EFFECTS OF WET AGING ON THE DAMAGE PROGRESS IN MULTI-DIRECTIONAL GFRP COMPOSITES UNDER FATIGUE LOADING

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ABSTRACT

The impact of harsh environments with variable temperatures, high humidity, or direct water contact poses a challenge for lightweight construction materials, such as polymer composites, because of their hydrophilic and temperature-dependent properties. Since the most widely used glass fiber reinforced polymers are known to suffer from the above-mentioned conditions, precise knowledge of their long-term durability is of high interest. In addition to the pure change of mechanical properties due to water absorption or temperature, damage evolution and lifetime performance are important. This is what the present study addresses, bringing together the initiation and development with the effects on fatigue life. In contrast to what is regularly reported in the literature, a lifetime increase was found for a specific multidirectional (quasi-isotropic), epoxy-based laminate after short- and long-term wet-aging. The reasons for this outcome were investigated using interrupted fatigue tests, microscopy, and residual strength analysis. As a result, significant differences in damage initiation and progress could be identified and related to the quasi-static and fatigue result. The main reason for the increase in lifetime is expected to be the distributed energy dissipation in the wet-aged condition. Here, damage occurs simultaneously in several regions, which leads to the fact that the introduced energy is divided among numerous defects at the same time. In the dry case, on the contrary, the growth of the few, individual damages terminates in an early catastrophic failure.

1 INTRODUCTION

Glass fiber reinforced polymers (GFRP) have been used for structural components in offshore and marine environments for several decades, but their demand is still increasing. Due to technical progress and increasing requirements on components, it is becoming more and more important to raise the weight and performance efficiency in these industries as well. In this context, knowledge of the long-term properties during application is essential, e.g., to safely reduce safety factors. However, most investigations have focused on the static mechanical properties of GFRP after (accelerated) aging procedures [1,2]. In contrast, the database of environmental fatigue and lifetime properties is relatively small. Furthermore, the results of published environmental fatigue tests show in parts contradictory results. Although some could hardly show any influence, others found a significant reduction in lifetime [3–6], as can be seen schematically in Fig. 1. The reasons for these different outcomes could be found among the aging conditions and durations, the fibers and specific resins used, the lay-ups and loading situations [7]. But a detailed analysis of how aging influences failure progress in detail is most often still missing.

As the authors also found an unexpected lifetime increase in the case of cyclic testing of multidirectional GFRP laminates after wet-aging in distilled water at 50 °C, analyzing the evolution of the underlying damage appears to be even more important. Therefore, the effects of wet-aging on the damage progress during the fatigue life are investigated in detail in this study. Consequently, interrupted cyclic fatigue tests, residual strength investigations, and microscopy, as well as long-term wet aging (14 month) and re-drying of long-term aged specimens were used to analyze the effects on the damage progress and the lifetime.

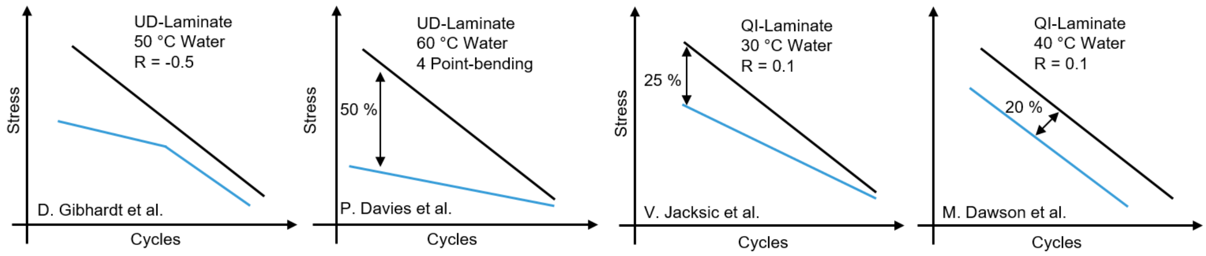

Figure 1: Schematic representation of environmental fatigue data published elsewhere by Gibhardt [3], Davies [4], Jacksic [5] and Dawson [6]. Black lines represent dry and blue lines wet-aged conditions.

2 MATERIALS AND METHODS

Multidirectional GFRP laminates were manufactured using a resin transfer molding process. Fabrics (G300U and G300BX from HACOTECH GmbH with PPG Hybon 2002 fibers) were stacked in a $[\pm45/(90)/0]_{2s}$ sequence and infused using Hexion EPIKOTE™ Resin MGS™ RIMR135 and EPIKURE™ Curing Agent MGS™ RIMH137. The (quasi) unidirectional fabrics are made of 86 % E-glass fibers in 0°-direction and 14 % backing fibers in 90°-direction, while the ±45° fabrics have an equal share in both directions. Laminates of 2 mm thickness have a total fiber volume fraction of 50 %. Infusion and curing were performed at 50 °C for 16 h, post-curing for 16 h at 80 °C. For fatigue investigations, rectangular specimens of 250 mm x 15 mm were tested using an Instron servo-hydraulic testing machine, equipped with a 100 kN load cell. All samples were tested load controlled under tension-tension loading with a R-ratio of 0.1 and at a frequency of 3 Hz. Damage progress was analyzed using optical inspection methods such as microscopy (Keyence VHX 6000, Japan) and high-resolution scans. Therefore, specimens were analyzed during defined points in time by interrupting the fatigue tests. Aging was achieved by total immersion of the samples in tempered water baths using distilled water at 50 °C. Fatigue tests were conducted after aging for 40 days (when saturation was reached) or after 420 days (to reveal the impact of long-term wet-aging). Additionally, a set of specimens was re-dried after long-term aging to investigate the permanent effects in the absence of absorbed water. To gain an insight into the progress of the damage, residual strength measurements were made after cyclic loading on dry and short-term aged specimens. During fatigue testing, all wet samples were covered with a bag containing moist fleece to counteract drying. Therefore, the loss of water during testing was limited to less than 0.25 m% (of the matrix fraction).

3 RESULTS AND DISCUSSION

3.1 Water absorption

The water absorption results for the neat epoxy resin and the QI-GFRP laminates are shown in Fig. 2. Saturation was reached for both types of specimens after about 40 days ($\sqrt{31}$ h) at 50 °C. The maximum weight gain of the epoxy was about 2.85 m% and thus slightly lower than expected from previous studies [3,7,8] (3.0 m%), which is probably an effect of incomplete drying before immersion. However, the drying process was similar for neat epoxy and GFRP specimens. A difference of approximately 0.15 m% (related to the matrix mass fraction) was found for the absorption within the GFRP during this period. The increased water absorption is typically due to effects in the fiber/matrix interphase and potential imperfections within the composite. However, a two-stage behavior or deviating water absorption could not be determined for short- or long-term immersion. The final water content inside the GFRP after 14 months (ca. $\sqrt{97}$ h) of aging was 3.15 m%.

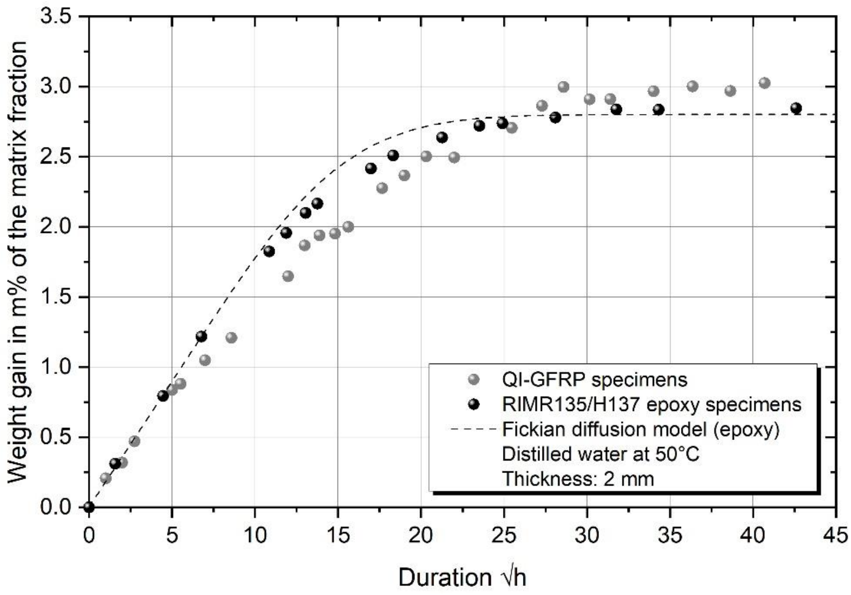

Figure 2: Water absorption of epoxy and QI-GFRP specimens until saturation in DW at 50 °C.

3.2 Quasi-static properties

The mechanical properties of the QI-GFRP specimens were determined by quasi-static tensile tests according to DIN EN ISO 527. As a result of the relatively high share of 0°-fibers, is the tensile strength of the dried specimens with 590±14 MPa comparably high for multidirectional GFRP laminates [9,10]. Conditioning of two weeks under laboratory conditions (22 °C and 50±10 % RH) leads to a slight decrease in strength due to moisture absorption. These conditioned specimens are referred as reference properties with a strength of 563±22 MPa. 40-day water bath aging until saturation causes a significant reduction in tensile strength of 36.5 %. The residual strength is 357±25 MPa in this case. To check the on the strain rate dependence of the strength, additional tests were performed at a loading rate of 315 mm/min, which corresponds to the rate during fatigue tests. Under these testing conditions, the reference strength increases by 15.1 %. Failure strains (2.7 to 3.0 %) and Young's moduli (21.6 to 22.7 GPa) are not affected by water absorption nor by the change in strain rate.

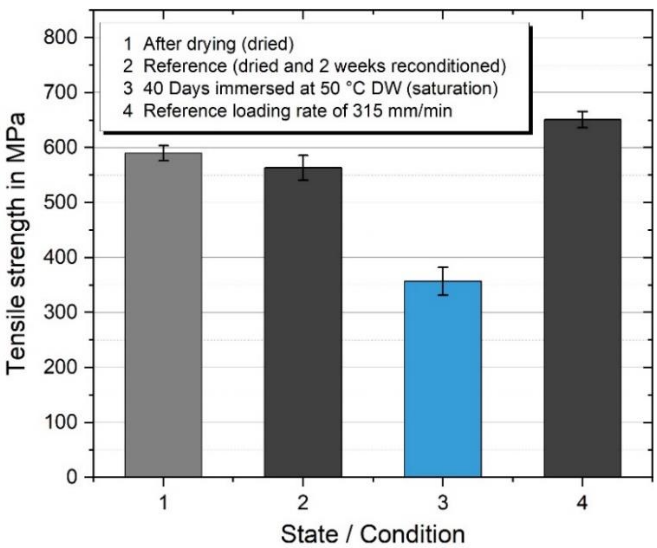

Figure 3: Quasi-static tensile strength in relation to the testing condition and aging-state.

3.3 Impact of wet-aging on tension-tension fatigue properties

In Fig. 4, the SN-diagrams of unaged, short-term (40 days) saturated, long-term (420 days) saturated, and re-dried specimens after immersion in 50 °C distilled water (DW) are shown. The Wöhler curves are fitted with a least squares method and are denoted in Table 1 in terms of:

$$\sigma(N_f) = \sigma_0 \cdot N_f^m, \tag{1}$$

where σ_0 is the axis passage, N_f are the cycles to failure, and m is the slope of the curve. While the fatigue lifetime of the unaged reference GFRP is comparable to other studies [11,12], it is surprising that the fatigue life of the short-term wet-aged composite is significantly increased at each load level up to 230 MPa, which is about 80 % of the wet and only 40 % of the static dry strength. These results are different to studies published before [3-6], where aging in hot/wet environments regularly decreased the fatigue lifetime. The fact that the lifetime after aging improves even though the static strength decreases drastically shows that it is often not possible to simply conclude from static to fatigue properties.

When also considering the long-term aged specimens, it becomes clear that there is a slight degradation over time, but that the fatigue lifetime, particularly for lower loads is still significantly increased. The amount of permanent damage and property deterioration can be estimated by contemplating the re-dried specimens. After re-drying, a slight reduction in fatigue performance compared to the reference can be identified. However, even though this reduction is significant, it is not as pronounced as the lifetime increase due to wet-aging. In general, it is evident that the Wöhler curves are declining in the wet-aged conditions compared to the dry ones. This results in a pronounced lifetime increase in the high-cycle regime at low loads and turns around for particularly high loads and low cycles ($<10^3$). Therefore, it can be concluded that a positive effect from water absorption predominates when the damage progress is driven by damage of the matrix and the interphases (inter fiber fracture and delaminations). On the contrary, a fiber driven damage progress (fiber strength related) at very high loads is negatively affected by the wet-aging. This is also in line with the significantly reduced static tensile strength, which fits well with the obtained Wöhler curves (Table 1).

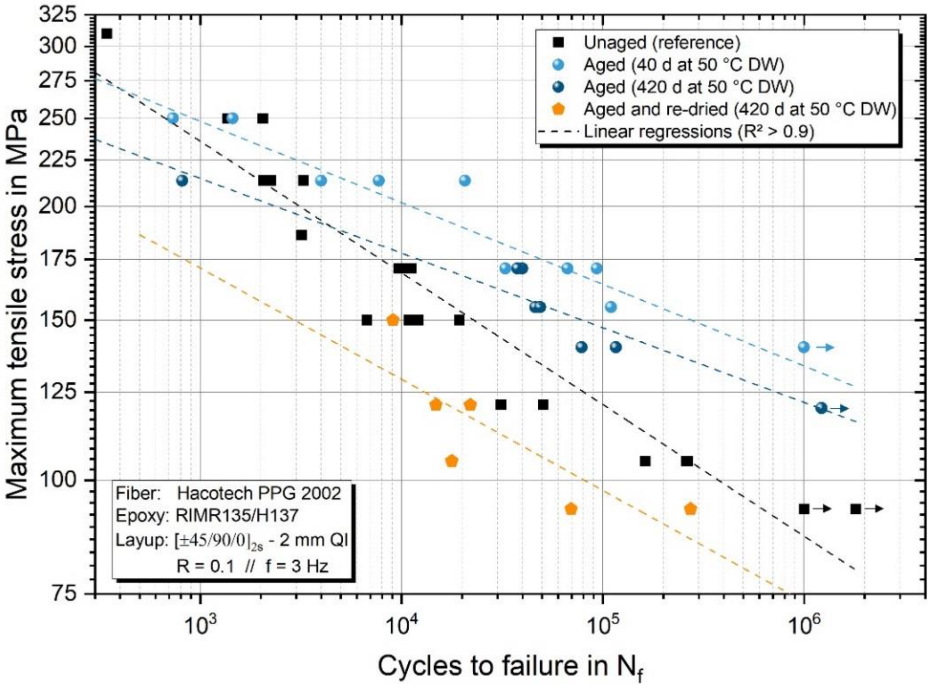

Figure 4: Tensile-tensile fatigue results and Wöhler curves for dry, wet-aged (40 days until saturation), wet-aged (420 days), and re-dried (420 days wet-aged, then re-dried) specimens.

Condition	Wöhler curve fit	$\sigma\ (10^6\ cycles)$ in MPa
Dry reference	$\sigma(N_f) = 641 \cdot N_f^{-0.145}$	87
40 days aged	$\sigma(N_f) = 461 \cdot N_f^{-0.090}$	132
420 days aged	$\sigma(N_f) = 378 \cdot N_f^{-0.082}$	121
420 days aged and re-dried	$\sigma(N_f) = 398 \cdot N_f^{-0.122}$	74

Table 1: Wöhler curve regressions and (calculated) load level to reach 10^6 cycles to failure.

To determine the factors that might explain these results in more detail, interrupted static and fatigue tests, dynamic modulus analysis, and optical damage studies were performed. Fig. 5 represents an example of the stiffness evolution of two samples (dry and short-term wet) tested at the same load level. Additionally, the extent of damage is shown for three points in time by high-resolution transmitted light scans. For aged samples, it can be seen that transverse cracks and delaminations occur much earlier and on a larger scale. Furthermore, especially the delaminations grow extensively and uniformly in the aged case. A growing delamination can be identified almost at every intersection. On the contrary, the unaged composite shows a relatively late and localized damage onset which rapidly leads to failure. In addition to the sheer number of growing damages, it is also important to note that water absorption can significantly change the fracture toughness of the matrix and the composite [13]. The progress of damage in terms of the stiffness evolution is also very dissimilar. While the stiffness in the dry condition decreases with an accelerating rate of about 400 MPa/1000 cycles, the evolution is both more steady and significantly slower with about 20 MPa/1000 cycles. This also indicates that in the wet case mainly interfiber fractures and delaminations occur, which have little influence on the stiffness. Under dry conditions, on the other hand, load bearing fibers are failing, which results in the acceleration of stiffness reduction.

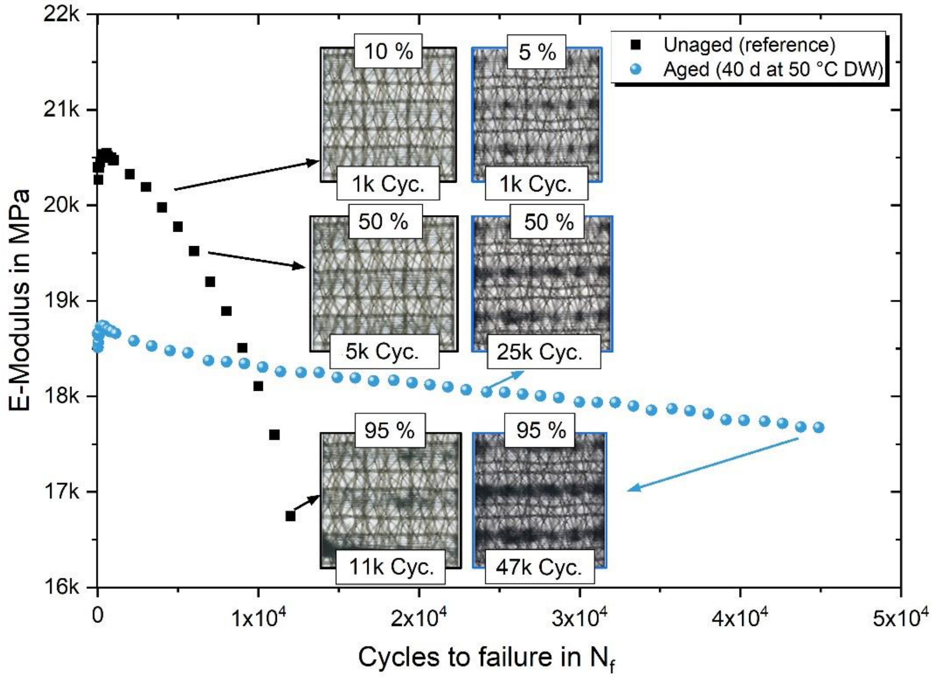

Figure 5: Stiffness and damage progress during fatigue testing at maximum tensile stress of 150 MPa for dry and wet (40 days aged) specimens.

In summary, it is hypothesized that although aging leads to an earlier and increased initiation of damage, local damage does not critically grow as a result. The reasons for this are, on the one hand, the reduced interfacial strength and increased matrix ductility and toughness, and on the other hand, the distribution of energy to numerous local defects. This is presented schematically in Fig. 6. However, even though the lifetime of coupon specimens is significantly increased during cyclic testing, it is a fact that the amount, initiation, and damage progress is drastically increased in the wet case as well. This fact should be critically considered for structures and parts in applications. As the lifetime of a coupon is limited by the geometry, which allows just a relatively small maximum damage increase until final failure, it might be very different in larger parts. Here, the significantly increased amount and characteristics of the accelerated damage growth could still be critical. Consequently, additional investigation is recommended on a part-size level based on the results of the coupon tests.

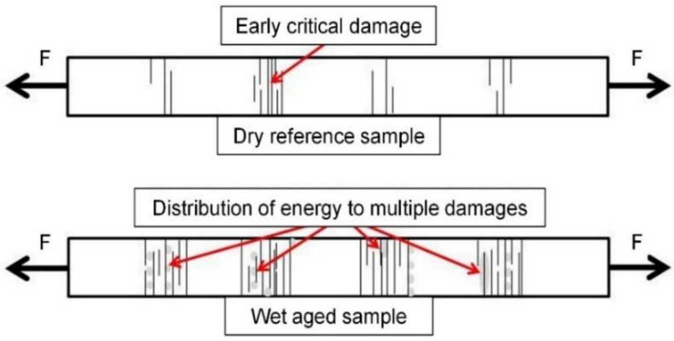

Figure 6: Schematic representation of damage growth as a result of energy distribution in dry and wet-aged samples.

3.4 Residual strength and one-parameter model

In addition to interrupted fatigue tests and evaluation of the stiffness degradation, residual strength tests were performed after cycling loading of dry and wet-aged specimens. Thus, quasi-static tensile tests were run after cycling loading on two different load levels and up to four different numbers of cycles. In Fig. 7, representative stress-strain diagrams (left) and the normalized stiffness degradation with corresponding residual strength values (right) are shown.

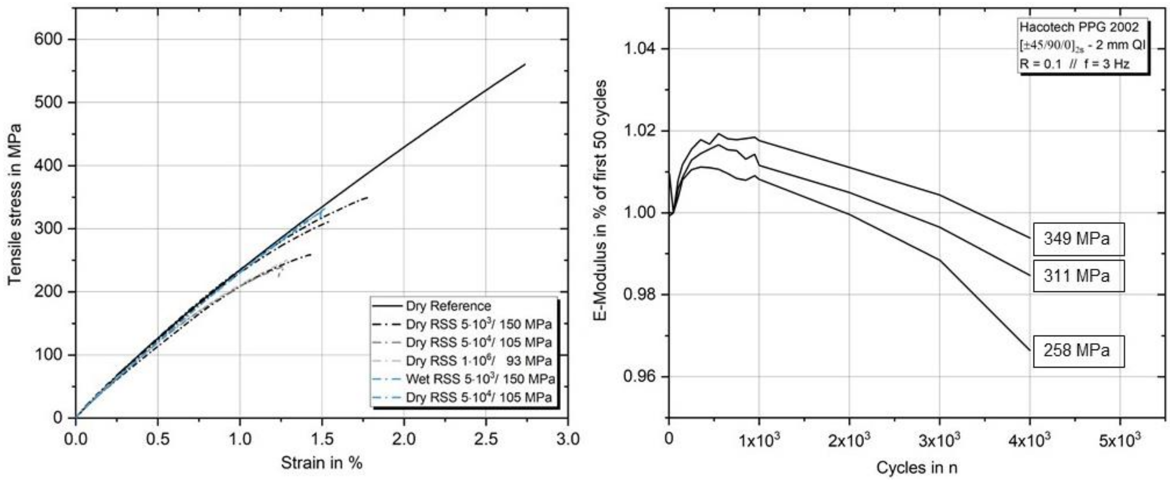

Figure 7: Representative stress-strain diagrams of residual strength after fatigue specimens (dry and wet) compared to the initial dry material.

From the stress-strain diagrams it is evident that, on the one hand, the behavior becomes nonlinear due to the initial damage of the fatigue load and, on the other hand, strength and strain to failure are significantly reduced. Based on the representation in Fig. 7 (right), it can be derived that the residual strength correlates with the relative loss of stiffness due to damage from cycling loading. In Fig. 8, the residual strengths of all tested configurations are presented as absolute (left) and relative (right) values.

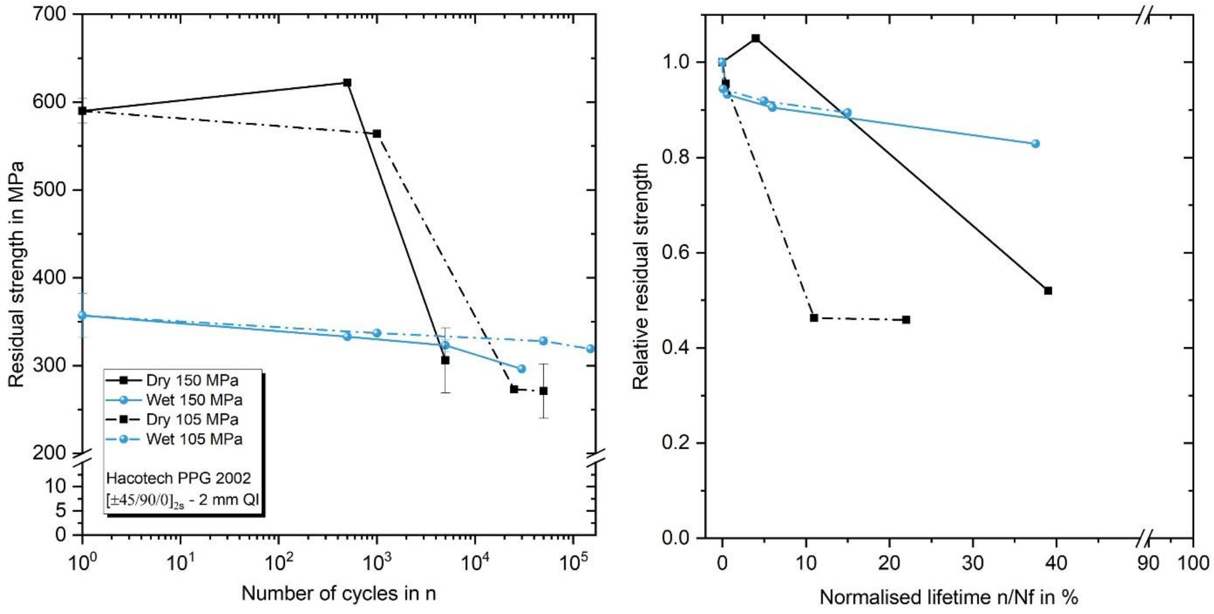

Figure 8: Absolute (left) and relative (right) residual strength vs. lifetime curves for dry and wet (40 days wet-aged) specimens. The lines are used to guide the eyes.

The residual strength tests again reveal that the damage progress is fundamentally different in dry and wet specimens. Thus, it can be seen that although the strength of the wet specimens is drastically reduced from the beginning, it is almost not further reduced due to cycling loading. The damage introduced (clearly visible on the scans), therefore, has hardly any influence on the residual strength. The situation is quite different for the dry samples. Here, a dramatic loss of residual strength can be identified as soon as the first damage is introduced and starts to grow (~ at 10^3 cycles for 150 MPa loading). As a result, the residual strength of the dry specimens drops below the wet-strength, which finally ends with a sudden catastrophic failure in cyclic loading.

An attempt to analytically describe the correlation between fatigue performance, static, and residual strength is to plot the data in a strength-degradation plot and describe the residual strength as a monotonic decreasing property [14]. Single-parameter models can typically differentiate between early damage, sudden failure, or linear degradation types of evolution. One proposed method is to describe the residual strength S_r as a function of the initial strength S_0 and the maximum stress during loading S_{max}. As described by Nijssen et al. [14] the evolution can be formulated as:

$$S_r = S_0 - (S_0 - S_{max}) \cdot \left(\frac{n}{N_f}\right)^C, \quad (1)$$

with the number of cycles n, the average number of cycles to failure N_f, and the degradation parameter C. From the plot in Fig. 9 it becomes clear that an approximation with a single-parameter model is possible, but not exceedingly precise. Similarly to the residual strength curves shown in Fig. 8, a classification is possible for an early degradation type behavior (C < 1) of the dry specimens. But however, the difference to the wet-aged specimens is not as clear as before by considering the C-value of about 1.2 (linear to sudden failure) for the wet-aged specimens. By extending the investigation to further load levels and increasing the number of residual strength tests, the difference might become clearer. Nevertheless, the investigations presented within this study reveal that, for future applications, a more sophisticated two- or multiparameter model is required.

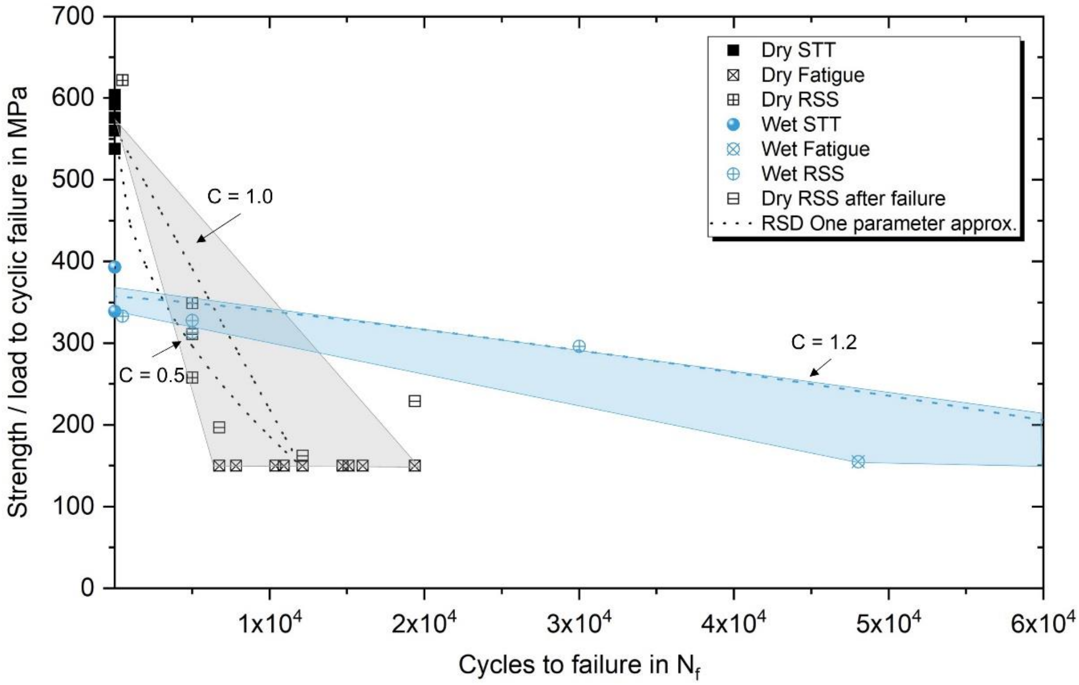

Figure 9: Residual strength degradation (RSD) plot and one-parameter approximation for dry and wet (40 days wet-aged) specimens.

4 CONCLUSIONS

The presented study on the effects of wet-aging on the lifetime and fatigue properties of multidirectional, epoxy-based GFRP laminates revealed a partially unexpected increase in the lifetime of wet coupon samples. Contrary to results usually reported in the literature, the energy dissipation due to extensive multi-local damage initiation and progress in combination with an increased fracture toughness of the wet epoxy resin extends the fatigue lifetime of the GFRP specimens in classical cyclic testing. In the specific example, based on the accompanying investigations and residual strength tests carried out, it is reasonable to hypothesize that interfiber damage occurs mainly, but much more frequently, in the wet case. The load carrying 0°-fibers appear to be less affected by the resulting damage. The situation is different in the dry case. Here, significantly less interfiber damage occurs, but the damage that arises affects the load-bearing fibers to a greater extent. Long-term aging of up to 14 months in 50 °C distilled water showed only little additional degradation compared to short-term aging of 1.5 months. Likewise, re-drying after long-term aging resulted in a slight lifetime decrease compared to the initial dry reference condition. Thus, it could be shown that permanent damage is caused by wet aging, but this is outweighed by the positive effects of water absorption. However, since damage such as interfiber fracture and delaminations occurred considerably earlier, to a greater extent, and more rapidly growing in the wet case, the lifetime results should be checked on part-sized samples in the future.

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