SYNTHESIS REPORT

FOR PUBLICATION

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TITLE. ADHESIVE BONDING OF ENGINEERING PLASTICS (ABEP)

PROJECT COORDINATOR N MAAN
NEDERLANDSE PHILIPS BEDRIJVEN B V
CENTRE FOR MANUFACTURING TECHNOLOGY
5600 MD EINDHOVEN
THE NETHERLANDS

PARTNERS. NEDERLANDSE PHILIPS BEDRIJVEN B V.
CENTRE FOR MANUFACTURING TECHNOLOGY
EINDHOVEN, THE NETHERLANDS

ROBERT BOSCH GMBH
CORPORATE DEPARTMENT PLASTICS
WAIBLINGEN, GERMANY

CIBA POLYMERS
STRUCTURAL ADHESIVES
CAMBRIDGE, UNITED KINGDOM

UNIVERSITAET-GH-PADERBORN
LABORATORIUM FUER WERKSTOFF- UND FUEGETECHNIK
PADERBORN, GERMANY

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Synthesis Report of Brite/EuRam Project 'Adhesive Bonding of Engineering Plastics'.

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Addresses:

Nederlandse Philips Bedrijven, CFT
Patterns & Layers
PO Box 218/ SAQ 1449
5600 MD Eindhoven
The Netherlands

Robert Bosch GmbH
Corporate Department Plastics
PO BOX 1131
71301 Waiblingen
Germany

Ciba Polymers
Struti Adhesives
Duxford
Cambridge CB24QA
United Kingdom

Universität-GH-Paderborn
LWF
Pohlweg 47-49
33908 Paderborn
Germany

Partners / Edited by N. Maan, Philips, CFT.

1. ABSTRACT.

The essence of the work performed in the Brite/EuRam project 'Adhesive Bonding of Engineering Plastics' (Contract No. BRE2-CT92-0203, Project No. BE5365/92, Period 1 January 1993-31 December 1995), has been directed towards the improvement of the joint quality and the development of adhesive bonding processes for engineering plastics/(dis-) similar material bonds.

This has been realized by four main objectives, namely

1. Improvement of the joint quality for engineering plastics/(dis-)similar material bonds by
   1a. Surface analysis and modification%
   lb. Development of adhesive systems.
2. Improvement of the industrial adhesive bonding processes.
3. Predictive meddling (development of new calculation methods to predict the joint strength).
4. Development of new test methods for joint strength measurements.

Note on la.
Surface analysis, before and after (various methods of) surface modification provides a means of (better) understanding of the chemistry of engineering plastic surfaces and their influence on adhesion.
The partners have used various techniques:

- FTIR to determine the type of additives in the plastic materials,
- XPS to determine the bonding state and concentration (atom '%.) of the elements in the plastic materials,
- SSIMS to determine the relative amount of oxygen present at the surface of the plastic material to be bonded.
- Corona, O_2-plasma and W-ozone equipment to pretreat the surfaces to be bonded.
- Contact angle measurements to determine the influence of surface pretreatment on the wetting behaviour.
- Wedge, tensile and torsion tester have been used to determine the strength before and after environmental tests.

Note on lb.
Adhesive systems have been developed for bonding the engineering plastics that fulfil the defined product and process requirements.
Three approaches can be distinguished

- Assessment of the performance of standard high performance adhesives for bonding engineering plastics, including resistance to thermal ageing.
- Development of new easily processable one component adhesives, that have good resistance to warm and humid environments.
- Development of new two component adhesives, that meet the special benefits (high strength, high temperature resistance) and preferably show a fast curing behaviour.

Note on 2.
Equipment and auxiliaries have been developed and modified for reproducible adhesive processes. This has been realized by the development of:

- A special mounting jig for lap shear specimens.
- (Two types of) mounting jigs for the rotationally symmetrical specimens.
- A dispensing unit, provided with a rotary table, meeting the required cycle times.
- A belt oven, meeting the required cycle times.

Note on 3.
A calculation method has been developed to describe the mechanical behaviour of long-term stressed adhesive joints and the extent of the stress level, using the ABAQUS finite element program system, combined with an optimization program.

Note on 4.
For the measurement of strength values of plastic materials a new test method and a new test geometry have been developed. The newly developed rotationally symmetrical test geometries have homogeneously distributed stresses during tensile, torsional or combined tensile-torsional loading.

From the investigations it has been shown, that

- Pretreatment, using O_2-plasma, W-ozone or corona, results in a considerable increase of the joint strength for plastic/adhesive combinations, in general and for polyphenylene sulphide/adhesive and polyetherimide/adhesive, in particular.
- Cleaning of the substrate with isopropyl alcohol is, in general, sufficient enough to obtain good joint strength values; however, pretreatment is essential to polyphenylene sulphide.
- Optimized pretreatment process parameters are available.
- For each type of engineering plastic material, glued to itself or to dissimilar materials, a suitable 1 and/or 2 component adhesive system, that fulfils the requirements with respect to strength and flexibility, before and after environmental testing, has been developed and tested.
- A new test geometry (NTG) has been developed, that fulfils the set demands and that is suitable for the determination of characteristic material parameters.
- A developed failure criterion, used in combination with the NTG, has proved to be a powerful tool for predicting the strength of multiaxial loaded plastic adhesive joints.

2. Introduction.

Adhesives are very often used fix the bonding of high-performance engineering plastics. For their specific benefits, such as high strength (e.g. lap shear $\geq$ 25 N/mm*), high temperature resistance (e.g. $\geq$ 150 °C) and high dimensional stability (in the order of 50 mm), the adhesives, available ox the market, do not guarantee reliable joints or do not fulfil the production requirements (e.g. cycle time < 5 s., no health affection and environmental pollution). Also lack of knowledge about adhesive and plastic surface properties and their influence on adhesion, lack of knowledge about adhesive processing (surface pretreatment, application and curing) often results in adhesive failures.

To make full use of the specific benefits (high strength, high-temperature resistance, dimensional stability), it is necessary to develop adhesive systems that guarantee reliable joints in structural applications and feasible industrial adhesive bonding processes.

The main objectives of this project are

- improvement of the joint quality for plastic/plastic bonds and plastic/dissimilar material bonds,
- improvement of the surface quality of the materials to be bonded (analysis of the surface layers, surface modifications and surface modification methods),
- development of new adhesive systems to meet the specific benefits of the engineering plastics (high strength, high temperature resistance and high dimensional stability).
- improvement of the industrial adhesive bonding processes,
- obtainment of application and curing methods that are industrial feasible, taken into account the avoidance of health and environment problems, for improved codes of practice within the partners' production facilities,
- transfer of knowledge (reports, data base, guide lines), for improved codes of practice within the partners' production facilities.
- predictive modelling (development of new calculation methods to predict the joint strength)
- investigation into and calculation of the mechanical behaviour (stress, strength, durability) of joints under mechanical and environmental load.
- development of new test methods for joint strength measurements.

The organization of this non-confidential synthesis report is based on the 'Tasks', as mentioned in the Work Programme.

- Task 1. Definite selection of engineering plastics. Material procurement.
- Task 2. Surface analysis and modification.
- Task 3. Development of adhesive systems.
- Task 4. Predictive modelling and test methods development.
- Task 5. Laboratory Set-Up.
- Task 6. Sample preparation.
- Task 7. Evaluation of joint performance and durability.
- Task 8. Report and data base.
- Task 9. Guide lines.

Remark: Tasks 6 and 7 have been considered as parts of tasks 2,3 and 4 and have been described in these chapters.

Each task description is subdivided as follows:

- Introduction.
- Technical description.
- Results/Conclusions.

Finally, acknowledgements have been mentioned in chapter 4.

3. Descriptions of the tasks.

3.1. Description of task 1. Definite selection of engineering plastics. Materials procurement.

3.1.1. Partners: Philips
Bosch

3.1.2. Introduction.

Task 1 is dealing with

- The proper choice of engineering plastics, also in combination with dissimilar materials, such as metal, glass and ceramic, from the most recent developments and needs in the partners' product development and production departments.
- The production of the test specimens of various engineering plastics.
- The literature study in order to obtain relevant information about already published results.

3.1.3. Technical description.

Philips/Bosch.

The following engineering plastic materials to be investigated have been chosen:

- Polycarbonate — Apee HT KU 1-9350; Apee HT KU 1-9354 (Bayer),
- Polyphenylene oxide — Noryl SE 1; Noryl PX 1751 (General Electric).
- Liquid crystalline polymer — Vectra A 130; Vectra A 230 (Hoechst).
- Polyphenylene sulfide — Ryton R-4XT (Phillips Petroleum Chemicals).
- Polyetherimide — Ultem 2300 (General Electric).

From these plastic materials a literature study has been made, with respect to their properties, applications and their gluing capabilities, before and after pretreatment. The specific benefits of the engineering plastics have been compared with those of the standard plastic materials.

3,1.4. Results/Conclusions,

The literature study resulted in a report, containing all relevant information about the engineering plastic materials, also in comparison with their 'standard' versions.

The literature study, with respect to the pretreatment and bonding of engineering plastics, resulted in an overview of pretreatment methods and adhesive systems, suitable for the engineering plastic materials under investigation.

The two main conclusions of this study are:

- Several pretreatment methods cause a large improvement of the joint strength,
- Up to now there are no adhesives available, that combine high strength at high temperature with good processability.

3.2. Description of task 2. Surface analysis and modification.

3.2.1. Partners: Philips
Bosch

3.2.2. Introduction.

Task 2 is dealing with

- The obtainment of comprehensive understanding of the material composition of surface layers (e.g. moulding skins), including the effects of environmental influences (e.g. humidity, temperature).
- The obtainment of relevant understanding of the influence of the nature of surface layers on the adhesion.
- The obtainment of knowledge of the effects of surface modification (e.g. O_2-plasma, corona, UV-ozone) on the nature of surfaces, including the effects of time (e.g. storage time) and environmental influences (humidity, temperature).
- The determination of the influence of surface modifications on adhesion, including effects of environmental influences,

3.2.3. Technical description.

Philips.
In order to gain more insight into the relation between adhesion and surface chemistry the surface of the chosen engineering plastics were analyzed. The engineering plastics have been investigated before and after solvent cleaning (isopropylalcohol) using X-ray Photoelectron Spectroscopy (XPS) and Static Secondary Ion Mass Spectroscopy (SSIMS).
The influence of several pretreatment (isopropylalcohol cleaning, corona, O_2-plasma, UV-ozone) on the wetting behaviour (contact angle) and the adhesion (lap shear joint strength) of the surfaces of the selected engineering plastic materials has been investigated.
At first, these joint strength experiments were carried out, using a well-known epoxy adhesive, because newly developed and/or modified adhesive systems were not yet available. Later on, newly developed and modified adhesive systems have been used.

Bosch.
Different pretreatment methods (e.g. corona, O_2-plasma) have been carried out to modify the surfaces of engineering plastics. The effects of pretreatment on the wetting behaviour of plastic surfaces have been investigated using the Wilhelmy Plate Method (WPM) and the Sessile Drop Method (SDM), the effects on the surface structure have been investigated using the Scanning Electron Microscope (SEM), the effects on the adhesion have been investigated using the Tensile Tester and the Wedge Tester.
Adhesion experiments have been carried out on both conditioned and not-conditioned samples.

3.2.4. Results/Conclusions.

Philips.

From the surface analysis, carried out with XI%, it can be concluded, that:

- The surface compositions of the not pretreated engineering plastic surfaces correspond to the main bulk component; only polyphenylene sulfide (Ryton) shows a slight oxidation of the surface.
- After isopropylalcohol cleaning, no contaminants were detected; only on the polyphenylene oxide (Noryl) surface some inorganic contaminants were found.

From the surface analysis, carried out with SSIMS, it can be concluded, that:

- Peaks, characteristic for each engineering plastic were observed.
- Cleaning of the plastic materials results in a considerable reduction of contaminants and additives at the surface.
- W-ozone oxidizes the polymer surfaces, resulting in new oxygen containing groups, characteristic for each polymer.
- W-ozone pretreatment on polyphenylene oxide (PPO) results in the formation of low molecular weight material, which partly can be removed by a solvent cleaning.
- Comparison of results from literature on O_2-plasma modification and the UV-ozone modification shows that both treatments yields similar modified polymer surfaces.
- Low molecular weight additives in plastics tend to migrate to the substrate surface. Enhanced temperature stimulates this migration. The occurrence of these additives at the substrate/adhesive interface may have a deteriorating effect on the bond strength.

From the investigation into the influence of pretreatment on the wetting behaviour, it can be concluded, that:

- The contact angle measurements show that corona, O_2-plasma and UV-ozone pretreatments have big influences on the contact angle. After 2 s (corona), 5 min. (O_2-plasma) and 10 min. (UV-ozone) the contact angle decreases from about800 to 30'.
- The influence of storage on the contact angle is different for the examined pretreatment methods. W-ozone shows hardly differences in contact angle during, at least, the first two days. However, corona and O_2-plasma show already after a few hours considerable increases, The effect of each pretreatment on the polycarbonate surface maybe different.

From the investigations into the influence of pretreatment on the joint strength, it can be concluded, that

- Corona, Q-plasma and UV-ozone pretreatment has hardly any influence on polycarbonate, polyphenylene oxide and polyetherimide. Only on polyetherimide pretreatment results in an increase in bond strength, when W'-curable or UV-gelable epoxy adhesives were used.
- It is difficult to determine the influence of pretreatment on the joint strength of liquid crystalline polymer, because in almost all cases cohesive substrate failure occurred. This means that bond failure occurs at the interface between the surface layer of LCP and the bulk material.
- Pretreatment has a tremendous influence on polyphenylene sulfide. The initial strength as well as the strength after environmental testing increases strongly.

Isopropylalcohol extracts of the plastics were analysed by FTIR.

It appeared that:

- All plastic materials contain aliphatic fatty ester, possibly an antioxidant.
- Noryl contains diphenyl cresyl phosphate, a flame retardant.

Bosch.
From the investigations into the optimization of the surface pretreatment, it can be concluded, that:

- The surface state of these engineering plastics improves by both corona and O_2-plasma treatment.
- An increasing surface tension, caused by both treatment methods, is measurable by the Wilhelmy Plate Method (WPM) as well as the Sessile Drop Method (SDM); however, the reproducibility of the Sessile Drop Method is much better.
- Both treatment methods have an influence on the surface morphology of the engineering plastics; the changes in surface morphology depend very strongly on the type of engineering plastic.

From the investigations into the influence of surface pretreatments on the adhesion, it can be concluded, that:

- Pretreatment causes an improvement of the adhesion for liquid crystal polymer (LCP), polyetherimide (PEI) and polyphenylene sulphide (PPS).
- The adhesion improvement depends on the type of adhesive.
- The lap shear strength values of (anisotropic) LCP is rather low, which is caused by the delamination of the upper layers, resulting in a cohesive substrate failure.
- The wedge test gives a better indication of the strength behaviour than other strength test, because the wedge test shows the behaviour of the joint under mechanical load combined with climatic conditions. As a result a definite statement about the bonding characteristic can be given within a comparatively short test time.

From the investigations into the influence of conditioning, it can be concluded, that:

- The moisture content in PEI strongly depends on the conditions; moisture content, temperature and sample geometry have a large influence on the dehydration sped
- At curing temperatures ≥ 120 °C pores in the adhesive layer can be formed, if PEI is not predried.

3.3. Description of task 3.
Development of adhesive systems.

3.3.1. Partners:

Ciba,	sub-tasks 3.1, 3.2, 3.3.
Philips,	sub-task 3.4.
Bosch,	sub-task 3.4.

3.3.2. Introduction,

Task 3 is dealing with

- The screening and evaluation of the performance of standard high performance adhesives.
- The development and testing of new one component easily processable adhesives.
- The development and testing of adhesives with improved properties - which could cover two component adhesives.
- The verification of targets, as defined in the above mentioned sub-tasks.

3.3.3. Technical description.

Ciba.
As the first stage of adhesive development, it was necessary to study the perfikm.ante of misting commercial adhesives on the selected Engineering Plastics, in order to establish the current capabilities, and to provide information upon which further development could be based.
The adhesives were evaluated by simple lap shear tests using degreased substrates, tested after bonding and again after thermal ageing. Full definition of the lap shear strength test methods and thermal ageing conditions are described elsewhere.
A summarised process is as follows:

- Standard 120 mm x 25 mm x 2 mm test specimens supplied by Philips and Bosch
- Surface to be bonded washed with isopropyl alcohol and air dried
- Adhesive applied to one cleaned surface
- Spacer wires or Ballotini applied to the adhesive film
- Second substrate surface applied with 5 mm overlap, and clamped in place
- Adhesive cured under prescribed conditions
- Lap shear tests pulled at 23 °C and a speed of 5 mm/min
- Thermal ageing in ovens at prescribed temperature for 100,250,500 or 1,000 hours
- Lap shear tests pulled at 23 °C as above
- Failure modes recorded as defined in ISO 10365

Adhesives were selected from the current Ciba commercial range to exemplify one and two component epoxy adhesives, two component polyurethane adhesives and UV curable epoxy adhesives.
Based on the results from Sub-task 3.1 a variety of thermally cured and UV activated/thermally cured epoxy adhesives were developed with several being selected as being of interest.
Based on the initial work described in Sub-task 3.1, a limited effort was made to produce improved two component epoxy adhesives Two component adhesives can be applied easily as a ready mixed material by use of cartridge/mixer technology similar to that used in the existing Araldite 2000 range, or using specialist mix/metering machines.

Philips.
Philips has verified the adhesive systems, developed by Ciba, before and after pretreatment. Moreover, the strength of the adhesive joints (lap shear specimens and rotationally symmetrical specimens) have been measured before and after environmental testing (dry heat test, damp heat test and rapid change of temperature test).
The 'best' adhesive systems have been tested the rotationally symmetrical specimens, made of polycarbonate, polyphenylene oxide and liquid crystalline (table 1).

Table 1. Test matrix for new test geometries.
(rotationally symmetrical specimens)

Adhesive	Type	PC	PPO	LCP
Araldite 2005	2 c epoxy	x		
Araldite 2007	1 c epoxy		x	x
Araldite 2011	2 c epoxy		x	
LMD 1313/1314	2 c epoxy			x
LMD 1193/1194	2 c polyurethane	x		

The adhesive systems, developed for the chosen engineering plastic materials are very often used in combination with dissimilar materials, such as steel, aluminium, ceramic or glass. For that reason, the performance of the developed adhesives on non-plastic materials is of interest. The best performing adhesives were used to determine the joint strength behaviour of these materials in combination with engineering plastics (table 2).

Table 2. Chosen adhesive systems for (dis-)similar materials.

adhesive	type	curing conditions
Araldite 2007	1 c epoxy	150 °C/15 min.
LMD 1301 + XD 4416	2 c epoxy	100 °C/20 min.
LMD 1221	UV-gelable epoxy	130 °C/20 min.
LMD 1193/1 194	2 c polyurethane	120 °C/20 min.

Differences in thermal expansion between the (dissimilar material) substrates give rise to internal stresses in the adhesive layer. The magnitude of these effects depend on different parameters, such as overlap length, glass transition temperature (T_g) of the adhesive, flexibility (E-modulus) and thickness of the substrates and the adhesive layer.
Calculation of this internal stress is possible, but the calculated values usually tend to be rather pessimistic. Therefore, the flexibility of adhesive joints between dissimilar materials was examined quantitatively as a function of overlap lengths, thicknesses of the non-plastic material and environmental tests.

Bosch.
The development of adhesive systems required the measurement of the bond strength of the selected plastic materials (in dependence on the surface treatment, the ageing conditions or the test temperature) bonded with proper standard and newly developed adhesives (proposed by Ciba).
Based on these investigations adhesives could be chosen, respectively developed which are suitable for bonding the selected engineering plastics.
For the investigation into the properties of glued (dis-)similar materials different material combinations of engineering plastics, glass, alumina and aluminium were tested. The properties of these joints were evaluated with lap shear tests and deformation measurements.

3.3.4. Results/Conclusions.

Ciba.
From the tests the following standard adhesives were selected as being of interest:
- Araldite AV4076/HY4076 (2005) - Noryl SE 1, Noryl PX 1751
- Araldite AW106MV935U (201 1) - Noryl SE 1, Noryl PX 1751
- Araldite AV119 (2007) - Noryl SE 1, Noryl PX 1751
- Araldite 2018 - Noryl SE 1, Ultem 2300, Apec HT KU 1-9350, Apec HT KU 1-9354
- Araldite 4004 - Noryl PX 1751, Apec HT KU 1-9350, Apec HT KU 1-9354.

From the development and testing of new one component easily processable adhesives the following thermally cured and UV activated/thermally cured epoxy adhesives were developed.

- LMD1204 - Vectra A 130, Vectra A 230, U'Item 2300
- LMD1369 - Vectra A 130, Vectra A 230, Ultem 2300
- LMD1196 - Ryton R-4XT, Vectra A 130
- LMD1312 - Ryton R-4XT, Ultem 2300
- LMD1197 - Noryl PX 1751, Apec HT KU 1-9350, Apec HT KU 1-9354
- LMD1221 - Noryl PX 1751, Ultem 2300> Apec HT KU 1-9350

From the development and testing of adhesives with improved properties the following two component adhesives were developed.
The selected new two component epoxy adhesives give results of interest on the following substrates:

- LMD1313/LMD1314 - Noryl SE 1, Noryl PX 1751, Ryton R-4XT
- LMD1301/XD4416 - Noryl SE l, Noryl PX 1751, Vectra A 130, Vectra A 230, Ultem 2300.

Based on the initial work described in Sub-task 3.1, a higher Tg variant of two-component polyurethane XD4436/XB5304 was developed in a 1:1 mixing ratio, suitable for cartridge application or use with mix/metering equipment.

LMD1193/LMD1 194 has a cured Tg of 83 "C, which has a high tensile strength and modulus, is of interest on Noryl SE 1, Apec HT KU 1-9350 and Apec HT KU 1-9354. The very short potlife (< 10 min.) of this adhesive is a disadvantage.

A variety of optimisation experiments were carried out on the selected adhesives. Generally these did not result in any significant improvement to the original formulations.
Optimisation experiments included

- Araldite AV119 - level and combination of curing agent and accelerators;
- Araldite 2011 - plasticiser content;
- LMD1204 - level and combination of curing agent and accelerator;
- LMD1369 - level of thixotropy;
- LMD1196 - level and combination on curing agent and accelerator;
- LMD1197 - optimisation of formulation with_ to "curing window".

From the investigations into the development of adhesive systems, it can be concluded, that

- Both from the screening programme and the subsequent development pmgramme, good progress has been made in the bonding of engineering plastics over their workable temperature range, the lists showing the selected adhesives indicate the most successful candidates for each substrate and it is encouraging that there are adhesives suitable for joining all five types of material.
- It is not possible to define adhesive chemistry which can be used to bond all of the substrates in the project in the form of one adhesive formulation.
- In some cases two component epoxy or PUR adhesives give better results than one component products.
- Apec polycarbonates are more suited to bonding by two component polyurethane adhesives, than by one or two component epoxy products.

- UV curing epoxy adhesives can be used to bond Noryl, Ultem and Apec.
- Flexible polyurethane Araldite 2018 can be used to bond some dissimilar substrates since the low Tg permits differential expansion of the component parts.
- Two component acrylic/methacrylic adhesives evaluated do not give adequate thermal ageing characteristics on the selected plastics.
- The failure mode and therefore long term performance could be improved in many cases by a suitable pretreatment method.
- Chemical toughening of the adhesives is not always beneficial in the thermal ageing performance of the adhesives, probably depending on test geometry.
- Fast cure of the adhesives can either be achieved using a thermal cure of a two-component adhesive, or, it must be accepted that a minimum cure temperature of 120 °C must be used for curing of one component adhesives due to considerations of storage and transport. Only W activated products can combine ambient temperature stability and rapid cure at 40 - C .

Philips/Bosch.
From the verification of the joint strength, using the new and old test geometry, as well, it can be concluded that:

. The new test geometry (rotationally symmetrical specimens) is better suited for strength measurements of adhesive joints than the old test geometry (lap shear specimens). In almost all cases much higher values, before and after environmental testing (dry heat test, damp heat test), are obtained.

From the investigations into the bonding of non-plastic materials, it can be concluded, that:

- Most of the adhesives that show good results on the selected engineering plastics, also perform well on steel, aluminium, ferroxcube and glass.
- The 1 and 2 component epoxies perform well on all substrates; either substrate failure or adhesion failure at strength > 30 MPa occurs.

. The 2 component polyurethane and the UV-gelable epoxy perform somewhat less; bond strengths > 20 MPa can be obtained if acid pickled aluminium is used.

. The UV-initiated epoxy performs very moderately, which is probably caused by the initiation of this adhesive, which up to now is not very reproducible.

From the investigations into the bonding of dissimilar materials, it can be concluded, that:

- In bonding dissimilar materials large internal stress in the adhesive layer may occur, resulting in strength values, which are lower than on the individual materials. On glass very low values are obtained and in several cases the bond strength is zero.
- Gap widths of 300 mm instead of a 100 mm adhesive do not change the bond strength significantly.

It is recommended to use flexible adhesive systems (low or moderate E-modulus), if stiff materials and/or large overlap areas are used.

From the investigations into the flexibility of adhesive systems on the adhesive joint strength, it can be concluded, that:

- In general, the internal stresses in the adhesive layer are not sufficient to destroy the bond; only at lower temperatures the internal stresses in the adhesive layers become so large that joint failures occur.

- In case of large overlap length and/or stiff substrate materials, best results are obtained with adhesive systems with a relatively low modulus of elasticity (E) and a low glass transition temperature (T_g).

3.4. Description of task 4.
Predictive modelling and test methods development.

3.4.1. Partners:

Partners:	Uni-Paderborn,	sub-tasks 4.1,4.2,4.3,4.4,4.5, 4.6,4.7.
	Philips,	sub-tasks 4.2, 4.3, 4.5, 4,6,4.7.
	Bosch,	sub-tasks 4.2, 4.3.

3.4.2. Introduction.

Predictive modelling for adhesive joints is much less advanced than for other joining techniques. Moreover, most of the knowledge available is developed for adhesively bonded metals. For plastic materials much larger deviations from linear behaviour are expected, which hamper modelling. Therefore it was decided, and in addition due to the still increasing use of engineering plastics, to develop calculation methods for adhesively bonded plastics.
Nevertheless predicting the mechanical characteristics of adhered components can be extremely difficult due to the complex material laws.
Task 4 is dealing with:

- The development and verification of calculation methods that predict, for the given plastic materials, the mechanical behaviour of the adhesive joint.
- The development of test methods and equipment, that matches better the strength properties of the adhesive joints than usually standard test methods and equipment for metals, for verification of the predicted joint strength values.
- The development of test methods and equipment to determine creep.
- The determination of the creep behaviour and its influence on the joint strength

3.4.3. Technical description.

Uni-Paderborn.
With the Finite-Element-Analysis it is possible to calculate plastic adhesive joints under multiaxial stress conditions if the material parameters of the adhesives are known. Otherwise the quality of adhesive bonds can only be judged either with large-scale component tests or in assistance with standard lap shear tests. These experiments only produce an average value of failure. The single lap joints show a strong deformation under load due to their inferior stiffness, which makes them useless for the determination of material parameters. Therefore a new test geometry has been developed.
This new test geometry also allows to determine, in a very easy way, the coefficients of the failure criterion, created for plastic adhesive, using the results of uniaxial tests.
For obtaining material parameters to describe the behaviour of long term stressed adhesive bonds an optimization algorithm has been developed.

Philips.
Philips have developed creep measuring methods and equipment to determine the creep parameters under combined loads (tension, torsion, tension/torsion).
Together with the Uni-Paderborn creep experiments have been carried out.

Bosch.
For the production of test specimens (NTG) an ejector pin type injection mould was made.

3.4.4. Results/Conclusions.

Uni-Paderborn.
The new developed test geometry, fig. 1, represents an excellent tool to get material parameters of adhesives in order to calculate bonded structures very fast.
In contrast to the standard lap shear specimen the new developed test geometry fulfils following demands:
- Producibility of uniaxial and defined multiaxial stress conditions.
- Homogeneous stress conditions inside the adhesive layer, fig. 2 (next page).
- Maximum stress in the adhesive layer, so that in most eases the adhesive fails instead of the specimen.

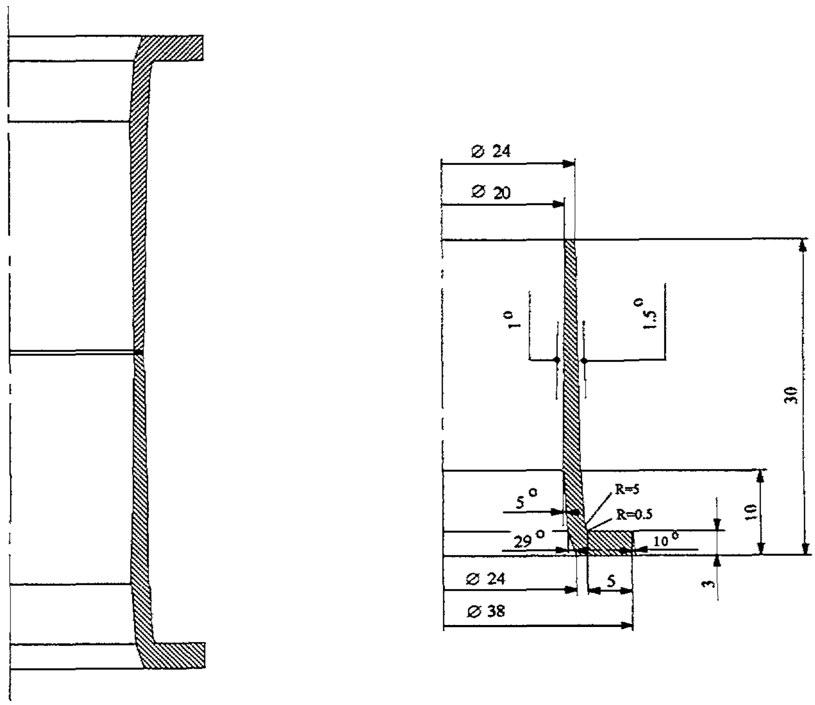
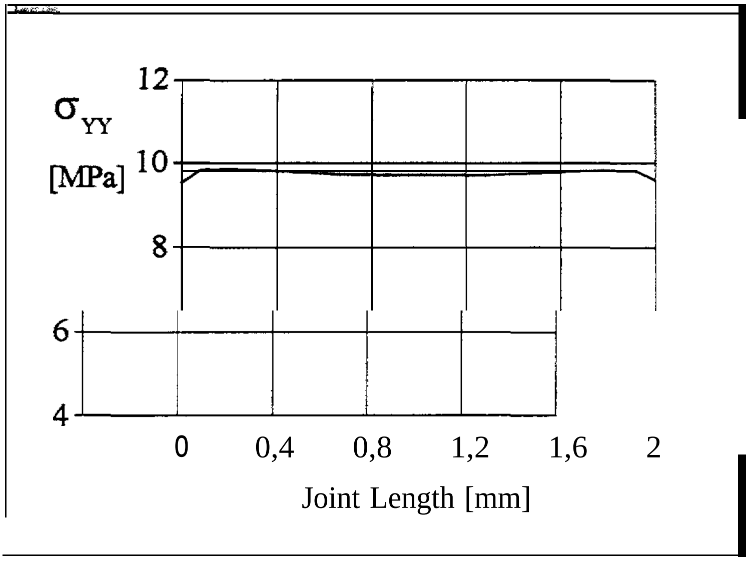
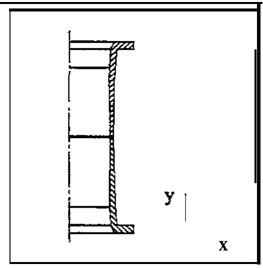


Figure 1. Stress optimized specimen for measuring the joint strength of high performance engineering plastics.



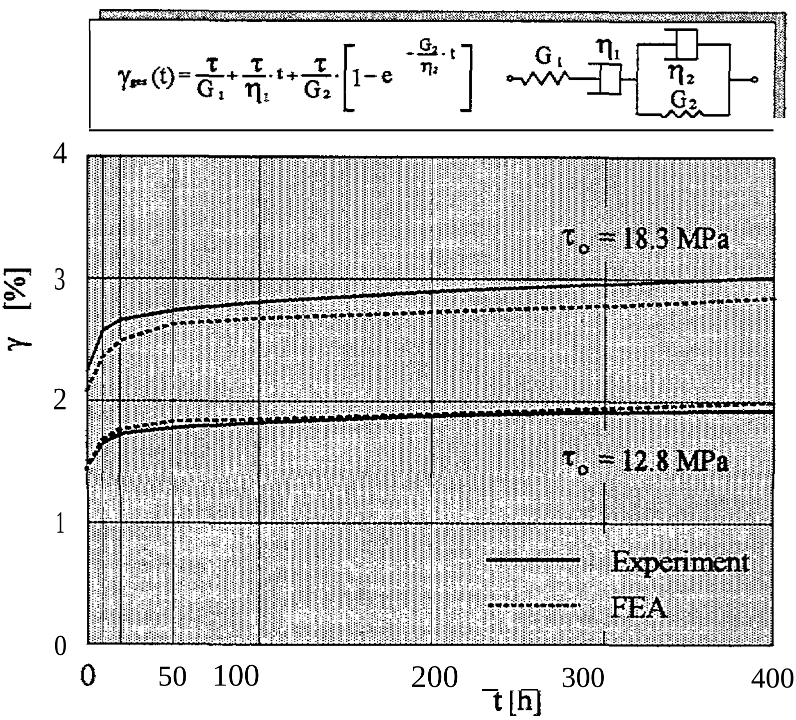


Young's Modulus
- Adherends: 2250 MPa
- Adhesive: 1500 MPa

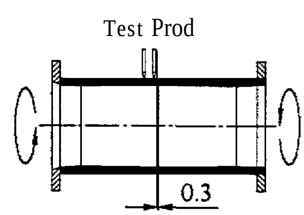
Bondline Thickness; 0.3 mm

Fig. 2: Butt tubular joint under tension (bondline stresses).

Figure 3 shows the good results of the optimization programme. The curves of the calculated creep values fit rather well to those of the experimental values.



$$\gamma_{res}(t)=\frac{\tau}{G_1}+\frac{\tau}{\eta_1}\cdot t+\frac{\tau}{G_2}\cdot\left[1-e^{-\frac{G_2}{\eta_2}\cdot t}\right]$$



Load: Torsion

Temperature 23° C

Humidity: 50 %

Adhesive: AV 119

Adherends: NORYL SE1

Figure 3, Plastic butt tubular joint under torsional load.

Experiments also showed, that in some cases it is possible to use material parameters of adhesives, obtained from tests with metal specimens, for plastic adhesive bonds (figure 4).

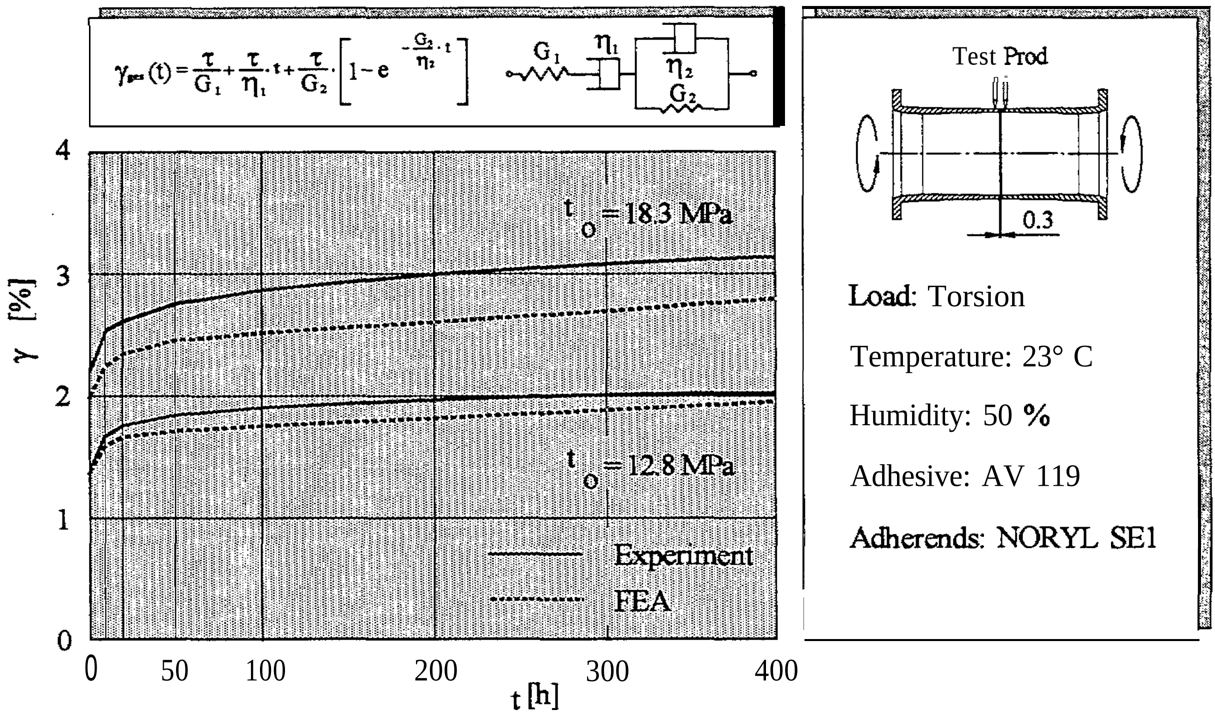


Figure 4. Comparison between predicted and real creep behaviour of torsional loaded plastic butt tubular joints, using material parameters, obtained from adhesively bonded metal tubes.

From the development of the calculation methods (predictive modeling), it can be concluded, that:

- With the aid of the developed tool for predictive modelling, using the new test geometries and methods material parameters on the basis on experimental results can be determined.
- With the determined material parameters stresses, strengths and deformations of adhesively bonded joints in loaded constructions can be calculated

Philips.

From the development of creep methods and equipment and from the investigations into the creep behaviour, it can be concluded, that:

. The (torsional creep) equipment, we built in the beginning, is not suitable for our investigations, because creep results, obtained from these creep experiments, cannot be used to predict the creep behaviour under combined loads.

- The measuring device, we built for the new test geometries, is able to measure the rotational displacement reasonably well; however, it does not fulfil the expectations with respect to the axial displacements and, as a consequence the combined displacements.

. Therefore, only torsional creep tests executed with the device have scientific meaning.

Bosch.
From the experiments, it can be concluded, that:

- The joint strength, using the new test geometry, is independent on the used test equipment.
- The new test geometry is very useful to determine" the strength under tensile, torsion and combined loads.

3.5. Description of task 5. Laboratory set-up.

3.5.1. Partner: Philips.

3.5.2. Introduction.

Task 5 is dealing with:

- The development of mounting equipment, that allows reproducible production of 'old geometry' test samples.
- The development of mounting equipment, that allows reproducible production of 'new geometry' test samples for the verification of predicted stresses and displacements in adhesive layers.
- The development of mounting equipment, that allows reproducible production of 'new geometry' test samples for 'mass production.
- The development/modification of an application system for circular adhesive beads.
- The development/modification of a curing system to fulfil the process times.

3.5.3. Technical description.

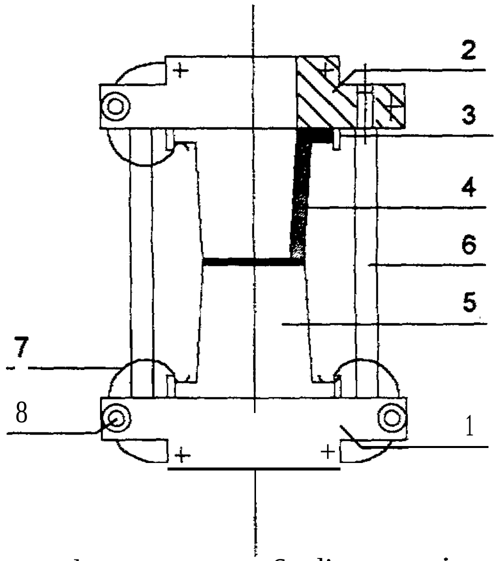
For the production of lap shear specimens the Bosch concept has been used. This means, that samples have been made with an overlap of 5 mm in stead of 12.5 mm to prevent cohesive substrate failures.
For the verification of the predicted strength of an adhesive joint a special mounting jig has been developed and produced. This mounting jig should guarantee a constant, accurate gap width during curing.
For the production of test samples to determine the strength of adhesive systems, a simpler version has been developed and produced.
For these three types of mounting jig a manual has been written.
For the application of the adhesive systems on rotationally symmetrical samples (circular beads) a standard dispense unit, provided with a rotary table has been modified to enable homogeneous circular beads.

3.5,4. Results/Conclusions.

From the development of mounting jigs, it can be concluded, that

- The mounting jig for modified lap shear specimens allows very well the comparison of the results of the different partners.
- The mounting jig for the new test geometry to verify the calculated values {figure 5, next page) allows a very reproducible preparation of very accurately defined adhesive layers.

. the mounting jig for mass production allows the quick preparation of test samples for only one gap width.
- the video film clearly shows the benefits of the newly developed mounting jigs and auxiliaries, compared with the 'usual method'.



1. ring-shaped lower plate
2. ring-shaped upper plate
3. positioning pin
4,5.test specimen halves
6. distance piece
7. spring
8. bolt

Figure 5. Mounting jig for plastic tensile-shear and torsional-shear specimens.

3.6. Description of task 8. Reports/Data base.

3.6.1. Partners: Philips
Bosch
Ciba
Uni-Paderborn.

3.6.2. Introduction.

One of the objectives of this project was to disseminate the knowledge about adhesive technology with respect to joint design, engineering plastic materials, adhesive materials, surface modification, application and curing of the adhesive systems and calculations, obtained during the project. It was proposed to disseminate this knowledge by means of reports and a data base.

3.6.3. Technical ascription,

AU results of completed (sub-)tasks have been reported in technical reports by all partners and have been sent to Brussels, as agreed. These results have also been elucidated to and discussed between the partners during the half-yearly progress meeting.
A start was made with respect to the database. After a very carefully executed inventarisation, it pointed out, that there are no suitable data base systems available for our purposes, i.e. a system, that allows a quick answer with respect to the adhesives to be used for different engineering plastic materials and their respective adhesive technologies. In the meantime a data base system was in preparation within Philips and we have proved the suitability of this system. This system consists of a part 'Description of Configuration' containing the adhesive technology, in general, and four parts 'Knowledge Base'. Up to now only the part 'Diagnosis', containing the ABEP test-observations, is ready and this part only shows the causes of possible failures and does not allow the choice of proper adhesive systems for chosen constructions and materials. Because the finishing of the part 'Design' takes too long, the participants have agreed to use the different reports, in which the results of the investigation are mentioned, as a data base, i.e. to use for the proper choice of the adhesive systems.

3.6.4. Results/Conclusions.

From the work, carried out on reports and data base, it can be concluded, that
. The results of all (sub-)tasks have been extensively reported by all partners.
. All reports have been passed to Brussels.
• The data base could not be finished, because suitable systems are not available yet.

3.7. Description of task 9.
Guide lines.

3.7.1. Partners: Philips
Bosch.

3.7.2, Introduction.

This task is dealing with the transfer of knowledge to design and development engineers of production facilities about proper production methods and equipment, with respect to surface modification, application and curing of the adhesive systems, taken into account the prevention of health affection and environmental pollution.

3.7.3, Technical description.

Philips/Bosch.
All items, which have been part of the investigations during the project have been laid down in guide lines.

3.7.4. Results/Conclusions.

Philips.
The guide lines give

- A description of the mounting jigs to obtain reproducible test samples for
  - plastic lap-shear specimens
  - plastic tensile and torsional shear specimens for
    - the verification of the predicted joint strength
    - for 'mass production'
- Description of the adhesive technology with respect to adhesive bonding of engineering plastics for
  - the pretreatment of the surfaces to be bonded, including measuring methods to determine the influence of the pretreatment
  - the application methods, including equipment to realize reproducible joints, using the new test geometries
  - the curing methods to realize optimally cured joints
- a description of the safety and hygienic precautions.

In addition, the moulding jigs and the moulding conditions to obtain

- Reproducible test sample geometries for
  - lap shear test specimens (the so-called 'old test geometries')
  - rotationally symmetrical test specimens (the so-called 'new test geometries').

Bosch.
The guide lines give descriptions of

- The surface treatment of the adherends
- Storage of adhesives till processing
- Processing with adhesives
- Disposal considerations for adhesives
- Instructions and advices to the industrial and health safety
- Standards for adhesive bonding technology

4. Acknowledgements.

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