INFLUENCE OF THE MANUFACTURING PARAMETERS ON MECHANICAL PROPERTIES OF POLYURETHANE BASED COMPOSITES

S. Gloggnitzer^{a*}, T. Buchsteiner^a, G. Pilz^a

^aInstitute of Material Science and Testing of Plastics, Department of Polymer Engineering and Science, University of Leoben, Otto-Glöckel Straße 2, 8700 Leoben, Austria. *stefan.gloggnitzer@unileoben.ac.at

Keywords: polyurethane resin, glass laminates, mechanical properties, fatigue.

Abstract

Recently, composites based polyurethane matrix resins are of growing interest because of the possibilities for reducing curing times and manufacturing cycles. In the present work, 8 unidirectional glass fiber reinforced laminates based on polyurethane resin system were manufactured along with a systematic variation of the curing parameters time, temperature profile and pressure profile. It is demonstrated that elevated temperature and pressure levels in the manufacturing process lead to a more compact laminate structure with a reduced void content. Regarding the structure-property relation this means primarily an increase of the short term properties such as elastic modulus and flexural strength as well as the low cycle fatigue strength, whereas an increasing void content leads to an improved high cycle fatigue performance.

1. Introduction

Fiber reinforced plastics are becoming more important in lightweight constructions due to their high mechanical properties combined with low specific weight. For an economic use of such material combinations, the curing time of the resin system is an important factor. Therefor especially polyurethane resin systems offer a high potential for reduced curing times and elevated curing cycles. However, for the save use of such novel composite materials, the major mechanical performance requirements have to be proofed. The mechanical properties in general, are decisively depended on structural parameters of the composite material such as fiber content and orientation, fiber-matrix bonding as well as resin inclusions and imperfections. The latter are strongly influenced by the processing conditions, hence the influence of the manufacturing parameters time, temperature and pressure must be investigated [1-5].

2. Experimental

2.1. Material

In the present investigation a special polyurethane resin system, optimized for hand lay-up processing, was used [6]. The fiber reinforcement, consisting of 4 layers bidirectional glass

fiber complex with mainly unidirectional fiber rovings, was impregnated by the resin system and cured to laminates using a platen press. By the systematic variation of the curing parameters time, temperature profile and pressure profile, in total 8 laminate states according to 3 processing profiles were manufactured. In Table 1, the processing parameters for the isothermal and isobaric curing programs are shown. In this program, the parameters curing temperature and curing pressure were kept constant for the whole curing time.

| Laminate | Curing temperature [° C] | Curing pressure [MPa] | Curing time [min] |
|------------------|--------------------------------|-----------------------------|-------------------------|
| PU_4UD_80_6_6 | 80 | 0.6 | 6 |
| PU_4UD_120_6_2.5 | 120 | 0.6 | 2.5 |

Table 1. Laminate designations and curing parameters for the curing program "isothermal and isobaric".

In the isobaric curing program with temperature ramp, the curing process starts at room temperature and increases to an isothermal temperature level at 80 respectively 120 °C. A curing pressure of 0.6 respectively 1.5 MPa acts on the laminates during the curing process. The processing parameters are shown in Table 2.

| Laminate | Curing start temperature [° C] | Curing end temperature [° C] | Curing pressure [MPa] | Curing time [min] |
|----------------------|--------------------------------------|------------------------------------|-----------------------------|-------------------------|
| PU_4UD_30/80_6_9.3 | 30 | 80 | 0.6 | 9.3 |
| PU_4UD_30/120_6_8.5 | 30 | 120 | 0.6 | 8.5 |
| PU_4UD_30/80_15_9.3 | 30 | 80 | 1.5 | 9.3 |
| PU_4UD_30/120_15_8.5 | 30 | 120 | 1.5 | 8.5 |

Table 2. Laminate designations and curing parameters for the curing program "isobaric with temperature ramp".

At the third processing profile, as shown in Table 3 the same temperature profile consisting of a heating ramp and an isothermal level was used. The difference was that the curing pressure starts at 0.1 MPa and was increased up to 0.6 MPa in form of a pressure step at the end of the heating ramp.

| Laminate | Curing start temperature [° C] | Curing end temperature [° C] | Curing start pressure [MPa] | Curing end pressure [MPa] | Curing time [min] |
|-----------------------|--------------------------------------|------------------------------------|-----------------------------------|---------------------------------|-------------------------|
| PU_4UD_30/80_1/6_9.3 | 30 | 80 | 0.1 | 0.6 | 9.3 |
| PU_4UD_30/120_1/6_8.5 | 30 | 120 | 0.1 | 0.6 | 8.5 |

Table 3. Laminate designations and curing parameters for the curing program "temperature ramp with pressure step".

2.2. Structure analysis

The structural characterization of 8 different laminate states was performed by light microscopy. For this purpose laminate specimens were taken out of the middle of each laminate state, embedded in 2 component resin system, grinded and polished. The used light microscope Olympus BX51 with a magnification of 50 was suitable to determine the overall

structure of the laminates especially regarding fiber distribution as well as resin inclusions and imperfections.

2.3. Mechanical investigations

The mechanical tests under monotonic and cyclic load conditions were carried out in form of 3 point bending tests. For this kind of tests a servo-hydraulic universal testing machine, type MTS 858, was used. Test specimens, in form of beams with length of 100 mm and width of 10 mm were cut out of each laminate. For the monotonic tests, the test speed was 20 mm per minute, with a support distance of 40 mm. The same support distance was used for the cyclic tests, at stress ratio R of 0.1 and test frequency of 10 Hz. To carry out a lot of tests within a reasonable period of time, the maximum number of cycles was limited to 10^6 cycles.

3. Results and discussion

3.1. Laminate structure

3.1.1. Influence of the curing temperature profile

The influence of curing temperature profile on the laminate structure is illustrated by light micrographs in Figure 1. At the micrographs you can see, that the laminate PU_4UD_80_6_6 (Figure 1a), isothermal cured at temperature of 80 °C has a high void and defect content within the glass fiber rovings and also in the resin rich areas between the rovings. An increase of the isothermal curing temperature to 120 °C leads to a reduced defect content (Figure 1b). The laminates which were cured with a temperature ramp in the curing profile show no significant differences in the laminate structure compared to the isothermal cured laminates (s. micrographs in Figure 1c and 1d).



Figure 1. Light micrograph of the laminate **a**) PU_4UD_80_6_6, **b**) PU_4UD_120_6_2.5, **c**) PU_4UD_30/80_6_9.3, **d**) PU_4UD_30/120_6_8.5.

3.1.2. Influence of the curing pressure level

The curing pressure level has a significant influence on the laminate structure as shown in Figure 2. At the used curing temperature profiles, with a temperature level of 80 respectively 120 °C, the increase of the curing pressure from 0.6 MPa up to 1.5 MPa leads to a reduction of the void content at both investigated temperature levels. The most compact laminate with the lowest void content, laminate PU_4UD_30/120_15_8.5 (Figure 2d), was cured at highest pressure and highest temperature level.



Figure 2. Light micrograph of the laminate **a**) PU_4UD_30/80_6_9.3, **b**) PU_4UD_30/120_6_8.5, **c**) PU_4UD_30/80_15_9.3, **d**) PU_4UD_30/120_15_8.5.

3.1.3. Influence of the curing pressure profile

The micrographs for the investigation of the influence of the curing pressure profile on the laminate structure are shown in Figure 3. For the curing temperature profile ending at 80 °C, the pressure step in the pressure profile has no significant influence on the laminate structure. For the laminate cured at a temperature of 120 °C, the pressure step leads to a higher void content, compared to the isobaric cured laminates.



Figure 3. Light micrograph of the laminate **a**) PU_4UD_30/80_6_9.3, **b**) PU_4UD_30/120_6_8.5, **c**) PU_4UD_30/80_1/6_9.3, **d**) PU_4UD_30/120_1/6_8.5.

3.2. Mechanical short term properties

3.2.1. Influence of the curing temperature profile

Figure 4 shows the influence of the curing temperature and the curing temperature profile on the mechanical short term properties. An increasing curing temperature from 80 °C up to 120 °C significantly improves the flexural strength and modulus and leads to a reduction of laminate thickness. In the curing temperature profile, the temperature ramp up to 80 °C has a positive effect on the mechanical short term properties, whereas for the temperature ramp up to 120 °C is this effect was not observed. These results can be attributed to the laminate structure. A higher curing temperature leads to lower defect content and improved mechanical short term properties.



Figure 4. Flexural strength, modulus and laminate thickness for laminates, isobaric cured with different temperature profiles ending at 80 °C and 120 °C.

3.2.2. Influence of the curing pressure level

The influence of the curing pressure level on the short term mechanical properties is shown in Figure 5. An increasing pressure level up to 1.5 MPa leads to higher flexural strength and higher flexural modulus levels for both investigated curing temperatures. The highest flexural strength and modulus values were determined for the laminate PU_4UD_30/120_15_8.5, cured at a temperature level of 120 °C and at a pressure of 1.5 MPa. These findings are direct related to the laminate structure, which means that the laminate cured at the highest temperature and pressure level has the lowest void content of all investigated laminates.



Figure 5. Flexural strength, modulus and laminate thickness for laminates, isobaric cured at pressure levels of 0.6 MPa and 1.5 MPa and curing temperatures ending at 80 °C and 120 °C.

3.2.3. Influence of the curing pressure profile

Within the investigation of the influence of the curing pressure profile on the laminate structure the curing temperature is an important factor. Increasing mechanical short term properties as shown in Figure 6 were determined at a curing temperature of 80 °C, when a pressure step is used in the curing pressure profile. On the other hand at the curing temperature of 120 °C a decrease of the mechanical properties was observed in which also can be explained by the laminate structure based on the manufacturing parameters. At the higher curing temperature of 120 °C the low curing pressure at the beginning of the curing process leads to a kind of foaming up the polyurethane resin system with a resulting high void content in the laminate structure along with lowered mechanical properties.



Figure 6. Flexural strength, modulus and laminate thickness for laminates, cured with different pressure profiles at temperatures ending at 80 °C and 120 °C.

3.3. Fatigue behavior

3.3.1. Influence of the curing temperature

The dynamic stress vs. loading cycle curves (S-N curves) for the laminates which were cured isobaric with a temperature profile ending at 80 respectively 120 °C are shown in figure 7. Depending on the short term mechanical properties the laminate PU_4UD_30/120_6_8.5 has higher fatigue strength at a low number of cycles compared to the laminate PU_4UD_30/80_6_9.3. However, the influence of the stress amplitude on the fatigue strength of the laminate PU_4UD_30/120_6_8.5 is distinctly stronger, so that for a longer dynamic loading period, above 10^5 cycles, the laminate PU_4UD_30/80_6_9.3 shows a higher fatigue strength level. At the end of the experimental test range at 10^6 load cycles, the fatigue strength of the laminate PU_4UD_30/120_6_8.5 is about 35% less compared to the laminate PU_4UD_30/80_6_9.3.



Figure 7. S-N curves for laminates, cured isobaric with temperature profiles ending at 80 °C and 120 °C.

3.3.2. Influence of the curing pressure

The comparison of S-N curves for laminates cured with different pressure levels and profiles as illustrated in figure 8 shows that the laminate $PU_4UD_30/120_15_8.5$ which has the lowest void content also has the lowest fatigue strength especially for the long term loading, among the laminates investigated. The laminate $PU_4UD_30/120_1/6_8.5$ with the highest void content in the laminate structure shows elevated fatigue strength at cycle numbers above 10^4 which means that significantly higher void content leads to reduced dependency of fatigue strength on dynamic loading time. Consequently, higher defect content in the laminate structure leads to lowered short term mechanical properties with enhanced long term fatigue behavior.



Figure 8. S-N curves for laminates, cured with both, isobaric as well as pressure step in the pressure profile, at a temperature profile ending at 120 °C.

4. Conclusion

The present investigation of the influence of the manufacturing parameters on mechanical short term and fatigue behavior of polyurethane based composites pointed out a direct correlation between curing parameters, laminate structure and mechanical parameters. A high curing temperature level combined with a high isobaric curing pressure level leads to a compact, nearly void-free laminate structure which results in excellent short term properties, but limited fatigue behavior. On the other hand, imperfect laminate structures including a higher amount of voids show a reduced mechanical short term property level. However, the fatigue behavior of such imperfect laminates is elevated, with a lower decrease in fatigue strength with increasing number of cycles.

References

- [1] L. Liu, B. Zhang, D. Wang, Z. Wu. Effects of cure cycles on void content and mechanical properties of composite laminates. *Composite Structures*, 73: 303–309, 2006.
- [2] S. Husic´, I. Javni, Z. S. Petrovic´. Thermal and mechanical properties of glass reinforced soy-based polyurethane composites. *Composites Science and Technology*, 65: 19-25, 2005.
- [3] U. Younes. Development of PU-based RTM and VARTM Technology. In *Composites* 2010, 2010.
- [4] U. E. Younes and F. W. Bradish. Polyurethane composites for wind turbine blades. *JEC Composite Magazine*, 70: 29-34, 2012.
- [5] D. Bareis, D. Heberer, M. Connolly. Advances in urethane composites: Resins with tunable reaction times. In *Composites 2011*, 2011.
- [6] Hexcel Corporation. A world of composite technologies. http://www.hexcel.com, 19.02.2013, 2008.