Composites welding: characterisation and prediction of adhesion kinetics

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The TACOMA device was developed to predict the quality of thermoplastic composite adhesion, considering the influence of various composites material and process parameters. It provides the aerospace industry with the key data to ensure high-quality welding.

Thermoplastic composite welding is based on melting the matrix and consists in joining two composite parts without adding any external material. It is a viable alternative to traditional rivet assembly, particularly in the aeronautics and aerospace industries, where it can be used to reduce aircraft weight.
While it is a promising technique, it presents scientific and technological challenges for composites due to the mere presence of fibres: surface roughness, residual stresses, low matrix volume, thermal history (inducing matrix ageing), short processing time, etc. In addition, as continuous welding processes are adjusted to become faster and faster (e.g., tape placement), the adhesion step is expected to be as short as possible (between 0.1-10 sec).

Therefore, how can strong adhesion between substrates be achieved, when thermoplastic composite welding processes accelerate?

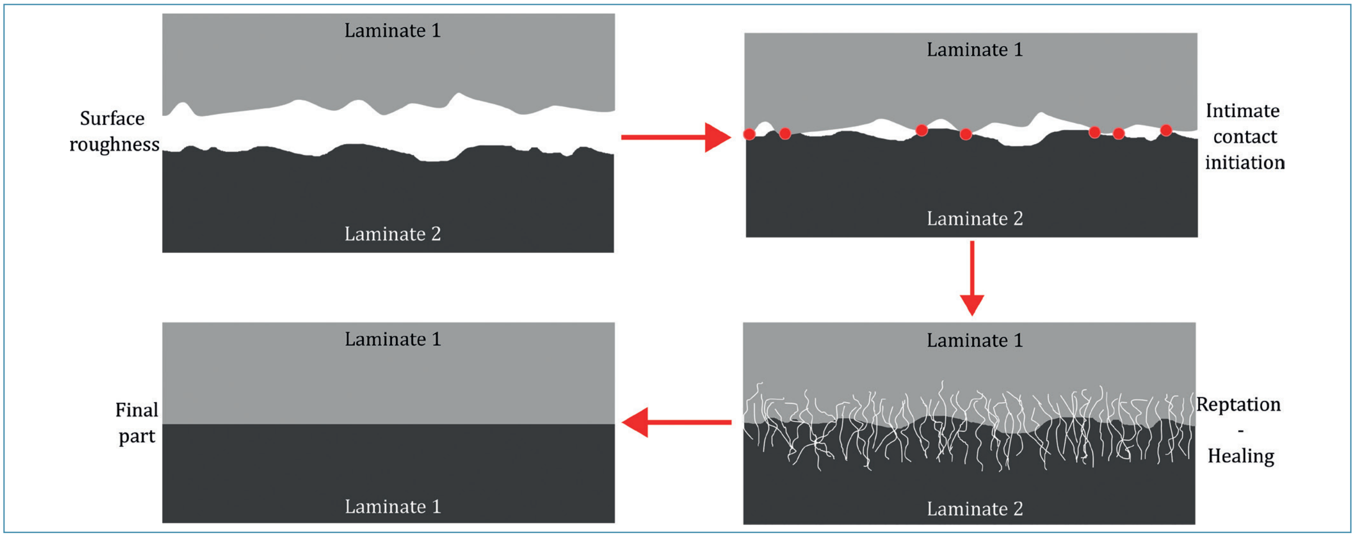

Fig. 1: Schematic description of the different phenomena involved in thermoplastic welding processes

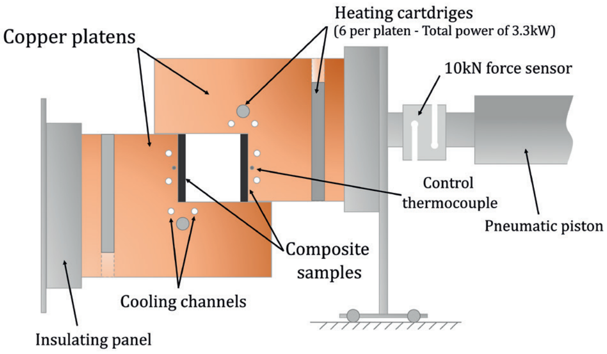

Fig. 2: Schematic view of the TACOMA welding bench

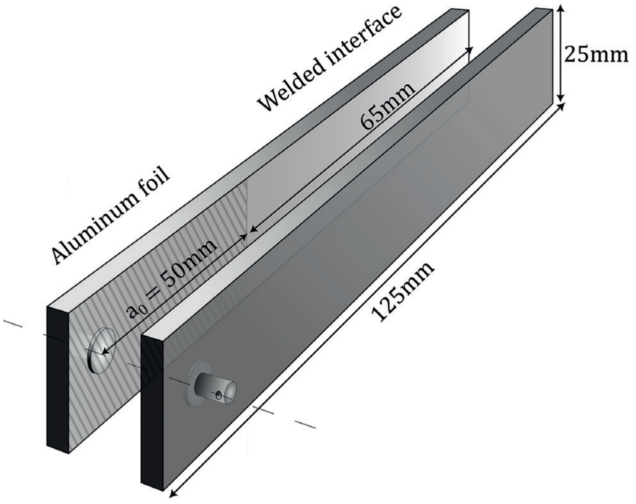

Fig. 3: Schematic view of the DCB samples and their dimensions

To meet this challenge, we've developed a unique device, the TACOMA (Thermoadhesion by conductive heating of composite materials) apparatus, in order to predict the quality of adhesion and ensure that the final composite assembly is strong.

Adhesion mechanisms

Classically, adhesion occurs as soon as the substrates are heated up above the matrix melting temperature and pressed together. Bonding quality results from the convolution of two main phenomena (Figure 1):
(i) intimate contact establishment at the interface driven by the applied pressure, viscosity and geometric roughness of the substrates; (ii) healing of the interface controlled by the diffusion of polymer macromolecules and modelled by De Gennes' reptation theory [1].

Mechanically, adhesion is quantified by using a non-dimensional parameter (D_h) called the degree of healing.

$$D_h = \left(\frac{G_{IC}}{G_{IC,\infty}}\right)^{1/2} \cong \left(\frac{t}{t_{w,\infty}}\right)^{1/4} \qquad (1)$$

$G_{IC,\infty}$ is the strain release energy of the bulk and $t_{w,\infty}$ is the welding time for which mechanical properties of the bulk are reached.
Characterisation of adhesion kinetics between thermoplastic composite parts over a wide range of parameters is a prerequisite for quality prediction of manufactured end- parts. Still, it remains a challenge as the adhesion must be characterised for times representative of those forming processes.

Identification of adhesion kinetics – Experimental protocol

TACOMA apparatus, coupled with mechanical solicitation of the welded interfaces, was developed to address this issue. The welding bench was developed by the LTEN [2] and with the funding of the IRT Jules Verne PERFORM programme. It was originally designed for welding composite parts for a finite amount of time under constant isothermal conditions.

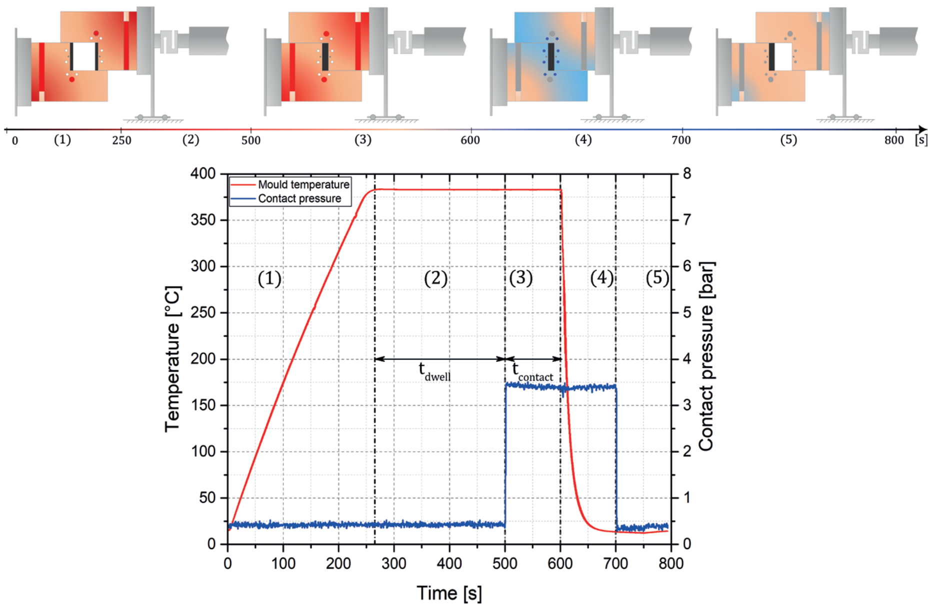

Fig.4: Experimental welding procedure used for healing kinetics identification

Welding tests can be performed for times as short as 1 second under finely-controlled process parameters (time – temperature – pressure). In addition to identifying adhesion kinetics, it makes it possible to quantify the influence of the material (nature of the matrix, surface enrichment, crystallisation, thermal history of the coupons, relative fibre orientation, etc.) and process (time, temperature, pressure) parameters.

TACOMA working principle and performance

The TACOMA bench (Figure 2) is composed of a copper mould with two L-shaped symmetrical platens, one fixed and the other mobile. Samples to be welded (Figure 3) are inserted between the platens (length =125 mm; width = 25 mm) to be heated and pressed together. It was designed so that it would control key process parameters perfectly and independently while welding high performance composites:

- ***Welding temperature***: Electrical cartridges inserted in each platen ensure heating, while two control thermocouples embedded near the cavity surface regulate temperature. Heat transfer has been modelled to guarantee a very precise and homogeneous temperature at the welded interface. Maximum welding temperature is 400°C and can be reached at a rate of 80K/min.
- ***Contact pressure:*** Once the targeted temperature is reached at the interface, pressure is applied with the moving platen. The applied pressure ranges from 1 bar to 8 bars (max.).
- ***Contact time:*** Contact time can be adjusted on demand and can be as short as 1 second before the sample cools. The wide range makes it possible to study both intimate contact development and healing of the interface.

Afterwards, the newly welded sample is cooled rapidly to stop adhesion development. The internal mould's cooling channels circulate water or air, so that available cooling rates range respectively from 720 $K.min^{-1}$ to 15 $K.min^{-1}$. By quenching, or slowly cooling the interface depending on the crystallisation kinetics of the materials studied, coupling between adhesion and crystallisation can be highlighted.

The traditional welding procedure used for kinetics identification is shown in Figure 4. After preparation and drying, the samples are positioned against each platen and the surfaces to be welded face each other, separated by a small air gap. The mould is heated up to a temperature higher than the melting temperature (1) until homogenisation occurs (2). Pressure is then applied for a finite amount of time defined as contact time $t_{contact}$ (3) and the process is quickly stopped as a result of cooling with water (4). Such a cooling rate limits the development of adhesion during this stage. Therefore, the measured adhesion degree after mechanical testing is solely due to the healing that occurs during the isothermal stage.

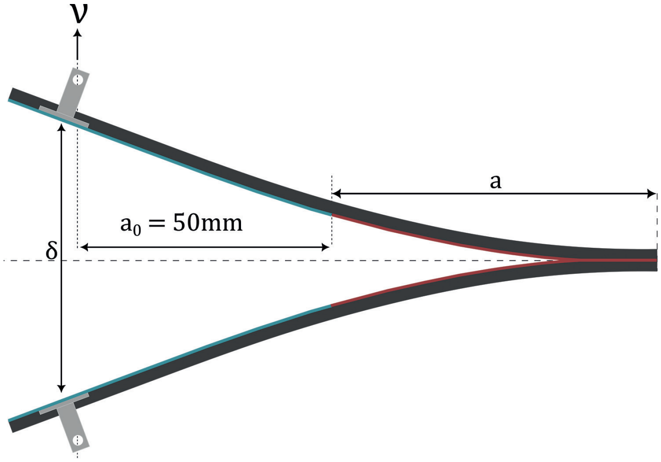

Fig.5: Mechanical testing set-up for the double cantilever beam fracture test

Mechanical testing of the welded interfaces

The interface mechanical resistance of the welded specimens is classically characterizsed using a mode I (opening) test, which is the double cantilever beam (DCB) fracture test (Figure 5). An aluminium foil is then placed between the samples prior to welding to initiate the crack, then and the strain release energy at the crack initiation point ($G_{IC,init}$) is calculated during the quasi-static displacement-imposed test.

$$G_{IC,init} = \frac{3P\delta}{2a_0 b} \qquad (2)$$

P and δ are respectivelyis the recorded force and displacement. a_0 is the initial crack length and b the width of the weld.

It should be noted that other types of mechanical characterisation can be implemented according to the needs of the customer and the type of stresses that the assembly will encounter in service. For example, single lap shear (SLS) testing has already been used successfully by our team at upon request of an aeronautics industrial leader.

High performance thermoplastic composites: healing kinetics

Figure 6 shows the evolution of the strain release energy resulting from DCB testing of a welded high performance CF-PEKK composite under three isothermal temperatures and over a wide range of contact times according to the experimental protocol presented above (Figure 4).

Several bonding regimes can be highlighted :

- ***Coupled regime***: For the shortest contact time, bonding energies are very low. This corresponds to a coupled regime where intimate contact is not fully established. Contact is probably initiated at some points of the interface where healing has already started but has not finished.
- ***Pure healing regime (autohesion)***: When intimate contact is fully established on the whole interface, G_{IC} presents a linear evolution with contact time. This is known as a pure healing regime (called autohesion) where macromolecular inter-diffusion occurs, resulting construction of mechanical resistance of the interface. Full adhesion is reached at the end of this regime when $G_{IC,\infty}$ is reached (Eq.(1)).
- For longer contact times, adhesion is characterised by a competition between matrix cohesive bonding and other phenomena (fibre bridging, segregation of polymer matrix). It results in higher values of G_{IC}, not purely associated with interface healing.

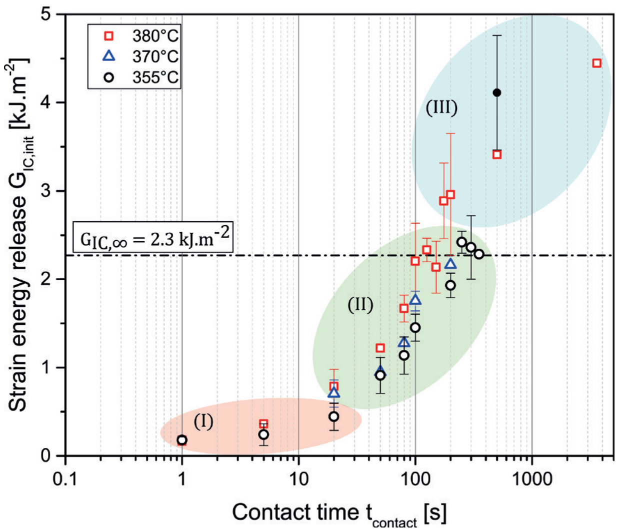

Fig. 6: Evolution of interlaminar fracture toughness (G_{IC}) with contact time at three isothermal temperatures

These experimental welding tests allow the healing kinetics to be determined by identifying the so-called "welding time" $t_{w,\infty}$ for which healing is complete at the interface. Such an analysis requires the healing degree D_h to be plotted over the fourth root of contact time as defined by the reptation theory (Eq.(1)).

For each isothermal temperature, a linear regression is applied to the portion of D_h corresponding to the regime (II) since only the healing regime is of interest. Regarding Eq.(1), the welding times $t_{w,\infty}$ are determined from the slope of this linear regression (Table 1).

Series of tests like these have already been performed by the Capacités team on other high performance composites for aeronautics industry leaders. They made it possible to identify the thermo-dependence of the welding time, which can then be implemented in various modelling tools to predict the evolution of adhesion between parts of any composite manufacturing processes and any anisothermal temperature cycle:

$$\frac{d\left(D_h^{\,4}\right)}{dt} = \frac{1}{t_{w,\infty(T)}} \qquad (3)$$

Towards the optimisation of welding processes

The previous section showed that time and temperature are key parameters for increasing adhesion bonding at the interface. Other parameters can also impact the process and interface performance, such as contact pressure during the process and crystallisation of the specimens. Series of tests shave already been carried out by the Capacités team on similar high performance thermoplastic composites using the TACOMA welding bench. In particular, the influence of the applied pressure and the so-called material parameters on the final quality of substrate adhesion has been quantified.

Tab.1: Identified welding times of a CF-PEKK high performance composite

Temperature [°C]	355	370	380
Welding time $t_{w,\infty}$ [s]	121	104	79

- ***Crystallisation and adhesion coupling:*** Extensive testing was carried out to study the influence of cooling rate (i.e., crystallinity rate). Preliminary work has been initiated with the LTEN on the welding under supercooling conditions (welding temperatures are lower than the melting temperature) which lead to strong coupling between adhesion and crystallisation.
- ***Welding pressure:*** By varying the pressure applied to the substrates during the welding process at iso-temperature and different contact times, one can assess its effect on bonding quality. Since intimate contact being proportional to the ratio between apparent viscosity and applied pressure, such studies can provide a good estimate of the characteristic time for intimate contact establishment t_{ic} [3]. Given that intimate contact is fully established at the beginning of regime (II) (Figure 6), the applied pressure also impacts the time for full adhesion completion.
- ***Fibrous architecture of the composite:*** The influence of the stacking and the relative orientation (angle) of the composite plies at the interface was successfully characterised by our team. It was also possible to study the addition of a pure matrix enrichment layer at the interface and the influence of its thickness.
- ***Heterogeneous welding***: The device offers the possibility to heat the two platens independently, i.e., to carry out the tests with different temperatures for each of the platens. Heterogeneous welding tests can then be carried out using two different materials with different characteristic temperatures.

Concluding remarks

The characterisation of adhesion under conditions similar to industrial processes is crucial. The TACOMA welding device has been developed at LTEN and is promoted by our team of research engineers. It makes it possible to anticipate how material and process parameters impact the quality of adhesion prior to large-scale production. Determining optimal parameters for complete adhesion at the interfaces, through experimentation, allows a process window to be defined.

Several studies have already been successfully conducted for clients in the aeronautics industry, for which the effect of most of the above-mentioned parameters were investigated thoroughly. ☐

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