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Relation between acoustic emission analysis during cure cycle and bonded joints performances

C. Santulli*, A.C. Lucia

European Commission, Joint Research Centre, ISIS-ATIA, Ispra, Italy

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Abstract

In 1993, a research program was started and developed in JRC Diagnostic and Reliability laboratories to assess the possibility to predict the structural behaviour of adhesive joints monitoring acoustic emission (AE) during the cure cycle leading to their production. Cure cycles have therefore been performed, either respecting the standards and guidelines established for the adherent materials, adhesive layer, timing, temperatures, or instead modifying one or more of these factors. The latter situation would presumably produce a joint which is unsuitable for use or at least with non-ideal characteristics. The idea was that acoustical activity during the different cycles should be discernible from a statistical point of view, presenting a relation with the quality of the joints finally produced. From experimental data some evaluation criteria were sorted out and a possible outlining of on-line joints' quality control through AE concepts is provided. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Adhesive joints; Cure cycle monitoring; Acoustic emission

1. Introduction

The possibilities offered by NDT techniques in controlling aluminium adhesive joints for aircraft structures are exhaustive for as regards the definition of local mismatches in joint structure as a result of the presence of voids, lack of adhesive or any zone of insufficient adhesion [1]. Here standardised methods, such as Fokker Bond tester, and tools of inspection based on ultrasonics [2] in the form also e.g., of Lamb waves analysis [3] or acousto-ultrasonic [4,5] techniques achieve this purpose with an acceptable accuracy. Some difficulties have besides been pointed out on particular goals e.g., enlightenment of cohesive problems [6]. Some new possibilities to industrialise the NDE on adhesive joints were provided in the late 1980s from the mechanical impedance method [7,8]. In that case advantages seem to be owing to the possible application of that technique to all types of bonded joints, although limitations were observed, when acting on flexible test-pieces.

Adhesive joints weakness owing to changes in temperature and moisture content can be excluded nowadays [9] from the onset of an efficacious control and a sounder knowledge on surface treatments (anodisation + primer application) together with severe guidelines for critical factors in cure cycle (temperature, pressure, adhesive storage and handling). The availability of highly reliable adhesives as modified epoxy resins with cure cycles at 120°C finally assured the development of industrial bonding. The aim was in that case to provide a low cost technology, whose successful use depends nevertheless on the accuracy in each phase of the process leading to adhesion. Such precautions were reflected, beyond to standards and industrial guidelines, in a great deal of literature [10-14].

An adequate quality verification does not imply that the adhesion process is completely lighted by current theories, mostly focused on wetting or mechanical interlocking models. In aircraft industry practice the exigency of high security levels makes in general complex both to evaluate defects' respective criticality and to reduce guideline requirements and hence intrinsic costs of adhesion process. A thorough knowledge of the most diffused alterations originating in joints production has to be attained, before proceeding to a confirmation or partial modification of guidelines, aiming to lead to a lower-cost management of bonding technology.

Here acoustic emission can play a role: from some examples of in-cycle cure [15,16] the adhesion process observation through AE was conceived. First results were the possibility of discernment between the different phases of cure cycle in AE cum. counts curves [17,18] and the emphasis on signal features most influenced from local viscosity variation owing to polymerisation and to shrinkage effects

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* Corresponding author. Fax: + 44 0151 7944675.

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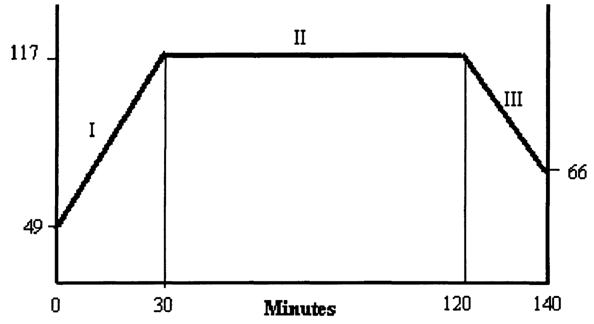

Fig. 1. Polymerisation cycle.

in the joint owing to adhesion [19,20]. A farther step [21,22] brought us to the creation of defects, whose importance for the completion of curing was assessed by digital scanning calorimetry (DSC) technique. Each series of investigation was followed by joints mechanical validation by standard ASTM D1002 and D3163 shear stress tests.

2. Experimental procedures

2.1. Materials

Sheet type rectangular 2024 T3 aluminium alloy samples were used in a single lap configuration (overlapping area 25.4 × 12.7 mm). Normal dimensions of the specimens, suggested by ASTM D1002 standard, were 75 × 25.4 × 1.6 mm. Double length specimens were also tested; the standard length does not fit in effect with the positioning of two AE sensors on the specimen.

Surfaces have been treated prior to bonding (thorough cleaning, anodisation, obtained through phosphoric or chromic acid, and primer application) according to the relevant industrial standards. The adhesive was a modified epoxy (AF163-2K by 3M) with a nominal thickness of 0.19 mm and a density of 0.22 kg/m^2.

2.2. Cure cycle

Standard cycle applied in Italian aircraft industries envisaged:

Pressure 0.25(± 0.035) MPa

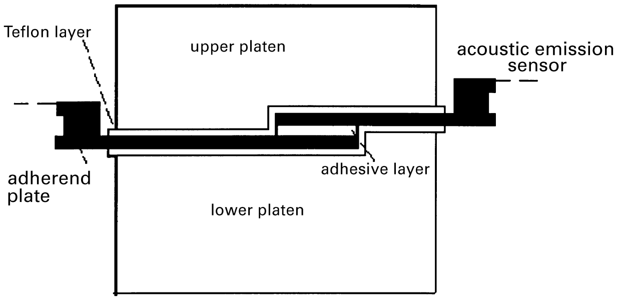

Fig. 2. Schematic test disposition.

Medium heating rate
2°C–3°C/min
Phase 1 Heating from 49°C to 117°C
Phase 2 Holding for 90 min at 117(± 3°C)°C
Phase 3 Natural cooling from 117°C to 66°C

The approximate polymerisation cycle employed is shown in Fig. 1.

The two adherents were wrapped together with ®Teflon, so to avoid the detection of spurious signals owing to electrical connection, and submitted to pressure in a hot press, whose platens temperature was measured by thermocouples. The acoustic emission sensors were applied as schematised in Fig. 2.

In Table 1 all the joints are summarised (each series 5 joints) whose AE behaviour during cure cycle was monitored. A-type joints were produced following the guidelines, although with some minor differences between them; to produce instead B and C joints a modified procedure were employed, leading in B-type *probably* to some quality degradation and in C-type joints *surely* to their inefficacy. Details are given in the legend.

2.2.1. Legend

Specimens:

- Normal: 75 × 25.4 × 1.6 mm 2024T3 aluminium alloy specimens
- Double length: 150 × 25.4 × 1.6 mm 2024T3 aluminium alloy specimens
- AISI316: 130 × 25.4 × 0.8 mm AISI316 steel specimens

Surface treatment:

CAA = Chromic acid anodisation
PAA = Phosphoric acid anodisation

Only a slight pickling was applied on steel adherends

Pressure

- Normal: as prescribed in the guideline i.e., 0.25 MPa
- Double: twice as prescribed in the guideline i.e., 0.5 MPa

Modifications from ideal procedure:

- Faster cooling (6°C/min.)
- Slower cooling (0.2 + 0.4°C/min.)
- 5 × 5 mm central defect
- 5 × 5 mm boundary defect
- Adhesive on half surface: the adhesive film was able only to half-cover the joint surface
- Fully defected area: among the adherend and the adhesive a sheet of Teflon® was inserted
- Expired adhesive: adhesive stored for twice the maximum shelf time was employed

Table 1
Details of all the joints whose AE behaviour was monitored

Series	Specimens	Surface treatment	Pressure	Modifications from ideal procedure	Envisaged result
a1	Double length	PAA	Normal	None	Good
a2	Double length	CAA	Normal	None	Good
a3	Normal	PAA	Double	None	Good
b1	Normal	PAA	Double	Faster cooling (6°C/min.)	Doubtful
b2	Normal	PAA	Double	Slower cooling (0.2 ÷ 0.4°C/min.)	Doubtful
b3	Double length	CAA	Normal	5 × 5 mm central defect	Doubtful
b4	Double length	CAA	Normal	5 × 5 mm boundary defect	Doubtful
b5	Normal	PAA	Double	Adhesive on half surface	Doubtful
c1	Normal	PAA	Double	Fully defected area	Bad
c2	AISI 316	Pickling	Normal	Expired adhesive	Bad
c3	AISI 316	Pickling	Normal	Mold release	Bad
c4	Double length	PAA	Normal	Mold release	Bad
c5	Double length	None	Normal	None	Bad
c6	Double length	CAA	Normal	Holding temperature 90°C	Bad

- Mold release: the two surfaces are covered with dybuthil ether, so to avoid the adhesion
- Holding temperature 90°C

2.3. Acoustic emission

A LOCAN-AT apparatus equipped with two 150 kHz sensors was used, with thresholds exceeding by 10 dB the electronic noise level of the acoustic emission system. Actual values of threshold varied from 28 to 33 dB, depending from the sensor used for the test. However no direct, but only normalised data comparison was envisaged; this because different *signal definition times* were employed. We intend with this locution the intervals used to divide the detected acoustical wave into acoustic emission hits: these are PDT (peak definition time), HDT (hit definition time) and HLT (rearm time), whose significance is widely explained in [23]. These were set at different values during the whole experimentation: PDT varied from 20 to 200 μs, HDT instead from 50 to 500 μs. Only HLT was always preserved as 300 μs, that was the minimum time allowed by AE system to process data and be able to acquire a further signal. Values of PDT and HDT were modified according to the sensors employed and to their calibration, aiming obviously to preserve the S/N ratio, so to obtain a quite equal level of sensitivity for the whole measuring chain. Hence can be deduced that any evaluation of the difference among the signal shapes on the base of AE data so collected has to be excluded. The interest is concentrated instead on the comparison between the different physical situations and their influence on the modifications in acoustical activity.

The same LOCAN apparatus was used in the whole test campaign, even if an up-to-date system would be available too. This because no reliable comparison between quantitative data from different AE systems could so far be obtained, especially for as regards released energy measurement [24]. The use during the whole experimentation of different sensors, although all with a 150 kHz resonance frequency, was not avoided, so to concretely assess the possibility of an evaluation totally independent from the probe employed in AE detection.

2.4. Tensile tests

All joints produced have been finally tested up to

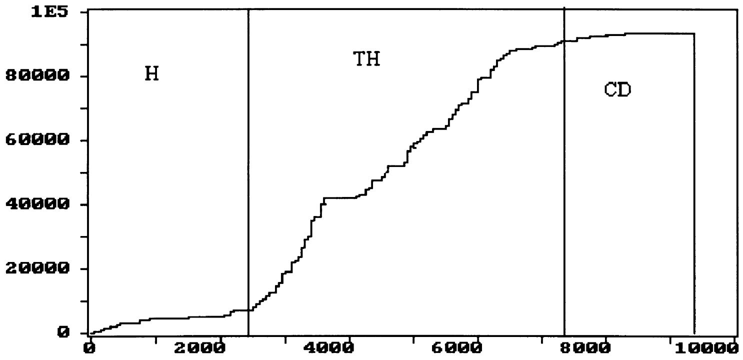

Fig. 3. AE Cumulative counts vs. Time (s) curve for case a1.

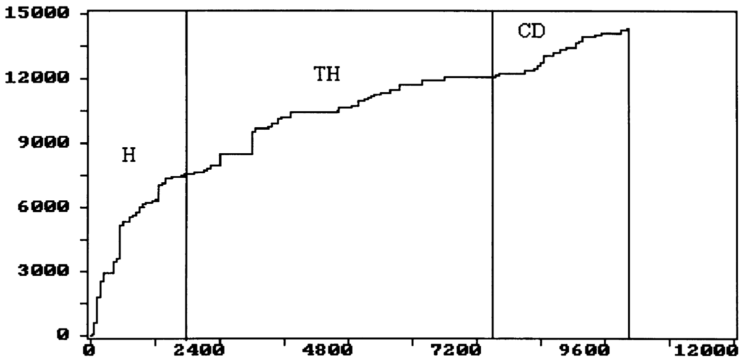

Fig. 4. AE Cumulative counts vs Time (s) curve for case c2.

their ultimate shear stress, following ASTM D1002 and D3163 standards, envisaging a 1.3 mm/min loading rate in displacement control. A thermomechanical ZWICK 1488 Universal Testing Machine was used, equipped with hydraulic grips exerting a 10 MPa pressure.

3. Results

3.1. First considerations

The first peculiarity observed in AE analysis from cure cycle monitoring was that the three natural phases (heating, temperature holding and cooling) could be more or less discernible from cumulative counts vs. time curves. A first hypothesis was then that *the grade of discernment of the phases could be linked to the intrinsic quality of the joint.* In Figs. 3,4 and 5 three different cases are reported, discussed here below; for a better evidence the three phases of the cure cycle are lettered with H (heating), TH (temperature holding) and CD (cooling down). A quite clear difference can be noted between Fig. 3 (joint type al: structure is ideal and phases are very evident in AE curve) and Fig. 4 (joint type c2: the structure is surely defective because the material does not fit, the adhesive is expired and the surface treatment is at least inadequate and phases are no more distinguishable in AE curve). Therefore the above explanation seemed reasonable.

The observation however of cases like that in Fig. 5 (joint type c1: the adhesion does not happen, but the phases are still discernible) raised some doubts, producing the need for a sounder analysis on the relation between this AE curve and the real mechanical behaviour of the joint.

3.2. Qualitative model

To enhance the comprehension of AE cumulative counts curves, a qualitative model was disposed. The main observation was at this regard that acoustic emission during joint production process may be generated by:

1. Movements related with defects present in aluminium and/or in the superficial layer
2. Change of viscosity in the adhesive owing to the polymerisation
3. Air trapping owing to pressure effect
4. Shrinkage owing to adhesion

These phenomena are of course not uniformly present throughout the whole cure cycle. The heating phase is dominated by movements inside adherends or even by pressure effect. Thus, AE activity during this first phase is mainly not linked with the creation of the joint interface. In Fig. 6 the

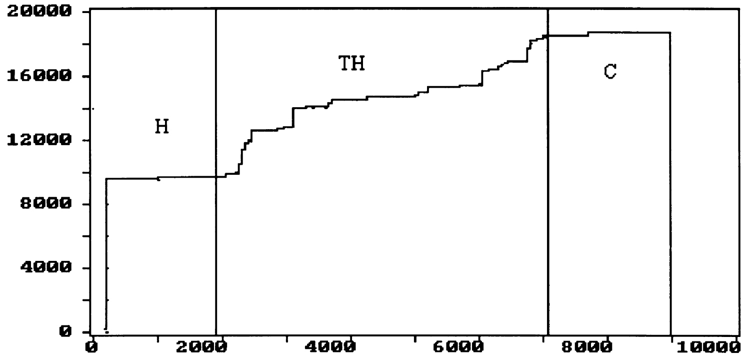

Fig. 5. AE Cumulative counts vs. Time (s) curve for case c1.

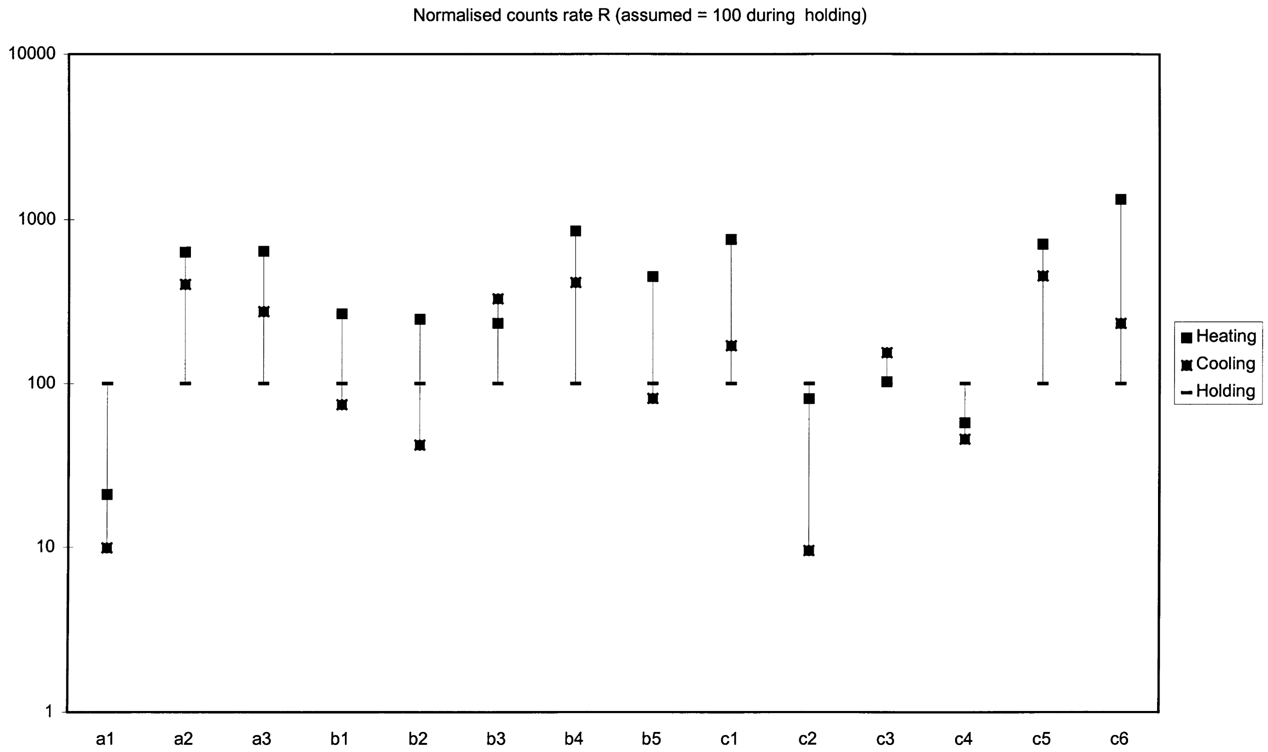

Fig. 6. Logarithmic counts rate for curve cycle phases ($C_t = 100$ for holding).

logarithmic curve of average counts rate C_t on each of the three phases is reported, as obtained by (1):

$$C_t = \frac{\sum_{1}^{5} C_c}{\sum_{1}^{5} t} \qquad (1)$$

where C_c = Cumulative counts t = duration (seconds) of the single phase of the cure cycle The number 5 is referred to the number of joints considered per each series and normalised by imposing C_t equal to 100 in the holding phase.

Table 2
Cure cycle phases in qualitative model

Phases	AE owing to	Approximate phase extension
A	3.(some) 2.(beginning) 1.(traces)	Heating from 49°C to 90°C
B	2.(increased) 1.(more evident)	Heating from 90°C to 117°C
C	2.(again increased) 1.(disappearing)	First half of holding phase (0–45 min.)
D	2.(reduced)	Second half of holding phase (45–90 min.)
E	4.(strong, if any) 1.(traces)	Cooling up to 90°C
F	4.(disappearing) 1.(traces)	Finishing cycle

A first comment on Fig. 6 would concern the fact that two phases out of three are absolutely not discernible for series c3 and discernible with many difficulties for series b5, c2 and c4. Some doubts could even be raised for series b1.

Going deeper into adhesion mechanism, we could assess that the three phases could be divided another time into six shorter phases, here below lettered from A to F and explained in Table 2.

Some concepts could be reported in order to justify this phase partition. Our testing experience has sorted out that standard guidelines for adhesive joints production in aircraft industries consider a time at least double than the real polymerisation time for temperature holding and the same idea could be kept for cooling down, where 90°C are reached at about half of cooling time. For as concerns instead heating, the adhesion effect is considered to be not relevant up to 49°C and begins in a very slight mode, so that we believed that at least 2/3 of total first phase time is not so meaningful for polymerisation.

Thus, phases A and even B, where phenomena extraneous to bonding are most probably dominating, have to be excluded in a quality control philosophy. This also because electronic noise coming from heating apparatus, although limited by the already mentioned precautions, was nevertheless present during these phases.

The remaining phases, from C to F, should present a comparable importance in order to attain a satisfying adhesion. An efficient joint would have reasonably a not very different level of activity in phases C and E and in phases D and F. Furthermore, the values have not to be dispersed. In

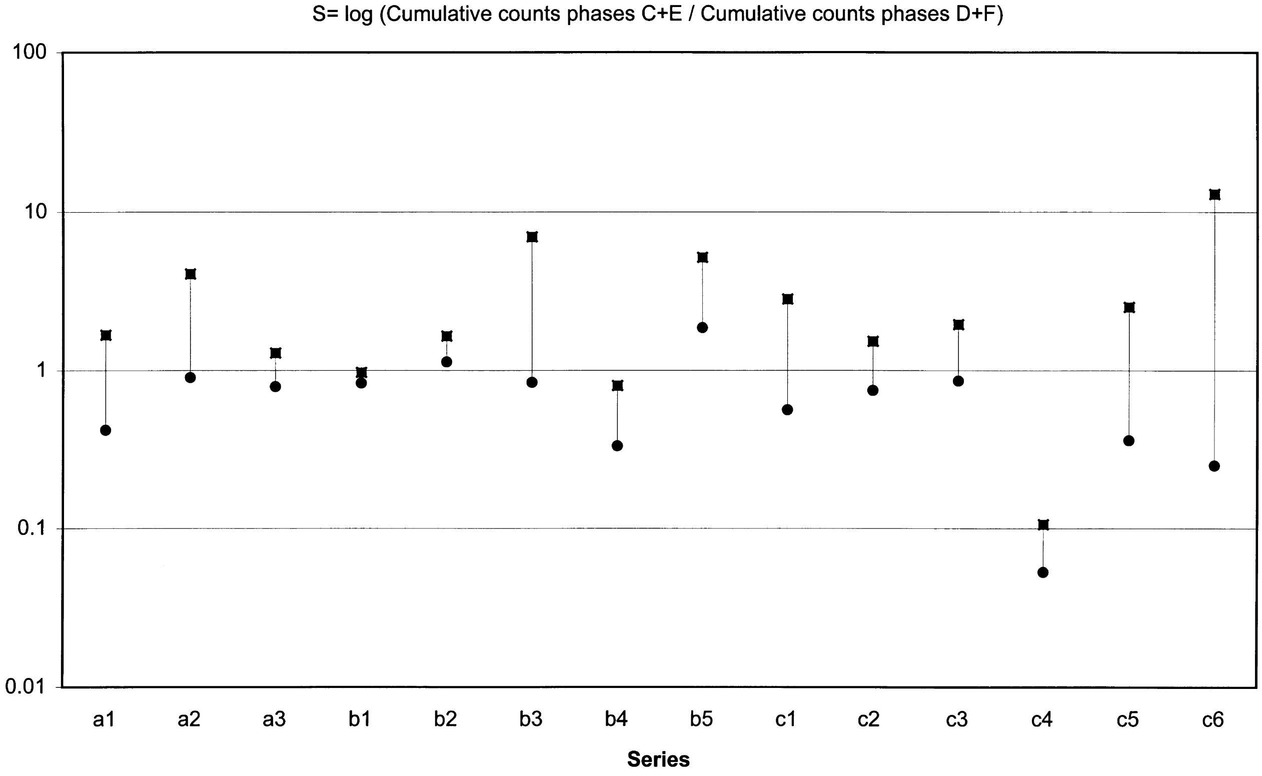

Fig. 7. S ratio values per each series.

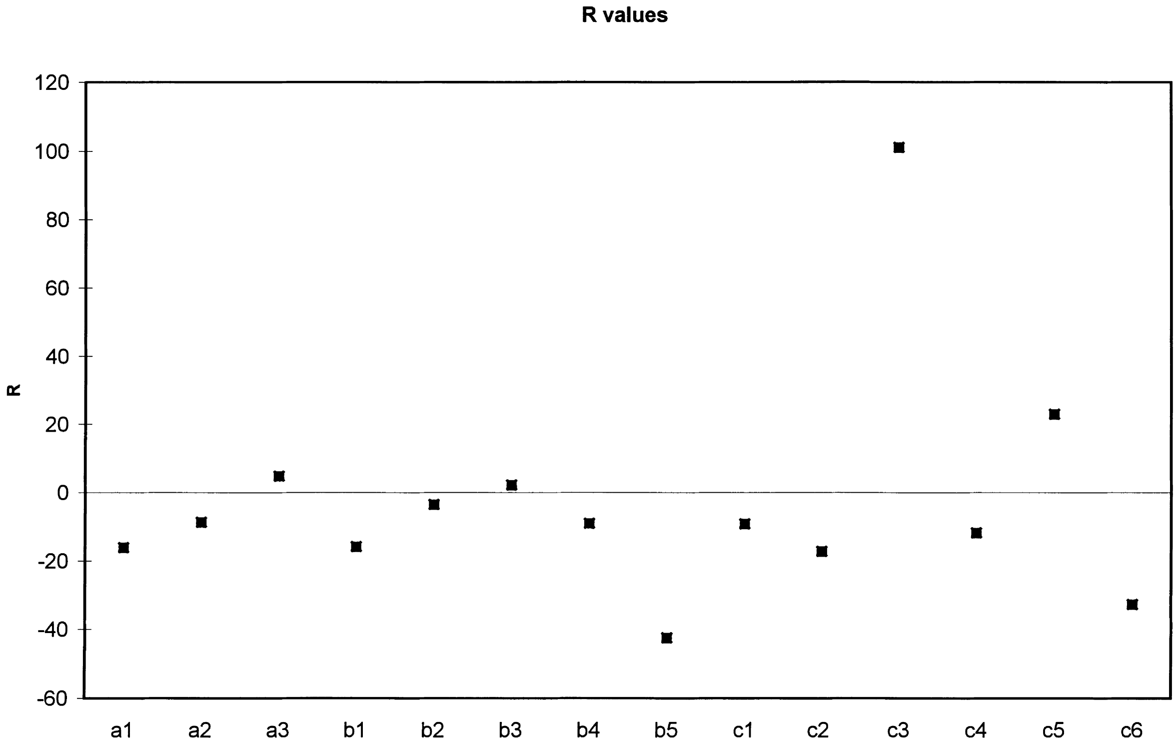

Fig. 8. Cum.counts/Cum.energy ratio: % difference from whole test and cooling only.

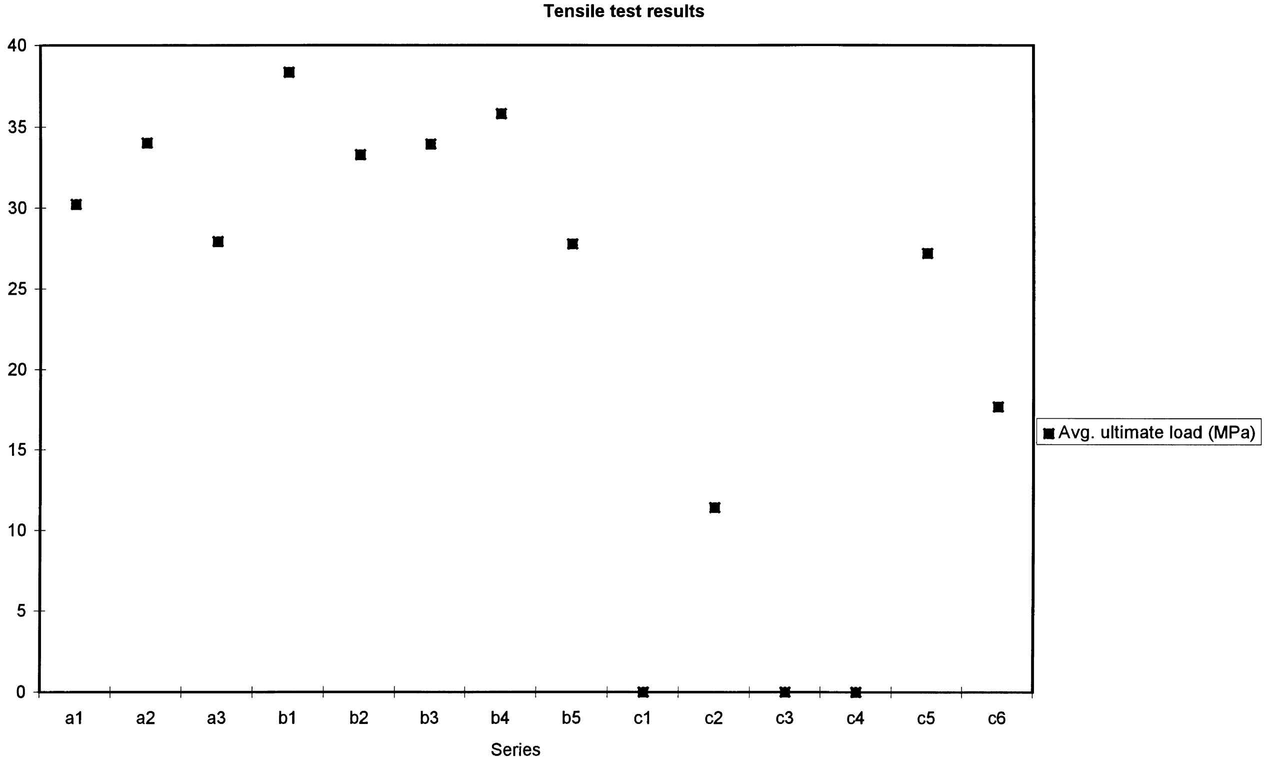

Fig. 9. Tensile tests results.

other words, if one compares the values of the ratio S between the cumulative counts in phases (C + E) and phases (D + F), represented in Fig. 7 (minimum and maximum value for each series), we will observe in what measure this idea is confirmed. Note that time was neglected in that analysis, owing to the fact that timing of phases (C + E) and phases (D + F) is ideally equal.

From that plot one should note that a much greater dispersion is showed by series c6, while in series c4 the importance of phase (C + E) is at least an order of magnitude greater than that of phases (D + F); in series b4 and b5 doubts could moreover be raised, since all values are situated in the same part of the plot, below (b4) or above (b5) the unit. Note that series b4 and series b5 both present adhesive on only a part of the whole joint surface, so that probably less time is needed to complete adhesion.

3.3. C_c/C_e = Cumulative Counts/Cum.Energy ratio behaviour

Observations on C_c/C_e = Cumulative Counts/Cum. Energy ratio (reported in [18]) showing a general decreasing trend for defective joints when considering C_c/C_e on the whole test and on cooling phase only are here better clarified. Phenomena related in an ideal cure cycle with in-service performances of the joint are change of viscosity in adhesive layer as a result of its polymerisation (above indicated as 2.) and adhesion induced shrinkage (above indicated as 4.). Such phenomena should be predominant during an ideal cure cycle.

A defective joint on the contrary is a joint in which either polymerisation does not take place (hence 2. is hardly observed) or adhesion is not sufficient to assure its reliability (hence the detection of 4. should be comparably poor). Air trapping and outgassing as a result of pressure application (above numbered as 3.) should instead have a relevant importance, *only during heating*: the sensitivity of acoustic emission to this aspect of cure cycle was already reported in [16]. To restrict therefore our analysis from the whole test to the cooling period only, considering that heating phase appears to be marginal for the final properties of the joint, would show the relative importance of phenomenon 3., whose excessive weight would most probably indicate that the joint is defective.

Hence we propose that Cum.counts/Cum.energy should present only a little variation whether considering it during the whole test or during the cooling phase only. The presence of extraneous events would be in fact immediately discovered through an unpredictable and broad change of that value. Fig. 8 depicts the situation for C_c/C_e ratio variation when the windowing is restricted from the whole test to cooling phase only. This variation is obtained through (2):

$$R = 100*\left[\left(\frac{\sum_{1}^{5} C_c}{\sum_{1}^{5} C_e}\right)_{WT} - \left(\frac{\sum_{1}^{5} C_c}{\sum_{1}^{5} C_e}\right)_{CO}\right] \Big/ \left(\frac{\sum_{1}^{5} C_c}{\sum_{1}^{5} C_e}\right)_{CO} \qquad (2)$$

Table 3
Series with deviations from normal behaviour

Criteria	Joint series showing deviations
I (Fig. 6)	c3, c2, c4, c5
II (Fig. 7)	c6, c4, b5, b4
III (Fig. 8)	c3, b5, b6, c5
IV (tensile test: Fig. 9)	c1, c3, c4, c2, c6

where

C_c = Cumulative counts
C_e = Cumulative energy
WT = whole test
CO = cooling phase only

Great variations of R value are showed by series c3 and also by series b5 and c6 and even the series c5 could be considered doubtful.

Final verification would be provided from tensile tests data (Fig. 9). Here joints showing such a unsatisfactory adhesion, that the compression into tensile machine grips is sufficient for their detachment, or even no adhesion at all, are reported with a 0 MPa ultimate stress. A considerable reduction of shear stress value was found, as largely expected for all joints series c# but joints c5, where the absence of surface treatment did not apparently generate a great loss in adhesion quality.

4. Discussion

Three experimental criteria were used to lead from acoustic emission data to a quality discernment; these criteria regarded in particular the possibility to recognise physical cure cycle phases into AE results and the presence of a real equilibrium among those phases without the presence of unexpected acoustic emission bursts; that equilibrium was expressed both from the consideration of a qualitative six-phases model and from the observation of variations in cumulative AE signal characteristics focusing our attention from the whole test to the bare cooling down period. The choice of such an analysis was lead first from the need to exclude through a widely used normalisation of every assumption whose significance would be linked to a particular AE set-up, and then from a consideration of physical phenomena involved into adhesion. The respect of such criteria, together with tensile tests data, would probably lead to an efficient quality discrimination. Table 3 completes our observations, where the series showing deviations from normal behaviour are listed. Greater deviations come first.

Results from tensile tests are confirmed by the global consideration of the three experimental evaluation criteria, with the exception of joint c1. An explanation for the lack of reliability of the model in this case could be that this series presented a complete polymerisation process, although, having covered one side of the adhesive layer with Teflon®, no adhesion could take place. The model raises further doubts about joints c5, b4 and b5, probably in the first case as a result of the formation of a less efficient interface (no surface treatment), the faster completion of polymerisation process allowed by the reduced joint area. Such phenomena could probably reduce the long term performances of the joint. No problems instead show up from joint b3, where the central position of the defect could decrease its significance. Series b1 and b2 seem again not seriously affected by changes in production procedures. This result is not completely unexpected: a slower cooling rate (b1) would not reduce quality, even if it is not convenient in an industrial philosophy, and in case of series b2 the faster cooling was not sufficient to produce any serious damage to the joint; a real thermal shocking was not obtainable with our experimental procedure.

5. Conclusions

Acoustic emission was in most cases able to recognise bonding quality through a wide consideration of real-time behaviour of joint surface during joint production itself. Some situations have to be explained better by a thorough investigation of the relative weight of security coefficient in that industrial practice. The significance however of that study lies in the possibility to apply statistical considerations into acoustic emission data analysis during cure cycle, after the assessment of their relation with physical phenomena. A further goal should be to apply this experimental philosophy into a low-cost and reliable monitoring of a real industrial procedure.

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