CURING MONITORING OF ADHESIVE BONDINGS WITH THE HELP OF AIR-COUPLED ULTRASOUND

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ABSTRACT

Curing processes of adhesives depend on application conditions such as temperature or humidity. Most of the time, manufacturer information like pot life and processing time are not directly transferrable to the actual application, so that large safety factors in the curing time are taken into account. They tie up resources such as production areas and delay subsequent process steps, which is accompanied by a significantly reduced added value. There are only a few non-destructive test methods that can be used to monitor the hardening processes of adhesives. Often laboratory methods are used which are not process-capable, have low penetration depths, high system costs and can only be applied to the adhesive itself and not to adhesive bonded component systems. As already shown in literature, air-coupled ultrasound overcomes these limitations. A corresponding test setup and an evaluation method for the determination of ultrasound parameters that allow conclusions to be drawn about the degree of curing are presented. There, the influence of a varying bonded sheet thickness and thus different wave modes, on the quality of the curing monitoring is described. The referencing is carried out by differential scanning calorimetry (DSC) investigations. The procedure is presented and discussed by means of adhesively bonded overlap joints.

KEYWORDS: air-coupled ultrasound, guided waves, wave modes, adhesive bondings, curing monitoring, viscosity

1. INTRODUCTION

In many industrial sectors, the use of lightweight components and composite materials is increasing. Adhesives are often used due to their economical application and to avoid stress concentrations [1]. However, adhesive bondings also require a non-destructive testing method both for assessing the final quality and for monitoring the actual adhesive curing depending on varying process conditions [2, 3]. The methods available so far are limited to laboratory measurements with the help of DSC [4], DMA [5, 6], NMR [7], terahertz [8, 9] or rheological investigations [6]. The following measurement approach using non-contact guided ultrasound waves, on the other hand, allows significantly greater penetration depths as well as applicability to real component structures and not only to the adhesive itself.

2. EXPERIMENT AND RESULTS

2.1 SIMULATIONS

In the first step simulations with help of the program GUIGUW were done to determine the possible order of symmetrical S and asymmetrical A wave modes, depending on the material parameters and the thickness of the investigated sheets. The dispersion diagrams and the calculated phase velocities for the different wave modes as a result of the simulations can be seen exemplarily for sheets of polymethylmethacrylate (PMMA) and thicknesses of 2 mm, 6 mm and 12 mm in figure 1 and table 1. The therefore needed longitudinal and transversal ultrasound velocity in PMMA was determined empirical at a frequency of 200 kHz, the velocity in air was assumed with 340 m/s and the density of PMMA was 1.18 g/cm³ according to the material parameters supplied by the material producer.
Fig 1 Dispersion diagrams for sheets of PMMA with thicknesses of 2 mm (top), 6 mm (center) and 12 mm (bottom). The phase velocity $c_{ph}$ is plotted against the ultrasound frequency. The used ultrasound frequency of 218 kHz is marked with the red line.

<table>
<thead>
<tr>
<th>Thickness (mm)</th>
<th>$A_0$ mode</th>
<th>S$_0$ mode</th>
<th>$A_1$ mode</th>
<th>S$_1$ mode</th>
<th>$A_2$ mode</th>
<th>S$_2$ mode</th>
</tr>
</thead>
<tbody>
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<td>1031</td>
<td>2364</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>6</td>
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<td>1483</td>
<td>2704</td>
<td>3350</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>12</td>
<td>1291</td>
<td>1305</td>
<td>1596</td>
<td>2225</td>
<td>2824</td>
<td>3044</td>
</tr>
</tbody>
</table>

Table 1 Overview of the theoretically existing modes and corresponding phase velocities of guided waves for PMMA sheets with thicknesses of 2 mm, 6 mm and 12 mm.

In the second step the optimal incidence angle $\beta$ to provide the best coupling condition for the ultrasound emitter and receiver was determined in dependence of the preferably propagated wave modes with the help of the corresponding phase velocity in air $v_{air}$ and in the component material $v_{material}$:

$$\beta = \arcsin \left( \frac{v_{air}}{v_{material}} \right)$$  \hspace{1cm} (1)
2.2 AIR-COUPLED ULTRASOUND MEASUREMENTS

To monitor the curing process of adhesives, the device Sonoinspect, Forschungszentrum-Ultraschall gGmbH, with transducers SONOSCAN CF 200, SONOTECH Ultraschallsensik GmbH, was used (see figure 2). The bonding to be monitored was excited with six square-wave bursts each with a center frequency of 218 kHz, a duty cycle of 50 % and a transmission voltage of 200 V. The data acquisition of the 70 dB amplified received signal was carried out with a sampling rate of 3.9 MHz. At a time interval of 30 s, an A-image averaged over 128 pulses with a pulse repetition rate of 50 ms was recorded in each case.

Complementary to the simulations, the mode-dependent phase velocities were determined experimentally. For this purpose, the distance between transmitter and receiver was varied continuously, the captured A-images were converted into corresponding B-images [10] and plotted over the transmitter-receiver distance (see figure 3).

The adhesive Technocill 9409-1, Rudner Klebtechnik GMBH, was examined in the form of overlap adhesive bondings of sheets with different thicknesses. The recorded measurement data were further processed with an evaluation software developed at SKZ. The time depended, relative attenuation \( \alpha(t) \) was determined by taking the maximum amplitude \( A_{max} \) and the current one \( A(t) \) of the corresponding wave mode into account:

\[
\alpha(t) = 1 - \frac{A(t)}{A_{max}}
\]
The signal intensity was not considered since different pulse trains of different wave modes might overlap in time domain due to the similar phase velocities (see table 1). Figure 4 shows the results of the curing process of bonded plates with thicknesses of 2 mm, 6 mm and 12 mm.

![Graph showing relative attenuation over time for different PMMA thicknesses](image)

**Fig 4** Behaviour of relative attenuation during the curing of Technicoll® 9409-1 adhesive, located between PMMA sheets of different thicknesses in an overlap bond. The manufacturer's information of “pot life” (red line) and “handling time” (red dashed line) are marked.

### 2.3 REFERENCING WITH DSC

For referencing the ultrasound results, additional DSC measurements were performed using the device 204 F1 Phoenix, Netzsch Gerätebau GmbH (see Fig. 5). The sample volume was 10 mg +/-0.01 mg. To measure the total enthalpy and thus the degree of hardening, two heating curves were run from -20 °C to 190 °C with a subsequent isothermal waiting time of 5 minutes. The
reheating served to check a complete reaction within the first temperature ramp. Both processes were carried out at a heating rate of 20 K/min.

3. INTERPRETATION

The dispersion diagrams show an increasing number of possible wave modes at the evaluated ultrasound frequency of 208 kHz with an increase of thickness of the PMMA sheets. This results in an increasing overlap of different wave modes in the measured A-Scans. However, due to the different phase velocities of the existing wave modes (see table 1) their separation in time domain was partially possible. Furthermore due to the different phase velocities the optimal incidence angle must be adapted in accordance with the preferably excited wave mode (see table 1 and equation 1). Here, the $A_0$ mode showed the best correlation with the curing status, determined with the help of DSC as well as physical models which describe the interaction of ultrasound waves with the chemical process during the curing [6,7].

Furthermore, in fig 3 it can be seen, that the signal-to-noise-ratio (snr) is decreasing with an increasing thickness of the sheets. This corresponds to physical models as well. The higher the number of propagating wave modes, the lower the snr of a single wave mode, in this case $A_0$, respectively. However, also in case of bonded 12 mm sheets the snr is sufficient to monitor changes in the curing process. While the manufacturer determined the “pot life” time at 15 minutes and the “handling time” at 40 minutes, the DSC measurements show slightly different times (see fig. 5). Here, the first turning point is at about 20 minutes and the second one at about 45 minutes. The reason for this difference might lie in a different temperature during the curing process. The results of the air-coupled ultrasound measurements (see figure 4) shows an analogous behaviour. The minimum attenuation is at about 20 minutes, while the turning point is between 50 and 60 minutes, depending on the thickness of the sheets and thus also the snr. From a practical point of view the amplitude of $A_0$ mode offers the possibility to determine the current curing status but the behaviour shown in figure 4 does not corresponds to the theoretical models described in literature [6,7]. There, the attenuation is increasing during the curing process due to an increase of viscoelastic material properties and finally decrease continuously. That can be justified due to the temporally changing state of matter from viscous to solid. At the end of the further processing time, no significant change in the signal attenuation can be observed, which is accompanied by the progressive cross-linking and the manifestation of mechanical end properties. To verify the air-coupled ultrasound results, rheological investigation will be done perspectively. While DSC shows mainly differences in crystallinity, rheological investigations show differences in viscosity which correlates directly to interactions of ultrasound waves with material.

4. CONCLUSION

In summary, it was shown that guided waves and especially the amplitude of $A_0$ mode is basically suitable for monitoring the curing progress of an adhesive overlap bonding and can be verified with DSC measurements. Furthermore difficulties in signal processing appear with an increasing thickness of the bonded sheets due to the higher number of propagating wave modes. However, it was demonstrated, that even in case of 12 mm thick bonded PMMA sheets the curing process can be monitored with a sufficient snr. Thus, air-coupled ultrasound offers the possibility of non-contact curing monitoring, which...
is not limited to the adhesive itself, but can be applied on bonded components for the first time. In addition to extended signal processing with algorithms to be developed, further measurements using DMA, NMR, and rheological investigations are planned in order to verify the measurement approach presented here.

REFERENCES