives. There are no definitive processing conditions regarding parameters such as contact time and pressure. They vary from process to process and one needs to experiment in order to achieve the optimal result for these novel heat-activated adhesives.

## 7. Conclusions

Polypropylene is a very high tonnage polymer which has found use as a raw material for a wide range of industrial products and the total usage is still increasing. It is generally recognised that PP is very difficult to bond to itself. Bonding PP to other engineering materials such as steel and aluminium is even more challenging and desirable. A new range of thermoplastic heat-activated adhesives has been developed to meet such a demand. It has been shown that high strength joints between thermoplastics and metals of 'engineering standard' can be produced with these novel, user-friendly, one-part heat-activated adhesives.

## References

- 1. Aubrey, D.W., *Bonding in flexible joints*, Joint Symposium on Adhesive Bonding of Flexible Materials, The City University, London, (25 September 1985).
- 2. Brown, M.W.R., Johnson, A.F. and Tsui, S-W.; *Reactive polypropylene yarns*; <u>International patent</u> <u>application number PCT/GB96/01025</u>; (1996).
- Brown, M.W.R. and Johnson, A.F.; *Modified polyolefins*; <u>International patent application number</u> <u>PCT/GB96/01026</u>; (1996).
- 4. Brown, M.W.R., Johnson, A.F. and Tsui, S-W.; *Reactive polymer processing*; <u>International patent</u> <u>application number PCT/GB97/02615</u>; (1997).
- 5. Johnson, A.F. and Tsui, S-W., Patent pending.
- Hoult, A., Pelaprat, J-M., Tsui, S-W., White, H. and Johnson, A.F; *Bonding polypropylene to metals using diode lasers and tailored heat-activated adhesives*, 7<sup>th</sup> International Welding Symposium, Kobe, Japan, (20-22 November 2001).



Figure 1. Effect of temperature on the performance (shear strength) of a film-grade heat-activated adhesive.



Figure 2. Effect of temperature on the performance (peel strength) of a film-grade heat-activated adhesive.

Pressure / MPa	Peel strength / Nmm <sup>-1</sup>
0.02	4.8 (Result from Section 6.1.2)
0.2	9.3

Table 1. Effect of contact pressure on peel strength. (Adhesive: one layer of heat-activated adhesive film of  $60-70\mu$  in thickness. Adherend: untreated aluminium.)

No. of layers of adhesive film (60-70µ each)	Peel strength / Nmm <sup>-1</sup>
1	9.3 (Result from Section 6.2)
2	12.4

Table 2. Effect of adhesive thickness on peel strength. (Adhesive: heat-activated adhesive film. Adherend: untreated aluminium.)