CRITICAL STEPS IN ADHESIVE BONDING PROCESS FOR SPACE APPLICATIONS

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GLOSSARY

2-C	Two component
AF	Adhesive failure
CF	Cohesive failure
CME	Moisture expansion coefficient
CMR	Carcinogenic, mutagenic or toxic to reproduction
CTE	Coefficient of thermal expansion
ECSS	European Cooperation for Space Standardization
EWF	European Federation for Welding, Joining and Cutting
HT	High Temperature
MPTB	Materials and Processes Technology Board
RH	Relative Humidity
RT	Room temperature
SCF	Superficial Cohesive Failure
TDS	Technical Datasheet
Tg	Glass transition temperature

ABSTRACT

The surface preparation and curing regimes belong to the key steps for success in the case of adhesive bonding and are currently under investigation in ESA/ESTEC in collaboration with Rescoll. A general overview of the adhesive bonding for space applications and its criticalities in this context is summarised in this work, focusing in particular on the importance of surface treatment, accelerated exposure conditions and impact of the curing regime on adhesive joint performance.

Experimental part consisted of series of mechanical tests on standard lap shear samples which were performed before and after exposure to humidity. This paper also summarises results from tests performed so far on “difficult” substrates such as FeNi36 alloy and standard aluminium AL 2024 T351 substrate.

Further work will continue to support the MPTB initiative to assess the impacts of formulation changes anticipated by key adhesive manufacturers (optimisation of manufacturing process, obsolescence and changes driven by REACH restrictions). Preliminary results from the “formulation change” assessment for selected adhesives are also part of this work.

1. INTRODUCTION

Use of adhesive bonding for space applications brings advantages, such as flexibility, joining of dissimilar materials and mass savings, but it is also a common source of failures. The root causes of these failures are often attributed not only to performance losses due to exposure to simulated space environment during testing phases, but also due to inefficiency of surface treatments of the substrates prior to bonding, improper curing and exposure of the manufactured joints to ambient atmosphere during the entire on-ground life cycle.

In particular, adhesive bonding is frequently used in the design of optical instruments and may represent significant challenge for the manufacturers. Such bonding must be able to withstand the harsh environmental conditions, such as enhanced humidity, thermal cycling between two temperature extremes, long-term thermal exposures in vacuum, the impact of vibrations and shocks during the launch phase, all without any significant drop in performance of the bonded optical assembly and with retaining of alignment and dimensional stability.

1.1. Quality assurance in adhesive bonding technology

There is also another source of challenges for adhesive bonding, which is the production of the adhesives themselves. Quality assurance aspects of the production can be questioned when it comes to long-term stability in performance of a product. Manufacturing processes, as well as techniques for raw and cured material characterisations evolve. Therefore it is necessary to stress the importance of unmixed and cured (bulk) characterisation when selecting the adhesive and during its utilisation [1-2]. The selection of suitable adhesive for any bonding process is one of the most critical steps in the adhesive bonding qualification and therefore it will

Proceedings of the 14th ISMSE/12th ICPMSE, Biarritz, France, 1 to 5 October 2018.

be addressed in the new product assurance standard ECSS-Q-ST-70-16C [2].

1.2. Progress on ECSS-Q-ST-70-16C, Adhesive bonding for spacecraft and launcher applications

High demand for a better quality and reliability in adhesive bonding technologies resulted in setup of a European Adhesive Bonder training programme and certification scheme under auspices of EWF [3]. Similar as per DIN EN 15085 „Welding of railway vehicles and components“, the adhesive bonding operators are recently being trained and certified according to the standard DIN 6701 „Adhesive Bonding of Rail Vehicles“ which became the mandatory standard for application of adhesive bonding technology on railway. ESA together with national space agencies and EUROSPACE initiative set up the ECSS working group to draft the standard dedicated to adhesive bonding. This product assurance standard, ECSS-Q-ST-70-16C, *Adhesive bonding for spacecraft and launcher applications*, is currently under a public review [2] and is dedicated to an adhesive bonding process, listing requirements and recommendations specific to the space sector.

1.3. ESA's contribution to MPTB and current challenges with adhesive formulation changes

Due to regular changes (usually announced by the adhesive manufacturers) in the formulation of some of the 2-C (two component) epoxy adhesives, ESA in collaboration with external partners runs extensive testing of the most common cases and assesses the criticality and impact of the change in commercially available aerospace grade products. Lately also some of the 3M's 2-C epoxy aerospace grade adhesives underwent changes in the adhesive manufacturing process, and after the testing of "pilot" scale batches, 3M proceeded with the commercialisation of the modified products (industrial scale). The assessment of these cases is ongoing as a part of support to the MPTB initiative. Philosophy of the "formulation change assessment" testing is typically based on comparison of the mechanical performance of the "current" and "modified" adhesive version. While some of the adhesive manufacturers clearly identify their "modified" products after formulation change by changing the name/code (e.g. the latest change of Huntsman's hardener HV998 and CMR-free version HV998-1), others do not. This also depends on the extent of the changes. The lack of clarity in traceability of the "modified" products may cause a lot of confusion on the market among the distributors and challenge the end user who is trying to characterise the "new" formulations within his internal delta-qualification exercise. Part of this paper is focusing on assessment of the formulation changes in two 2-C epoxy resins, which were identified by the MPTB community to be the most critical ones.

2. EXPERIMENTAL

There are three major paths to characterise the adhesives:

- <u>characterisation</u> of the <u>unmixed materials</u> (viscosity, colour, density etc.),
- <u>characterisation of the curing process</u> (viscosity, pot life, reaction heat, polymerisation volume shrinkage etc.)
- <u>characterisation of the cured material</u> (bulk properties such as: density, outgassing in thermal vacuum, tensile & shear strength, elongation at break, elastic moduli, Poisson ratio, glass transition temperature, cure degree/conversion ratio, post-cure shrinkage, CTE, CME, and performance in assembly, such as: lap shear strength, peel strength, fatigue & creep resistance, fracture toughness, resistance to specific environments etc.)

This paper is mostly focused on the performance of adhesive in bonded assemblies, in particular on adhesion strength, determined by standard single lap shear samples before and after exposure to humidity.

Part of this work is also dedicated to bulk characterisation of cured 2-C epoxy adhesives (tensile strength and elastic modulus, elongation at break & Poisson ratio) for the "current" and "modified" versions of selected adhesives. Major part of the tests and exposures was carried out by Rescoll. Test assessing the influence of the operator factor took place at ESTEC laboratories.

2.1. Efficiency of surface treatment of FeNi36 alloy (INVAR®) to improve adhesion strength

These tests focused on the assessment of lap shear strength as a function of surface treatment. The single lap shear samples were made of 1.6 mm thick FeNi36 alloy (INVAR®) plates (Annealed Deep Draw, ASTM F1684). The surface conditions were the variable parameter, following: 1, 1+2, or all three consecutive treatment steps (1+2+3):

Treatment 1: Grinding paper 320 & acetone bath
Treatment 2: Treatment 1 + chemical: degreasing bath
Treatment 3: Treatment 2 + priming: EC3901 primer

The 3M's ScotchWeld^{TM} 2216 B/A Gray structural adhesive was cured at room temperature (RT), followed by post-cure at 65 °C, for 2 hours. There were two sample sets (6 samples per set) per surface treatment type. The fracture surfaces were analysed according to NF EN ISO 10365 for definition of fracture modes. There were 12 samples per each sample category out of which 6 samples were exposed to elevated temperature and humidity ("hot-wet") conditions (7 days, 95% RH, 55°C) and reconditioned to bring the sample moisture content into equilibrium with the ambient laboratory conditions. The reconditioning phase after humidity exposure lasted 72 hours and efficiency of reconditioning was monitored by mass monitoring of the adhesive in-process samples. The mechanical test was conducted according to the standard test EN 1465.

2.2. Effect of curing temperature on the durability of the 2-C epoxy adhesive joints

This part of the study focused on the combined influence of the curing temperature and surface treatment on durability of adhesively bonded joint on two different types of metallic substrates. The single lap shear samples were made of 1.6 mm thick aluminium plates (AL 2024 T351), as well as 1.6 mm thick FeNi36 alloy (INVAR®) plates (Annealed Deep Draw, ASTM F1684). The surface treatments followed the same philosophy as the one described in previous paragraph with differing surface treatment quality. The 3M's ScotchWeld™ 2216 B/A Gray structural adhesive was applied and cured at RT; some lap shear samples were cured for 2 days at RT and post-cured at 65 °C, for 2 hours. There were 12 samples per each sample/curing category out of which 6 were exposed to "hot-wet" conditions (7 days, 95% RH, 55°C) and reconditioned afterwards. The reconditioning phase after humidity exposure lasted 72 hours. During reconditioning the mass change of the in-process sample (mass travellers) was monitored. The mechanical test was conducted according to the standard test EN 1465.

2.3. The influence of the formulation changes on the "as-cured" properties

To assess the formulation changes of the recently modified EC2216 B/A Gray and EC9323-2 B/A Black, tensile strength, elastic modulus, elongation at break and Poisson ratio were determined according to ISO527-2 [3], sample type 1B. Each sample set included 7 samples for each formulation and adhesive type. Samples were all cured at room temperature for a minimum of 7 days. Testing speeds 1.0 mm/min (modulus) or 20.0 mm/min (break) for EC2216 and 1.0 mm/min (modulus) or 2.0 mm/min (break) for EC9323-2. Engineering stress and strain were recorded.

2.4. The influence of the operator's workmanship on the results of the single lap shear strength EN1465

Within this experiment the key variable was the factor of bonding operator. Three operators were given the same task: to prepare 5 standard lap shear samples for EN 1465 using the same tools, surface treatment procedure, cleaning and adhesive application, including curing. The substrates were made of aluminium AA6082 (1.5x100x25 mm). The substrates were treated within three consecutive steps:

- Cleaning/degreasing (with isopropanol) to remove contaminants.
- Mechanical (grinding paper Struers FEPA P #80, grain size 82 microns, 40 strokes each direction in 45° angle between longitudinal axis and the grinding direction).
- Another cleaning cycle, ultrasonic cleaning with acetone.

Operators used 3M's ScotchWeld™ 2216 B/A Gray, mixed and applied by spatulas and cured at 65 °C, for 2 hours in the box oven at ambient pressure. Each operator had to prepare his own mix, using analytical balance and instructions from the manufacturer's datasheet. After curing and 72 hours recovery the samples underwent dimension inspection (thickness and overlap) and were submitted to mechanical test according to EN 1465, using a Zwick/Roell tensile machine equipped with a 100 kN load cell with pull rod speed 5 mm/min. The fracture surfaces of the sample with the weakest and strongest joints were analysed in detail with Keyence VHX-2000 digital microscope in order to determine the relation between the lap shear strength and the portion of adhesive/cohesive fracture modes.

3. RESULTS

3.1. Influence of the surface treatment on adhesion strength

The effect of surface treatment on adhesive joint strength is demonstrated in Figure 1. The results show that more careful and complex surface treatment leads to higher adhesion strengths obtained for FeNi36 alloy (INVAR), but also improves the quality of fracture surfaces, shifting from partial adhesive failure (AF) for basic surface *Treatment 1* to fully cohesive failure (CF) for fully treated and primed surface (*Treatment 3*).

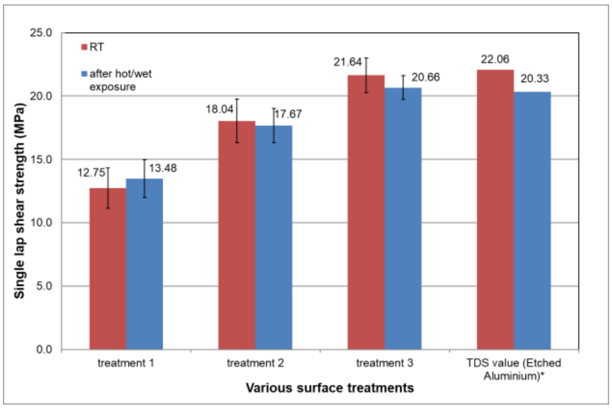

Figure 1: Single lap shear strength for post-cured ScotchWeld 2216 on FeNi36 alloy (INVAR®) before (red bars) and after exposure to "hot-wet" conditions (blue bars) as a function of surface treatment[1]

1 *NOTE: TDS category in Figure 1 is taken from the TDS [4] for Al-Al joint, cured for 2 hours at 66°C, tested in ambient conditions (red) and after hot-wet exposure 14days, 100%RH, 49°C (blue). Used as a reference for shear strength.

Such improved bondline quality results also in excellent resistance to “hot-wet” conditions (nearly equivalent for “ideally treated” aluminium substrates, as per datasheet data for the adhesive [4]).

On the other hand, these results also show that only relatively low adhesion strength can be achieved for the items for which the ideal surface treatment is unachievable (shear strength after *Treatment 1* ~ 60% of strength achieved after the treatment 3).

3.2. Durability of adhesive joints with various surface treatments and curing temperatures

The results of this type of tests clearly demonstrated that the 2-C epoxy adhesive may become very sensitive to “hot-wet” exposure conditions in case there is no further post-cure regime involved before the exposure of the joints (Figure 2). Unlike in the previous study of surface treatment efficiency (Figure 1), the RT-cured 2-C epoxy joints appeared more sensitive to moisture; surface treatment quality seems to improve the apparent shear strength before exposure, but the joints are not as durable as those, which were post-cured at elevated temperatures. Figure 2 shows that *Treatments 1 and 2* resulted in almost the same shear strengths after “hot-wet” exposure. Priming (*Treatment 3*) improved resistance to “hot-wet” exposure, but the obtained strengths were not equivalent to those obtained from samples post-cured at high temperature with the equivalent surface treatment quality (compare the results for *Treatment 3* in Figure 1 and in Figure 2).

Note that the maximum shear strength obtained for the unexposed samples which were not post-cured (Figure 2 red bars for *Treatment 2 & 3*) did not reach as high values as for their post-cured equivalents (Figure 1, red bars for *Treatment 2 & 3*).

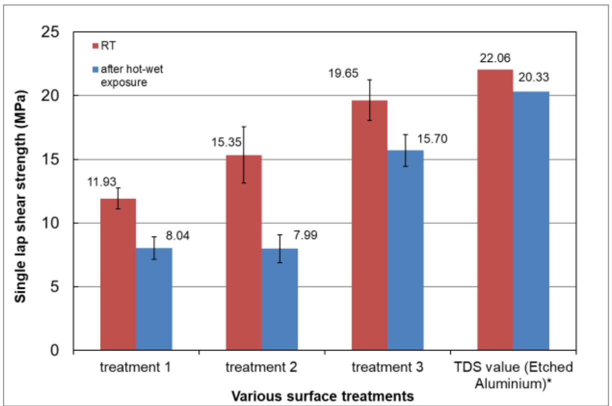

Figure 2: Single lap shear strength of RT-cured ScotchWeld 2216 on FeNi36 alloy (INVAR®) before (red bars) and after “hot-wet” exposure (blue bars) as a function of surface treatment.

A similar susceptibility to adhesion loss for RT-cured INVAR samples was observed after exposure to “hot-wet” conditions also for RT-cured aluminium samples (Figure 3). Summarising findings from all three series of tests for both INVAR as well as aluminium the strength of the joint is extremely dependent on the surface treatment quality. This effect is enhanced after exposure to hot-wet exposure conditions. The durability of the joint increased greatly if RT-cure was followed with post-cure regime at elevated temperature (2 h, 65 °C). Poor adhesion strength may be associated with susceptibility of unterminated polymer fragments in not fully cured adhesive to bond to the hydroxyl groups from ambient moisture instead of hydroxyl groups on the substrate. This effect is probably enhanced by diffusion of moisture along the imperfect bondline areas.

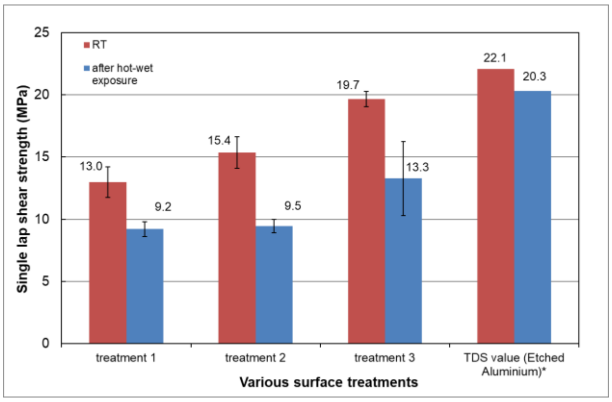

*Figure 3: Single lap shear strength of RT-cured ScotchWeld 2216 on aluminium substrate before (red) and after “hot-wet” exposure (blue) as a function of surface treatment *.*

Further work should be dedicated to these phenomena to develop a model for deterioration of adhesive bondline and models predicting lifetime performance of adhesively bonded joints.

3.3. Preliminary results from formulation change assessment of EC2216 B/A Gray and EC9323-2 B/A black

The results from the tensile tests according to ISO527-2 for EC2216 B/A Gray and EC9323-2 B/A Black (7 days RT-cured) are listed in the following tables with standard deviations in brackets. The impact of the formulation changes verified for the new formulations is more consistent for the “new” version of EC9323-2 (Table 2) than for EC2216 (Table 1). EC9323-2 new formulation displays a slightly negative trend in all measured characteristics (Table 2, modulus and strength) when compared with the old one.

Nevertheless, none of the measured characteristics changed significantly. Absolute changes were for some parameters in the range of standard deviations.

The work on bulk property characterisation for these two adhesives is ongoing to provide the results from thick adherend test, single lap shear tests, fracture energy characterisation and immersion wedge test and to further assess the impacts of formulation changes.

Table 1: The averaged bulk properties of RT-cured EC2216 (average) with "new" and "old" formulation changes.

Adhesive	Modulus (MPa)	Poisson ratio	Max. strength (MPa)	Strength at break (MPa)	Strain at break (%)
EC2216 OLD	118 (6)	0.408 (0.021)	17.2 (0.3)	17.1 (0.4)	41.1 (1.8)
EC2216 NEW	133 (10)	0.420 (0.015)	18.2 (1.1)	17.9 (1.2)	39.8 (4.1)
Relative change	+12.7%	+2.9%	+5.8%	+4.7%	-3.2%

Table 2: The averaged bulk properties of the RT-cured EC9323-2 with "new" and "old" formulations.

Adhesive	Modulus (MPa)	Poisson ratio	Max. strength (MPa)	Strength at break (MPa)	Strain at break (%)
EC9323-2 OLD	1281 (50)	0.390 (0.022)	27.2 (0.7)	27.0 (0.7)	3.01 (0.56)
EC9323-2 NEW	1158 (101)	0.398 (0.016)	24.8 (0.5)	24.8 (0.6)	3.24 (0.43)
Relative change	-9.6%	+2.1%	-8.8%	-8.1%	-7.1%

The performances in thermal vacuum with respect to exposure temperature, surface treatment and pre-load stress could be part of future work. A curing shrinkage and viscosity evolution during the cure are lately also key properties for many applications and further assessment should also consider them.

3.4. Operator's influence on shear strength of the bonded joints

The results from the tests dedicated to assessment of the influence of the operator factor on adhesive bonding shear strength are shown in the Figure 4.

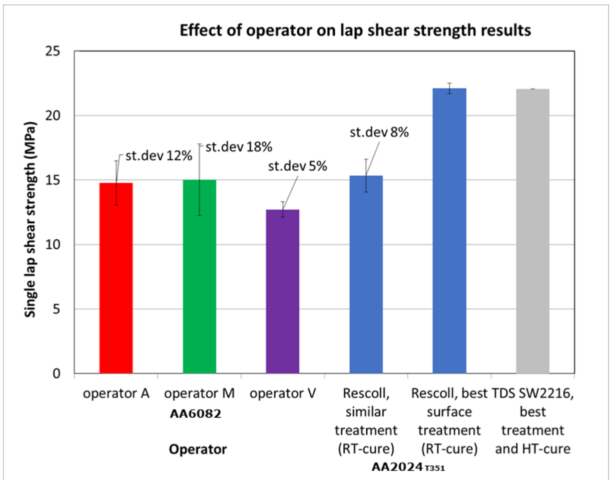

Figure 4: The operator's influence on the adhesive bonding reflected in the adhesive shear strength (first three from the left), compared with reference values from other tests (blue bars) and TDS value (grey bar).

Note that the key characteristic for the selection and acceptance of the bonding operator is not only the achieved average shear strength but also the reproducibility (lowest standard deviation). The operator "V" (Figure 4, purple column) could be considered as better choice for the applications, where standard deviation is affecting further calculation of design allowables.

Comparison of fracture appearances of the strongest and weakest bond is shown in Figure 5.

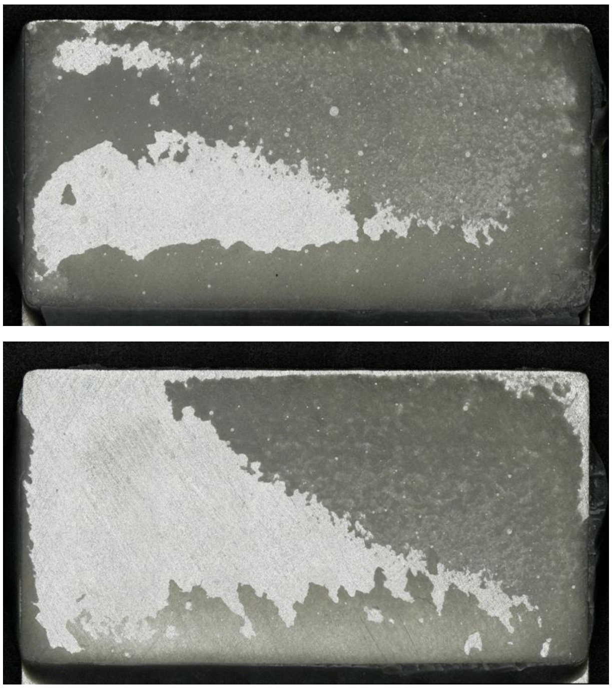

Figure 5: The analysis of fracture surfaces from the samples with the highest (top) and the lowest (bottom) shear strength.

The highest and the lowest single lap shear strength recorded among all three sets of tested samples was 18.8 MPa (with 51.2% CF) and 12.0 MPa (38.9% CF) respectively. This finding suggests that there is a clear relation between portion of cohesive fracture mode and obtained shear strength value.

CONCLUSION

This work summarises the recent developments in certification, standardisation and quality assurance in adhesive bonding technology, focusing on adhesives relevant for space applications.

Experimental work on 2-C epoxy adhesive (ScotchWeld 2216) proved the importance of the surface treatment and the curing temperature, identifying the most efficient processing among the selected ones.

The assessment of the recent formulation changes in ScotchWelds EC2216 Gray and EC9323-2 so far did not reveal any significant impact on the studied properties (tensile strength, elastic modulus, poison ratio and elongation at break), but further testing is needed for the completion of the assessment.

The influence of the operator's workmanship on lap shear strength and reproducibility in obtained results were studied. Both average strength as well as standard deviation are relevant parameters within an assessment of adhesive bonders.

ACKNOWLEDGEMENT

The authors would like to thank to all ESA's and Rescoll's teams involved in adhesive formulation change assessment. Special thanks to Mickaël Lafourcade (Rescoll) for his involvement in "surface treatment efficiency" exercise, and to George Hunt (ESA) for fracture surface analyses.

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