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# Nonlinear time dependent behaviour of epoxy resins

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**Abstract.** The nonlinear behaviour of epoxy resins is studied on standard tensile tests. A strain field measurement system is applied (Aramis) in order to monitor local strains. The residual strain is measured by recovering the specimens for up to 68 hours after unloading. The time span the specimen is exposed to load has a large influence on the creeping process and the residual strain after recovering. This is studied by comparison of instantaneous unloading with keeping the specimen under permanent load for thirty minutes. It is shown that moderate differences in the initial strain can lead to large differences in the creep behaviour as well as in the residual strain.

## 1. Introduction

The failure initiation of fiber reinforced composites is strongly influenced by the mechanical behaviour of the matrix. Failure of plies with a high loading transverse to the fiber direction is initiated on microscale either by interface or matrix failure. For the simulation of microfailure a district description of the matrix material behaviour is needed. Since epoxy resins are widely used in high strength materials applications e.g. in aircraft, wind energy and automotive industry the present study is focused on them.

Epoxy resins possess a complex mechanical behaviour [1-5]. They exhibit a strong nonlinearity, they are highly time dependent and they can undergo large deformations. They behave as viscoelastic as well as viscoplastic solids. The superposition of time dependence and plasticity makes the determination of the respective material properties very elaborate. A further problem arising in the determination of material properties is that the deformation in tensile specimens is not homogeneous. The occurrence of large deformations is limited to a small zone. As a result, in one region the material may behave predominantly viscoelastic while in another region it behaves viscoplastic. This gives the classical tests such as relaxation and creep test a different meaning. The typical values of strain to failure found in literature range from four to eight percent. These values presumingly are not taking into account the local nature of plasticising. Tensile tests show that at a certain load large deformations develop leading to local strains of more than 50%. The contribution of these high deformations to the total change in length of the specimen, however, is small due to the short length of the zone. It must be doubted that all strain to failure values given in literature are indeed true local strains but rather averages over the specimen length.

The stress/strain behaviour depends on various parameters. That means, for example, that the residual strain left after unloading depends on the level of the strain applied, the duration of the loading process and the time the material could recover after unloading. When we classify the material behaviour as viscoelastic or viscoplastic we have to keep in mind that these material models are still idealised models of the actual mechanical behaviour. The present study does not deal with the

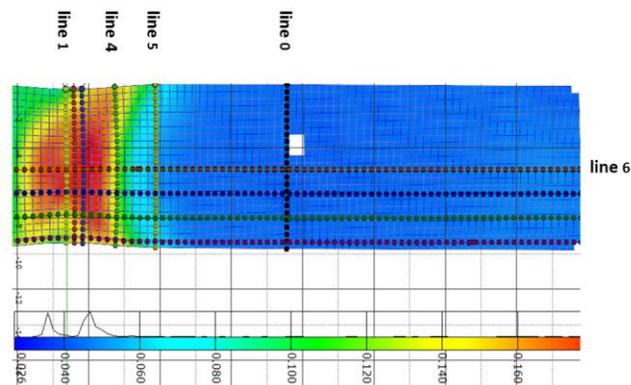


development of constitutive equations but is restricted to a phenomenological level. The tests performed so far do not represent a full matrix of parameter variations. The study aims to identify the governing parameters in order to design a test program in an appropriate manner.

## 2. Specimen preparation

The resin used for the present study is a L135/H137 epoxy system manufactured by Hexion. The tests are performed using standard dogbone tensile specimens with a total length of 150 mm, a width of 10 mm and a thickness of 4 mm. The specimens are cut out of plates by a milling process. The tests are performed by displacement control with a loading speed of 1 mm/min. Since the material can undergo large viscoplastic deformation the formulation utilises Hencky strains (logarithmic strains). In order to allow local strain measurement a strain field measurement method (digital image correlation) is applied. In the present study the Aramis system is used.

The specimens are prepared for the strain field measurement by applying a black/white patterns on the specimens. The patterns cover the full width and have a length of 65 mm. The distance of the grid lines of the Aramis digital imaging system is 1 mm in either direction. The strains are evaluated at specified lines in longitudinal and in transverse direction (figure 1). Line 1 is located in the center of the zone where large deformations appear. In this region the material behaves viscoplastic. If strain values are used in the following for the stress/strain curves the averages over the cross sections are taken. Line 4 and 5 are located on and near the edge of the viscoplastic zone. Line 0 is located in a region where the material behaves viscoelastically. In order to visualise the strain distribution in longitudinal direction of the specimen one line is located in the middle of the specimen (line 6). In this case the strain on every point is taken, this is, no averaging is done.



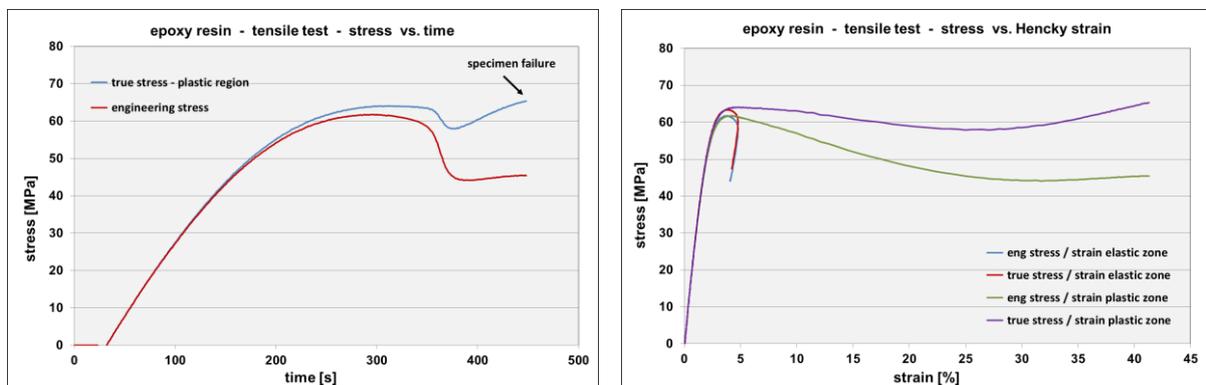
**Figure 1:** Positions of Aramis strain data lines

## 3. Stress / displacement and stress / strain curves

The typical stress/displacement behaviour at a constant loading rate of 1 mm/min is shown in figure 2. The specimen is loaded by displacement control with a rate of 1 mm/min. The engineering stress (red line) linearly increases in the first phase of the loading process which corresponds to a basically linear elastic behaviour with evanescent small time dependent deformation. Then the slope decreases and the curve becomes strongly nonlinear leading to a substantial decrease of the engineering stress. The nonlinear behaviour is caused by primarily three effects. First, the time dependence of the mechanical properties grows with the strain level. Second, additional viscoplastic deformations develop at a certain strain level. Third, the cross section decreases leading to an increase of the compliance. The decrease of the cross section necessitates the discrimination between true stresses and engineering stresses. The lateral strain is also measured by the Aramis system allowing the calculation of the true stresses in the high strain zone. Since the material is isotropic the change of the thickness can be calculated directly from the change of the width. The deviation of the true stresses (figure 2a, blue line) from the engineering stresses (red line) becomes significant after 150 seconds loading time which

corresponds to an average strain of roughly 2%. When the engineering stresses drop down the true stresses also decrease rapidly but only for a short time. Then the true stresses increase again until specimen failure where the stresses slightly supersede the prior maximum. At failure the difference between the true stresses and the engineering stresses is maximal reaching about 30%.

The different material behaviour in the viscoelastic and in the viscoplastic zone is visualised by the stress/strain curves in the respective zones (figure 2b). The engineering and the true stresses are plotted against the strain in the viscoelastic as well as in the viscoplastic zone. Since the maximum longitudinal strain in the viscoelastic zone is 4.7% the true stress is about 9% higher than the engineering stress. After the stress maximum is reached the strain in the elastic zone even decreases. This is caused by a strain redistribution in the specimen due to large deformations in the viscoplastic zone which is addressed in a later section. The longitudinal strain in the viscoplastic zone grows very rapidly and reaches more than 40% (figure 2b).

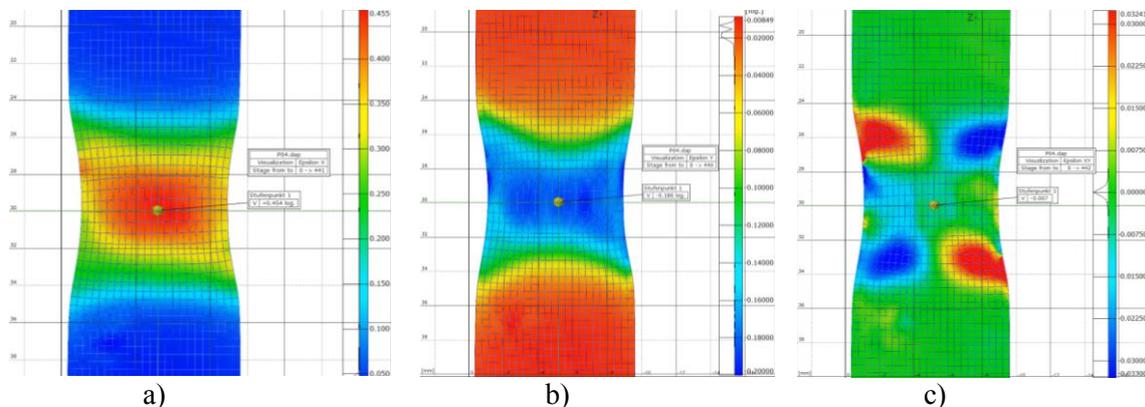


**Figure 2 a):** Stresses vs. displacement engineering stress (red line), true stress (blue line)

**b)** Engineering stress / true stress vs. strain viscoelastic zone (blue line, red line), viscoplastic zone (green line, violet line)

#### 4. Strain distribution in the specimen

The strain distributions delivered by Aramis in and around the high strain region are shown in figure 3. The plots correspond to the last step before ultimate specimen failure occurs. On the left hand side the strains in longitudinal direction (loading direction) are shown. The necking is very pronounced (true scale, no graphical enlargement). The strains decrease slightly towards the edges of the specimen. This is caused by the change from plane strain to plane stress conditions. The transverse strains are negative, this is, the lowest strains appear as red and vice versa. As expected, the transverse strain



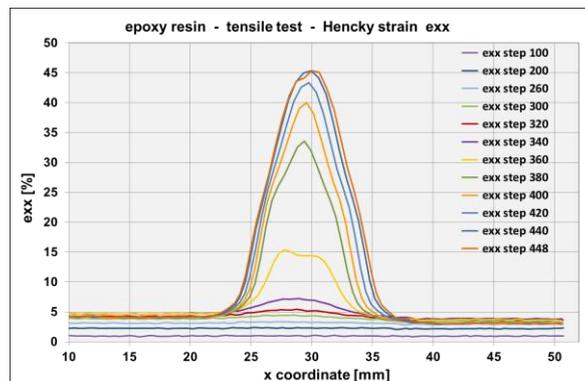
**Figure 3:** Strain distribution in the center part of specimen: a) longitudinal strain; b) transverse strain; c) shear strain

distribution in general is very similar to the longitudinal strain. On the rims of the viscoplastic zone (e.g. yellow region), however, the strains decrease a little faster towards the edges of the specimen. Substantial shear strains arise at the rims of the viscoplastic zone in the vicinity of the specimen edges which is due to the change of the width of the specimen. Even though the shear strains are much lower compared to the maximum longitudinal and transverse strains they are of the order of the longitudinal strains in the viscoelastic region.

#### 4.1 Strain distribution along the specimen during loading

In order to get an overview over the development of the longitudinal strains during the loading process the strain along the center line (line 1) is examined. Figure 4 shows the strains at different time steps. One has to keep in mind that the coordinate system Aramid refers to is the actual coordinate system, that is, the Eulerian coordinate system. This means that during the loading process a point fixed on the specimen, this is, in terms of continuum mechanics a material point, is shifted. The shift of the position however is moderate. The center point of the zone where large deformations occur shifts within the relevant time span (300s to 448s) by 1.6 mm.

During the increase of the load (applied displacement) a zone with large deformations forms at a certain load. The zone is very limited and virtually does not extend during the further loading process. A typical experiment is shown in figure 4. In the first phase of loading (strain below 4%) the longitudinal strain is distributed basically homogeneously in the specimen. The strain grows proportional to the applied displacement. When the strain exceeds 4% (time step 300, green line) the strain starts to grow faster in a limited zone of about 10 mm in length. With further increase of the applied displacement the strain rapidly grows and reaches a maximum of 46% just before specimen failure. This corresponds to a linear strain of more than 58%. The high strain zone does not substantially extend with the time. Outside this zone the strain changes only moderately. This shows that the epoxy resin has the ability to locally undergo large local deformations. The question is which portion of the deformation is viscoelastic, this is, will recover with the time and how big is the viscoplastic portion. These questions will be addressed later.



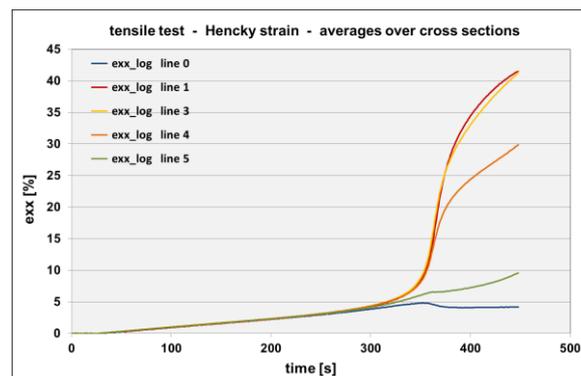
**Figure 4:** Strain along central line at different time steps (load steps)

#### 4.2 Time dependence of strains at different locations

The course of the strain versus time is analysed at different cross sections along the specimen (figure 5). The data represent the averages over the respective cross sections. Over a long time span the strains measured at any position increase linearly with the time. Since the tests are displacement controlled the time is equivalent to the applied displacement, e.g. to the load. At about 4% strain the curves begin to differ. The strains at two cross sections start to increase very rapidly. These lines correspond to the center (red line) and the rim (yellow line) of the viscoplastic zone. Both strains grow simultaneously up to about 12% which is reached at 350s. At this point the strains at the rim of the viscoplastic zone

start to grow slower. The strains outside the high strain zone even decelerate while the strain in the viscoplastic zone still rapidly grows.

The strain in the viscoelastic region (light blue line) only slightly increases after the 4% mark. When the viscoplastic strains start to grow very fast the strains in the viscoelastic region even slightly decrease. The strain in the cross section closer to the viscoplastic zone grows a little faster and reaches 10% in the moment of ultimate failure. The decrease of the strain in the viscoelastic region is caused by the over proportional growth of the strain in the high strain zone. Since the external displacement is prescribed a redistribution of the strain field has to take place.



**Figure 5:** Strain vs. time; average over cross sections at different positions along the specimen

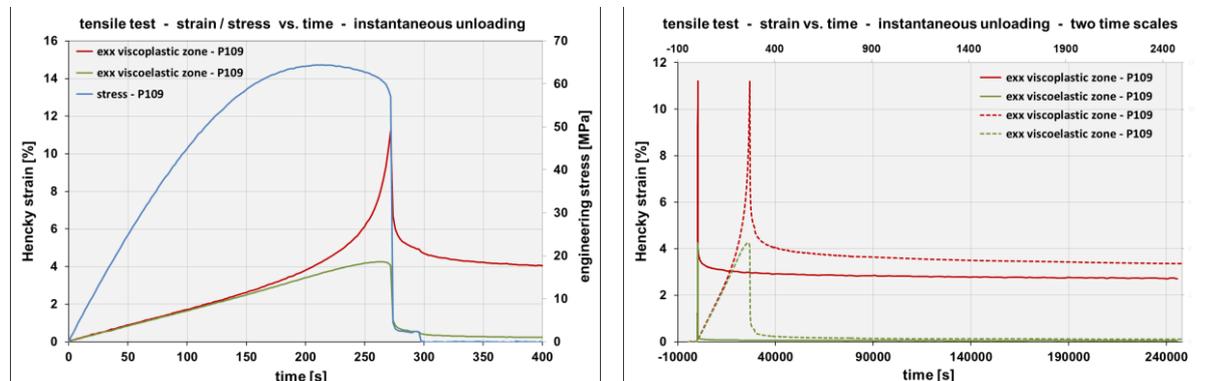
#### 4.3 Strain recovery after unloading

The strain in the high strain zone is a superposition viscoelastic and viscoplastic strain. In order to determine the viscoplastic portion a series of tests was performed where the specimens were unloaded and the strain recovery was monitored. Either the specimens were immediately unloaded or they were exposed to a permanent load before unloading. It's a matter of fact that the strain distribution along the specimen monitored by the digital imaging system is not known during the test. Accordingly it is not possible to reach a specified strain in the respective zones. As a result, the strain levels of different tests cannot be predefined and accordingly underlie some fluctuation. This reduces the accuracy in the estimation of the influence of parameters such as the duration of loading.

**4.3.1 Immediate unloading, high initial strain, small residual strain** First, the strain recovery is monitored for instantaneous unloading. At a certain displacement, which corresponds to a maximum strain of 11.2% (see below) the specimen is rapidly unloaded with a rate of 85mm/min. Subsequently the strain is monitored over 68 hours. The strain versus time is evaluated for two different zones in the specimen (figure 6a), the viscoelastic (green line) and the viscoplastic zone (red line). For low strains the curves more or less coincide. After the strains have reached 2% the strain in the viscoplastic zone grows increasingly faster while the growth of the strain in the viscoelastic zone is decelerated. Just before unloading the strain in the elastic zone even decreases. This is due to the fact that the strain in the viscoplastic zone grows faster than the external displacement. As already mentioned, this causes a redistribution of the strain in the specimen

In order to improve the visibility of the loading process the strains are displayed on two different time scales in figure 6b. The dotted lines refer to the enlarged time scale representing the first 40 minutes of the recovery phase, the continuous lines correspond to the full time span of 68 hours. Just before unloading the strain in the viscoplastic zone is 11.2% while in the viscoelastic zone it is 4.1%. When the specimen is unloaded both strains instantaneously decrease. Within the first 40 minutes the strain in the viscoelastic region vanishes. The viscoplastic strain decreases to 3.4% which is a loss of 70%. At the end of the recovery process the strain in the viscoplastic zone is reduced to 2,7%. This shows that more than 75% of the initial strain is viscoelastic and the plastic portion of the strain is

rather small. A completely different behaviour is encountered if the specimen is further exposed to load which is studied in the next section.

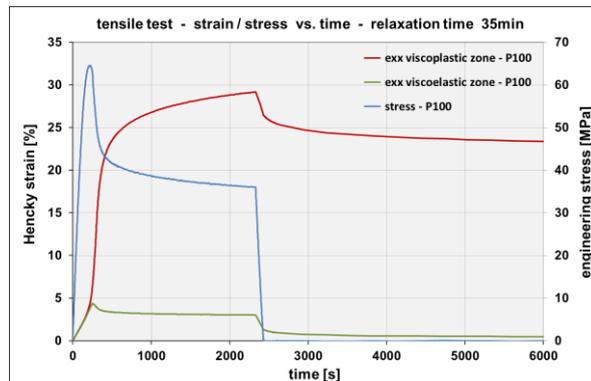


**Figure 6a):** Instantaneous unloading: stress/strain vs. time in viscoplastic and viscoelastic zone; initial phase **b)** Instantaneous unloading: strain vs. time in viscoplastic and viscoelastic zone; displayed on two different time scales

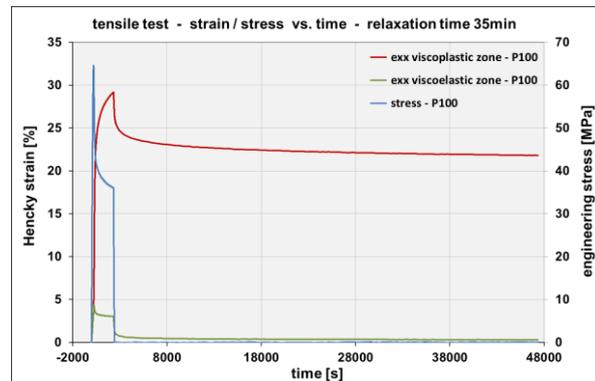
*4.3.2 Permanent load, higher initial strain, high residual strain* In a second test the influence of the time the specimen is exposed to load is studied. To this end the applied displacement is kept constant at a certain level for a specified time span. Keeping the displacement fixed does not give a classical relaxation test since the strain in the specimen is not homogeneous. As a result, it is not possible to control the strain of the total specimen. Alternatively the strain in a certain region could be controlled by using an additional strain measurement tool such as a strain gauge. However, different strains cause a different creep behaviour. Keeping the strain constant, e.g. in the viscoelastic region, does not prevent creeping in the viscoplastic zone. As a result, the stress relaxation is not only due to the relaxation in the region of interest, i.e. where the strain is kept constant but an additional decrease of the stress occurs due to the loss of stiffness caused by creeping in the viscoplastic region. Another alternative is to control the external displacement which is equivalent to prescribe the average strain in the specimen. In this case again a redistribution of the strains occurs in the specimen also resulting in different creep and relaxation processes in the respective regions.

The specimen is loaded up to a specified displacement (4.2 mm) corresponding to a stress of 62.1 MPa (figure 7a, blue curve). The displacement is held constant for 34 minutes. During this time span the stress relaxes down to 36.1 MPa which is a loss of more than 40%. In the moment when the displacement is kept constant the strain in the viscoplastic zone is 6.7% while it is 4.3% in the viscoelastic zone. While the strain in the viscoelastic zone decreases to 3.0%, which is a loss of 30% the strain in the viscoplastic zone enormously grows due to creeping and reaches 29.2% at the end of this phase. This means that the strain grows by 330%. The creeping of the viscoplastic zone enforces a decrease of the strains in the viscoelastic zone since the average strain is constant. This means that three processes are active simultaneously: relaxation, creep and strain recovery.

After the end of the load phase, this is, the increase of the external displacement together with the holding phase, the specimen is unloaded and the strain is monitored for another 12 hours. The strains in the viscoplastic and in the viscoelastic zone decrease instantaneously by 1.7 and 2.5%, respectively (figure 7a). The complete recovery process is shown in figure 7b. While the strain in the viscoelastic region levels off during the recovery phase the strain in the viscoplastic zone recovers by only 5% yielding to a residual strain of 22%. This means that during the permanent loading phase the material has experienced large plastic deformations.

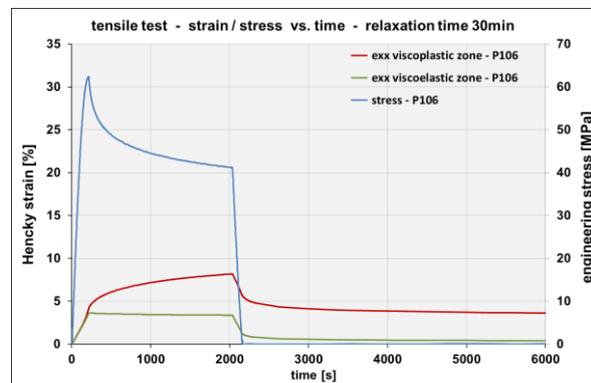


**Figure 7a):** Holding time 30min: stress/strain vs. time in viscoplastic and viscoelastic zone; initial phase

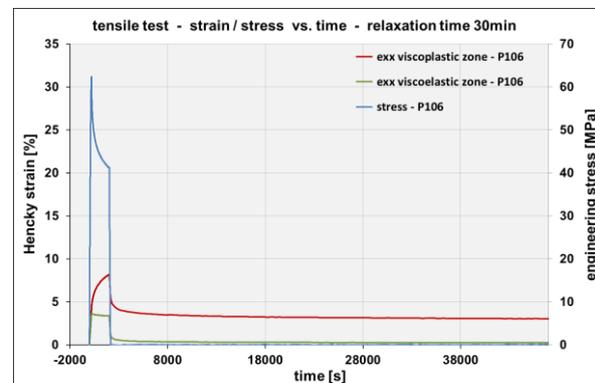


**b)** Holding time 30 min: stress/strain vs. time in viscoplastic and viscoelastic zone; initial and recovery phase

*4.3.3 Permanent load, medium initial strain, small residual strain* A similar test is performed with a slightly lower external displacement of 3.55 mm. Even though the reduction of the applied displacement is just 15% it has major consequences for the strain in the viscoplastic zone. The corresponding stress is even a little higher (62.3 MPa) since in either tests the stress is already beyond the maximum (figure 8a). The displacement is held constant for 30 minutes. During this time span the stress relaxes down to 40.7 MPa which is a loss of 35%. At the end of the loading process when the phase of constant displacement begins the strain in the viscoplastic zone is 4.2% while it is 3.6% in the viscoelastic zone. While the strain in the viscoelastic zone slightly decreases to 3.3%, the strain in the viscoplastic zone increases substantially and reaches 8.7% which is a growth of about 110%.



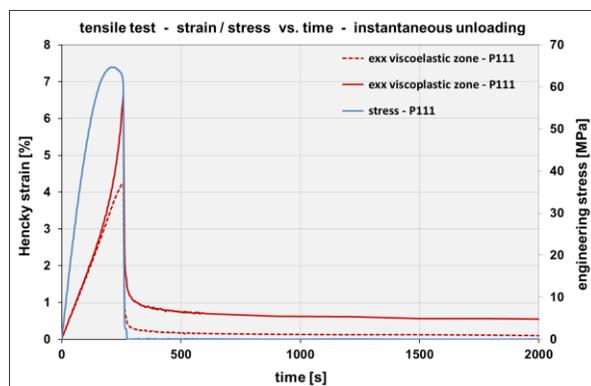
**Figure 8a):** Holding time 30min: stress/strain vs. time in viscoplastic and viscoelastic zone; initial phase



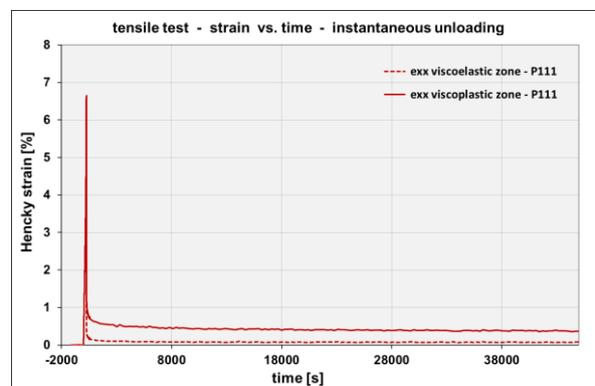
**b)** Holding time 30 min: stress/strain vs. time in viscoplastic and viscoelastic zone; initial and recovery phase

Again, the specimen then is unloaded and the strain is monitored for 12 hours. The strains in the viscoplastic and in the viscoelastic zone decrease instantaneously by 1.1 and 2.7%, respectively. The creeping process is shown in figure 8b. After 12 hours recovery time the residual strain in the viscoplastic zone is only 3% while in the viscoelastic zone it has vanished. In the first experiment with permanent loading (section 4.3.3) the initial strain before permanent loading is 6.7% while in the actual test it is 4.2%. During the phase of permanent loading the strain in the actual experiment reaches 8.7% while the other specimen ends up with 29.2% strain. After unloading and strain recovery the residual strain is 3% and 22%, respectively. This shows that a moderate increase of the initial strains has a large influence on the creep and recovery behaviour in the viscoplastic zone. Obviously the material is very sensitive against small changes of the strain level.

**4.3.4 Immediate unloading, medium initial strain, small residual strain** In a fourth test, a specimen loaded up to a maximum strain of 6.7% (figure 9a) which equalises the strain in the first test at the end of the loading process. It should be noticed that the scale of the strain is strongly enlarged in figure 9a. The corresponding minimum strain is 4.2%. In contrast to the former test the specimen is rapidly unloaded with a loading rate of 85 mm/min. The strains instantaneously drop by approximately 3%. After 12 hours recovery time the maximum and the minimum strain are decreased to 0.4% and 0.1%, respectively (figure 9b). In figure 9b for the sake of visibility the stresses are not displayed. The strain at the end of the loading process is the same as for the specimen studied in the preceding section where the loading process was followed by phase of permanent load. The instantaneous unloading obviously prevents the specimen to develop noteworthy viscoplastic strains. The specimen behaves predominantly viscoelastic. Comparison with the first test (4.3.1) shows that of course the residual strain increases with the magnitude of the initial strain. However, the influence of the time the specimen is exposed to load is much higher.



**Figure 9a):** Instantaneous unloading: stress/strain vs. time in viscoplastic and viscoelastic zone; initial phase



**b)** Instantaneous unloading: strain vs. time in viscoplastic and viscoelastic zone; initial and recovery phase

## Conclusions

The viscoelastic/viscoplastic behaviour of epoxy resin is very sensitive against the level of strain and especially the time the specimen is exposed to load. Extensive creeping can occur with local strains higher than 50%. In order to develop a constitutive equation taking into account the essential features of the material requires an extended test program.

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