

# **Advances in Joining Plastics and Composites**

Bradford, Yorkshire, UK, 10-12 June 1991

Technical Director — Martin N Watson



**International Conference**

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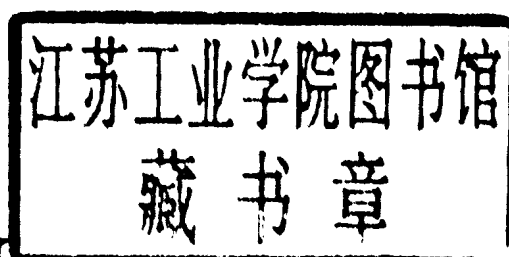
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# **International Conference ADVANCES IN JOINING PLASTICS AND COMPOSITES**

**Bradford, Yorkshire, UK, 10-12 June 1991**

**Martin Watson, BSc, PhD, MWeldI, AMPRI**  
Head, Plastics Joining Department, TWI

*Conference Technical Director*



**Organised by TWI**

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## FOREWORD

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The plastics industry is large, rapidly growing and has products whose performance covers an enormously wide range, from low cost 'commodity' plastics to high strength engineering plastics and others whose performance is tailored to meet high temperature or corrosive service conditions. The addition of reinforcing fibres further extends the service range of these materials. Today a greater tonnage of plastics than metals is used and they are a feature of almost all sectors of industrial production. As well as widespread use in domestic appliances, packaging and other mass production applications, these materials are of growing importance in vehicle manufacture, aerospace, electronics, marine and offshore, and chemical plant. Joining is a key technology in almost all areas of engineering production and the growing use of plastics and composites has highlighted the need for fast, reliable, safe, yet cost effective joining methods for them.

The pace of developments in this field is quickening, so the aim of this conference was to bring together researchers, technologists and users in the field to review recent developments across a broad spectrum of topics such as equipment, process technology, testing, materials aspects and applications.

The papers presented cover all aspects of the technology of joining plastics and composites, dealing with topics such as adhesive bonding, fusion welding, ultrasonic techniques, the joining of composites and the joining of pipes. I am sure that these proceedings will be of wide interest to those working in production as well as research and development.

I would like to thank the authors of the papers, the session chairmen and the TWI staff who organised the successful conference.

*M N Watson  
Head, Plastics Joining Department  
TWI*

# Advances in Joining Plastics and Composites

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## **Recent development in composite bonding with particular reference to large structures and unprepared surfaces**

W A Lees (PermaBond Division, National Starch & Chemical Ltd)

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### **SUMMARY**

The excellent performance record which has been established by composites over the last 30-40 years is acknowledged by an ever-increasing commitment to the creation of ever larger structures which must function effectively for prolonged periods. The construction of large monolithic hulls is now well founded, but even in the shipbuilding industry there is a need to fabricate large structures from panels. Elsewhere, the intricacies of design or transportation problems demand construction techniques based on the assembly of sub-components. Bonding is one method which can be used to assemble the parts and it is known to be very successful. However, in a world where production costs are forever under pressure it is essential to know the limiting parameters. Surface preparation being a case in point. Cleaning is expensive, how much is necessary? The paper explores basic issues such as this in relation to the assembly of large structures from panel-like components.

### **Definition**

For the present purpose a large structure is considered to be anything from a vehicle door upwards - a complicating issue for vehicle doors are likely to be fabricated from any one of a number of materials and potentially by several processes. But, despite a component's size, small parts may have to be attached and while they are considered in the paper emphasis will be placed on the assembly of large panels generally, but not exclusively, fabricated from polyester/glass laminates.

### **What is being bonded?**

The parts involved range from vehicle doors, tailgates, floor pans up through the body panels of vans, buses, coaches, articulated lorries, marine structures - including offshore platforms - ship's hulls and superstructures.

From hand-lay and spray techniques via the increasing sophistication of the various intermittent moulding methods to the continuous manufacture of pultrusion a multitude of individual matrix resins are seen. They can be resolved into the following major, but not exclusive, groups:

- |                       |            |
|-----------------------|------------|
| * Polyamide (Nylon)   | * Epoxide  |
| * Polypropylene Oxide | * Phenolic |
| * Polypropylene       | * Urethane |

Moulded parts are likely to be contaminated by mould release agent - not exactly the best foundation for long term bonding! Such release agents may be used either directly in the mould while others may be incorporated in 'self-releasing' formulations.

The composite may be bonded to other materials such as:

- \* Aluminium
- \* Zinc
- \* Steel
- \* Paint
- \* Another composite

Of the many paints that might be used only the electro deposited Cathophoretic paints applied to properly prepared surfaces are likely to provide a sufficiently stable base for structural bonding.

The compatibility of the various material substrates with a number of candidate adhesives is discussed in detail in Section 6 and summarised in Table 3.

### Design for bonding

Outside the aircraft industry, which is not under direct consideration, the larger the composite part the flatter and more geometrically simple it tends to be. This is important for there is benefit to be gained by designing a component in such a way that the adhesive will be supported by the part's geometry rather than being undermined by it. This is achieved by shaping the jointed area so that the forces experienced tend only to compress the adhesive or induce, if possible, both compression and shear loads. If this cannot be done the design should induce the unwanted cleavage forces away from the vulnerable edge of the joint, see Fig. 1.

The designer has another major consideration: the variable and potentially large gap which inevitably accompanies the assembly of large components.

At a simplistic level the adhesive has the advantage that it can be used to fill in the gap (hole) and both bond and seal the components. But many engineers have heard that 'the thinner the bond-line, the stronger the joint' and while not necessarily familiar with the subject they will be aware of the fact that stress distribution in a joint is not just a matter of averaging out the load over the area involved.

An example of the benefit obtained, in terms of reducing joint edge stress, by bonding Sheet Moulding Compound (SMC) with a ductile toughened acrylic adhesive rather than a stiff brittle polyester resin is clearly illustrated in Fig. 2.

Given the foregoing how can a designer ensure that he creates a structure which maximises performance while utilising a flat panel which, in itself, can contribute no geometric benefit to a bonded joint?

The ultimate performance may be gained by bonding such panel section into either an aluminium extrusion or into a composite pultrusion. Joints such as there, based on the concepts of Fig. 3, are virtually indestructible given an appropriate insert length.

### What is available?

Of the many adhesive types some five groups offer likely candidates for composite bonding. Two of these - the phenolic and polyamide types - tend to be used on very specialised work and for the bulk of applications three major families dominate. They are:

- \* epoxy - one and two part
- \* acrylic - true one part, pseudo one part and two part



- \* polyurethane (PU) - one and two part

The fundamental differences are:

- \* the basic chemistry of individual family type and -
- \* the mechanical properties of individual formulations within those families

### The family types

The reaction chemistry of the epoxy resins is much slower than the isocyanate group which drives the hardening reaction of the polyurethane (PU) adhesives. The isocyanate group will react with water - spontaneously in the case of atmospheric moisture. This has the advantage that single part sealants and low strength adhesives may be formulated using this reaction. They cure, rather slowly, by absorbing atmospheric moisture. Functionally more effective forms use the same chemistry but also incorporate the 'hot-melt' principle. The adhesives which rely on atmospheric moisture suffer the disadvantage that open containers can 'self-destruct'.

In practice, for a short reaction time to be achieved with an epoxide based adhesive, the material in question must be either warmed or heated. By contrast, the PU's driven by the isocyanate group will set in seconds at room temperature should this be desired. Although engineers always like to minimise production times, these extremely short periods are not normally invoked because of the time required to both apply the adhesive to the components and then put them together.

The need to slow the reaction rate to a point which will allow satisfactory assembly often has the effect of making the time required for the adhesive to acquire sufficient 'handling strength' - in order that it may be removed from the jig - too long. Consequently, jigs and clamps are often heated to approximately 100-120°C to allow rapid demounting during mass production operations. By contrast, the single part epoxides must be heated to a minimum of 100°C to ensure their proper curing. While frequently used for aircraft work these sorts of temperatures strongly inhibit the use of single part epoxides for large scale assembly elsewhere.

Normally, the acrylic adhesives are not heated. Essentially, there are three types of acrylic based adhesive.

The first are the cyanoacrylate (Super-Glue) adhesives which set in seconds using absorbed surface moisture as their curing catalyst. They are not normally considered to be load bearing materials and will not be discussed in detail.

The second are pseudo-one part, no mix types, which rely on the self-mixing of the initiating catalyst and the adhesive. The initiator is spread very thinly on one surface and the adhesive bead is placed on the other. Pressing the parts together during assembly gives sufficient mixing to effect cure. The reaction is fast giving short de-jig times.

The third are the part pre-mixed versions which are also fast. The reaction rate can be specifically modified to automatically increase when the joint is closed by the placement of the second component. This occurs because atmospheric oxygen can no longer get access to the adhesive and, as a consequence of the chemistry involved, can no longer slow the reaction. A summary of this behaviour is given in Table 1 for the materials Permabond V500 and V501.

Table 1 - Variable reaction rates

	V500*	V501
Open time (minutes)	10	20
Handling time (minutes)	5-15	10-30
Functional time (minutes)	30-60	60-120

- Open time            - maximum time which may elapse from the start of mixing and dispensing to the point at which the joint must be assembled.
- Handling time       - time lapse from joint assembly to the point where sufficient bond strength has been developed to allow careful handling.
- Functional time      - time taken to achieve working strength.

The relative cure rates at a variety of temperatures, appropriate to the adhesives involved, are given in Fig. 4.

**Their mechanical properties**

Figure 5 presents the main family types set out in terms of their shear moduli.

The moduli of the PU based adhesives span more than two orders of magnitude. No distinction is made between one and two part PU's. This is because their different formulations can take up almost any desired position on the scale. From the chemist's point of view it would be very easy to modify all the families so that they too could span the scale given - but economics and other aspects of working reality limit the possibilities to the broad outline given. Figure 5 indicates that the lower modulus materials (PU, acrylic and some two part epoxides) are best used for those applications where the unit loads being transferred from one component to another are very low. Furthermore, at extremely low moduli the materials typified by one part PU's are probably best considered as being no more than sealants.

Typical applications for the medium and lower modulus adhesives are the mounting of vehicle windscreens, composite body panels and tailgates.

One needs to be very careful when considering stress distribution with regard to an edge. If the edge represents a 'width' then the unit load is proportionately reduced by an increase in the dimension. If it is a 'length' then the unit load will only be proportionally reduced by a dimensional change if either the adhesive is extremely ductile or the lengths involved are very short. This follows from the disproportionate distribution of stress in joints as represented for an idealised lap joint in Fig. 6.

---

*\* If the open time is restricted to one minute then the time to acquire handling strength would be 6-16 minutes from the moment mixing commenced. By contrast, if the joint is not closed until some 10 minutes have elapsed since the adhesive was mixed and applied then the corresponding handling time will be 15-25 minutes.*

The low modulus of the PU adhesives enables them to bond materials of widely differing expansion characteristics without either inducing unacceptably high local stresses or cosmetically distorting any thin plastic panels being assembled. Such distortion can also be induced by the shrinkage of the adhesive during the curing process - again an issue readily avoided when low moduli materials are used.

Care needs to be taken to ensure that a low modulus adhesive is not overloaded in such a manner that it creeps over extended periods - particularly when exposed to elevated temperatures.

At the other extremity of the modulus spectrum lie the heat cured, single part epoxides. These adhesives are amongst the most durable and robust known. While the need for heat curing limits their use to the specialised applications, their stiffness and high strength makes them ideal candidates for the transmission of substantial loads. A prime example of this being their use to assemble the composite propeller shaft of the Renault Espace. Nonetheless, when materials with different coefficients of expansion are to be bonded, care needs to be exercised to ensure that unacceptable stress is not induced by their thermal cure (Fig. 7)

It is possible that in some designs a lower modulus material, such as a cold or warm cured two part epoxy, would be better. An example of this is given in Fig. 8 which compares the stress distribution generated by adhesives of different moduli under otherwise identical circumstances. It is clear that the lower modulus material is the more suitable for its elastic limit is not exceeded by the joint-edge stresses.

Co-axial joints are ideal candidates for the transmission of substantial forces, particularly because they do not induce peel/cleavage forces in either the adhesive or the composite surfaces being bonded. However, the most convenient joint is usually some form of lap joint in which cleavage forces are readily introduced unless care is exercised to ensure that they are either suppressed by a design feature or by some mechanical reinforcement (Section 3.)

The  $T_g$  of any adhesive will be reduced by water absorption and as a consequence there will also be a corresponding reduction in modulus. The modulus is itself a function of temperature and will change sharply if the temperature of the adhesive moves through its  $T_g$ . It should be noted that the stiffer, structural adhesives are intended to function below their  $T_g$  unlike elastomeric materials and some of the PU adhesives.

It is perhaps worth mentioning that PU adhesives are particularly susceptible to chain scission in the presence of moisture and while this may lead to a reduction in modulus some versions have even been known to stiffen up as the rendered polymeric chain re-formed into a more cross-linked structure.

Except in the more highly stressed joints - which do need careful design - such considerations may be considered academic. But, as composites are pushed into even more demanding situations and are used in direct competition with conventional materials these issues are becoming increasingly more significant and along with a general demand to improve the toughness of adhesives there are additional requests for even higher  $T_g$ s. The two do not go readily together for as  $T_g$  and stiffness climb there is a strong tendency for brittleness to increase.

### Material selection and production considerations

The designer's choice of material, its preparation for production bonding and that latter process itself are limited by the reaction chemistry of the main family types and the durability of the end product of that reaction. Durability is the key and its achievement a controlling factor.

#### **Epoxides**

There is no doubt that these adhesives in their heat cured, single part form are extremely durable. For example, they have been used to assemble millions of heat exchangers fabricated from a glass-filled nylon composite and aluminium tubes. There have never been any failures despite the fact that the joints have been exposed to a hot 25% glycol solution for many years.

Two part epoxies are also of good durability as their use in vehicle and shipbuilding industries testifies.

#### **Acrylics**

The fundamental form of the acrylic adhesive is far too stiff and brittle to be considered useful for structural purposes. Consequently, it is 'toughened' or flexibilised with a variety of rubbery additives.

A study produced by the Admiralty (AMTE (M) TM 79307) has demonstrated that these adhesives outperformed the polyester composites that they were used to assemble. As a consequence they have been extensively used for the bonding of composite and aluminium components in ocean-going ships.

#### **Polyurethanes**

The PUs have a particular problem - they are sensitive to chemical attack by water - especially when warm and particularly in the presence of metal or metal oxide when some form of catalytic degenerative process also appears to take place. This is exemplified in Fig. 9 which demonstrates the relative performance of an acrylic based adhesive by comparison with three different PUs (one a moisture cured single-part) from different manufacturers when assessed on steel testpieces.

PUs must not be used directly upon uncoated metal surfaces without a thorough prior investigation.

### Material selection

Table 2 will give a broad impression of the compatibility of a number of composite materials with the adhesives generally considered to be suitable for their structural assembly. Also included are other materials often used in association with composites. The information given should be interpreted carefully in the light of the following observations:

#### **Metal**

Both the heat cured epoxides and the toughened (Hypalon) acrylic types will bond oil contaminated metal surfaces very successfully. However, safety critical, loaded joints must

be properly prepared to guarantee a long durable performance. PU based adhesives must not be used directly on bare metal or oil contaminated surfaces.

#### Aluminium

Bonds well with the epoxides and particularly well with the pseudo single-part acrylics. Maximum durability requires either silane, phosphate or chrome passivation treatments.

#### Steel

Maximum durability requires silane or Accomet C treatment.

#### Zinc

This is a weak, readily oxidised metal that behaves badly unless properly prepared and loaded. Both Accomet C and chrome passivation give bonded joints of excellent durability though if these are to be loaded low modulus adhesives should be used. (Note: PUs will need a separate primer coating).

#### Thermoplastic

##### Polyamide (Nylon)

Bonds poorly unless bonded hot or pre-treated by shot blasting, oxidation or Corona Discharge\*. When hot bonding consider release of absorbed moisture.

##### Polyphenylene Oxide

Bonds poorly unless bonded hot or pre-treated by shot blasting, oxidation or better by Corona Discharge. When hot bonding consider release of absorbed moisture. May stress crack.

##### Polypropylene

Surface preparation needed; shot blast, better being, oxidation, Corona Discharge, or chlorinated primer.

#### Thermoset

##### Epoxy

Usually used for only the most demanding circumstances and consequently bonded with either the two part or single part epoxides. When the latter are used care should be taken to ensure that absorbed water is released. While the acrylics and PUs can bond them well the lower moduli of these adhesives generally makes them unsuitable for the type of work normally involved.

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\* Wherever Corona Discharge is mentioned plasma treatment may also be considered. Although the latter is said to be more effective it requires the use of a vacuum chamber - difficult or impossible for large structures.

Phenolic

Bonds very well with epoxides (ensure absorbed water released if heat cured versions to be used) but some types do not suit all acrylics and PUs quite as well. However, a point worth noting is that the PUs can release particularly toxic (-CN) derived fumes on burning. As phenolic composites are normally used because of their own low smoke and fume emission when burnt the use of a PU to bond them is questionable.

Polyester

Will bond well with all types of adhesive but single part epoxides not normally used because of the higher curing temperatures required. If single part epoxides are used then care should be taken over the release of absorbed water. Surface preparation needs very careful consideration.

Polyurethane (RIM)

Bonds particularly well with PU adhesives and selectively with epoxides and acrylics.

Paint

Catophoretic paints are probably the only paint materials with sufficient integrity to be used as a substrate for structural bonding. Even then, the underlying surface will need proper preparation if the paint is to function properly. Paint, or speciality primer, needs to be used to ensure the chemical stability of PU adhesives on metal

Table 2. Compatibility of the principal structural adhesives with a variety of composite and associated materials.

Material to be Bonded	Acrylic		Epoxy		PU
	Pseudo 1 part	Two part	One part (heat cured)	Two part	Two part
<b>Metal (also see Paint)</b>					
- Aluminium	1	1	1	1	4
- Steel	1	1	1	1	4
- Zinc	2	2	2	2	4
<b>Thermoplastic</b>					
- Polyamide (Nylon)	2	2	3	2	2
- Polyphenylene oxide	3	3	3	2	1
- Polypropylene	2	2	4	3	1
<b>Thermoset</b>					
- Epoxy	2	2	1	1	2
- Phenolic	2	2	1	1	2
- Polyester - hand lay	1	1	2	1	1

- VARI	1	1	2	1	1
- SMC	1	1	2	1	1
- cold press	1	1	2	1	1
- Polyurethane - RIM	3	3	4	2	1
Paint					
- Cataphoretic	1	1	1	1	1

Scale:

1 - Excellent

2 - Good

3 - Good but possible problems

4 - unsuitable

Note:

This categorisation is given in relation to the types of application usually seen in association with the materials nominated. Therefore, each line should be considered to be unique.

Set against this background it will be appreciated that the cheapest adhesives may not provide the most economical solution which is technically acceptable. Each application requires careful consideration of the costs incurred when extra painting processes may have to be invoked or heating equipment is required to cut production cycle times to an acceptable figure.

Production

Effective production requires the creation of a specified product within specification at minimum cost. Given that such cost incurring issues as design, compatibility and long term durability have been resolved then for a large structure the closing considerations are:

- ease of use
- the cost incurred when mixing is needed
- the cost of surface preparation

Surface preparation

The thermo-dynamics of thermoset composites favour the bonding process and were it not for the need to remove mould release agent and accidental contaminants then there would be no requirement to prepare their surfaces prior to bonding.

Mould release agents do not form a good foundation for adhesives and consequently European practice has always been to either remove them using very light abrasion, followed by a solvent wipe, or to suppress their influence by wiping over with chemically active primers. Such good practice has given faultless performance in road vehicles and their demanding circumstances over a 20 year period.

Recent work at Permabond has produced evidence that given suitably controlled production techniques the long term bonding of unprepared composite can be anticipated whether the bond line is heated or not. Figure 10 illustrates this point for a medium modulus, two part, toughened epoxy.

Thermoplastic materials are not generally so favourable and bonding may prove difficult unless the plastic concerned is treated in some way. This may range from shot blasting to

oxidation through corona discharge to the use of the chlorinated polyolefin primers which are now being introduced for the bonding of polypropylene based composites.

**Mixing and application**

The issues involved are perhaps best examined by considering two extremes. At one end of the scale stands the motor vehicle automatic component assembly line while at the other is the steady but slow assembly of a ship's panel work in an ever changing environment. This latter situation does not lend itself to major capitalisation except under exceptional circumstances. Yet, the quality demands on the workforce are much higher simply because of their circumstances and an environment which is much harder to control.

The assembly of big, awkward panels high above the ground requires simple, light equipment which will function well despite variable temperatures. Yet while the adhesive needs to be paste-like it will also need to be mixed and dispensed easily. These contradicting requirements may be resolved by the provision of correctly formulated materials which can be mixed at pre-determined temperatures by a fixed location mixer/dispenser which fills re-usable cartridges to be distributed on demand to the assembly team.

This system also has the advantage that air is not incorporated into the mixed adhesive as it would be were hand mixing and pasting employed.

As assembly conditions become progressively less arduous and demanding of the workforce, cartridge guns incorporating static mixers can be used directly. These have improved considerably of late and the better designs have given the adhesive manufacturers greater scope for versatility.

The mass production of large bonded composite panels required very careful control of the temperature and viscosity of a mixed adhesive system. Manufacturers contemplating the installation of a new line are strongly recommended to review their needs with their adhesive supplier.

**CONCLUSION**

Large composite structures, particularly those based on thermoset resins can be readily and durably bonded both to themselves and associated metal structures. A summary of the main characteristics, benefits and shortcomings of the adhesive types generally involved is given in Table 3.

Table 3. Characteristics of the principal structural adhesives for composite bonding

	Main Characteristics	Principal Advantages	Principal Disadvantages
PU 1 part	Low modulus very low strength	Very simple to use. Hot melt variants very convenient on suitably sized components. Fills large gaps. No mixing.	Sensitive to moisture. Not true structural adhesives. Slow curing. Must be applied to non-metal surface for long term durability.



PU 2 part	Very low to medium modulus. Very low to medium strength	Fast curing possible. Very good application characteristics. Fills large gaps	Sensitive to moisture. Often requires heating to achieve acceptable production times. Must be applied on a non-metallic surface for long term durability. Some versions cannot be considered to be structural adhesives. Must be mixed.
Acrylic pseudo 1 part	Medium modulus Medium strength	Very fast. Very easy to apply. Extremely durable. Bonds metal particularly well. Copes with contamination well. A true structural adhesive. No mixing	Needs good fit and narrow gaps to function effectively. Best below 1mm.
Acrylic 2 part (VOX)	Medium modulus Medium strength	Fast. Easy to apply. Benefit of delayed action cure (DAC). Extremely durable. Fills large gaps. Copes well with light contamination. A true structural adhesive.	Must be mixed
Epoxy 1 part	High modulus Very high strength	Fast curing. Easy to apply. Extremely durable with robust all round performance. No mixing	Needs to be heat cured.
Epoxy 2 part	Medium to high modulus. Medium to high strength	Easy to apply. Durable. Can be speeded by warming/heating. True structural adhesive.	Must be mixed. Slow curing.

As there are many factors to be taken into account the inexperienced designer is strongly recommended to review his application with an adhesive manufacturer before undertaking a commitment. Furthermore, it is also recommended that the proposal is also studied using one of the expert systems now available - PAL being a well known example.