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Lifetime Prediction with Combined Hygro-Thermo-Mechanical Influences on Thick Layer Adhesives

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Semi-structural bonded joints of steel components are exposed to long-term stresses in addition to water diffusion, temperature changes and varying external forces during operation. These hygro-thermomechanical loadings lead to successive degradation of stiffness and strength due to damage resulting from chemical aging and fatigue processes, causing failure of the bonded joint when its service life is reached. In the following contribution, a methodology is presented to predict the service life of hygro-thermo-mechanically loaded semistructural adhesive joints with transient FE simulation. The lifetime prediction is based on a constitutive model with a viscoelastic and a damage part. The first part for viscoelasticity consists of the generalized Maxwell model, in which the effects of temperature changes and varying humidity on the viscoelastic properties of the adhesive bond are captured by the time-temperature and time-water concentration shifts. The second part for the material damage is based on an ordinary differential equation for the damage evolution. It consists of a creep and a moisture damage part for the void developments caused by the local water concentration due to mechanical stress and chemical aging. Both damage parts are multiplied by an Arrhenius-type function to account for the effect of temperature change on the defect growth. The interaction of the creep and the moisture damage part is studied by applying the analytical and numerical solution of the damage differential equation. All parameters of the damage model are determined by using fracture times from creep tests under different climatic conditions and loading levels. The local water concentration in the adhesive is calculated by Fick's model and a concentration boundary condition. The diffusion parameters are determined by gravimetric measurements. A numerical example is setup to demonstrate the lifetime prediction methodology and to show the influence of viscoelasticity on the predicted lifetimes for different hygro-thermo-mechanical loading cases.

1 Modeling and simulation of moisture diffusion

In the course of the diffusion scenario, outlined in Fig. 1, it can be observed that water leads to swelling and to an increase of the weight of the adhesive specimen. In addition to these two effects, water diffusion also has an impact on the mechanical properties of the adhesive, and a differentiation is made here between the reversible and irreversible effects. The term reversibility here does not refer to a thermodynamic process and to the fact that no entropy is generated. Rather, reversibility in the following is to be understood as the reversibility of a chemical process in which a state of equilibrium is always aimed at. Accordingly, the meaning of the term reversibility is similar to the reversibility of a reversible chemical reaction.

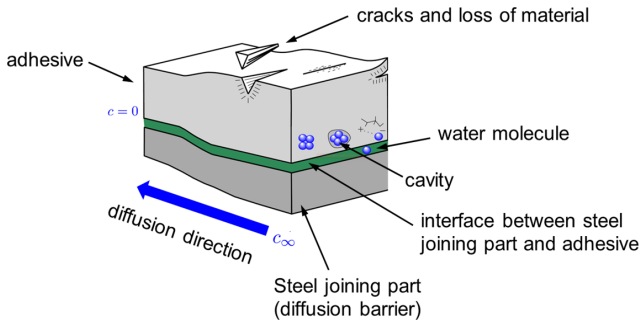

Fig. 1: Schematic process of moisture diffusion into adhesive / Image based on [7]

The mathematical description of the diffusion problem is based on the FOURIER heat conduction [2] analogous to first FICK's equation [1], [4], which gives the relationship between the concentration gradient, the diffusion coefficient D, and the diffusive flow or mass flow vector $\mathbf{J}$:

$$\mathbf{J} = -D\boldsymbol{\nabla} c \, . \tag{1}$$

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The diffusion coefficient D is a material parameter. It corresponds to a diffusion velocity and, therefore, indicates how fast a substance diffuses into another material. Together with the continuity equation [3]:

$$\frac{\partial c}{\partial t} = -\boldsymbol{\nabla} \cdot \mathbf{J} \tag{2}$$

the 1st FICK equation becomes the diffusion equation [3]:

$$\frac{\partial c}{\partial t} = \boldsymbol{\nabla} \cdot (D \boldsymbol{\nabla} c) \ . \tag{3}$$

If the diffusion coefficient D is constant and in particular independent of the location $\mathbf{x}$ and the concentration (see [4]), so the second FICK's equation results [1]:

$$\frac{\partial c}{\partial t} = D(\boldsymbol{\nabla} \cdot \boldsymbol{\nabla})c = D\boldsymbol{\nabla}^2 c \tag{4}$$

which is in the one-dimensional case [4]:

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} \tag{5}$$

The solution of the 2nd FICK equation can be obtained by the method of separation of variables [4, Section 2.3]. For the 1D diffusion problem shown in Fig. 1, the solution of the 2nd FICK equation yields for the 1D case the following trigonometric series [4] to determine the local concentration c resp. the relative local concentration c/c_∞ with the saturation concentration c_∞:

$$\frac{c}{c_\infty} = 1 - \frac{4}{\pi} \sum_{i=0}^{\infty} \frac{(-1)^i}{2i+1} \exp\left(-D(2i+1)^2 \pi^2 t / d_\mathrm{k}^2\right) \cos \frac{(2i+1)\pi x}{d_\mathrm{k}} \ . \tag{6}$$

The parameter identification for the calculation of moisture diffusion is carried out with test data from gravimetric investigations on adhesive substance samples from [3] (see figure 2) and inversely optimized with LS-OPT. Thus, the water absorption can be determined exactly for a certain adhesive and is used in a damage model as a damage-driving hygric parameter for the service life prediction in any adhesive compound sample or component. In the case of hygrothermomechanical loading, there is a so-called three-field problem with the three fields: hygric, thermal and mechanical. However, LS-DYNA does not have a diffusion solver, so the three-field problem is reduced to a two-field problem. This simplifying assumption is permissible, since the temperature field, in contrast to the other field, has adjusted itself quickly. The implementation into LS-DYNA is carried out by exploiting the thermo-mechanical solver, due to the analogous field problem. The finite element model is created with the commercial programme LS-PrePost. The round substance sample has a diameter of $d = 40$ mm and a thickness of $d_\mathrm{k} = 0.5$ mm. The diffusion problem was calculated under constant boundary conditions. Let the sample be completely dry at the beginning, because the initial concentration is zero. The diffusion problem is treated as a heat conduction problem in the nominal finite element program LS-DYNA. Therefore, the mass increase due to water can be read out as thermal energy. Since in the present calculation only the temperature brings an energy term with it, the total energy can be read out accordingly in LS-PrePost.

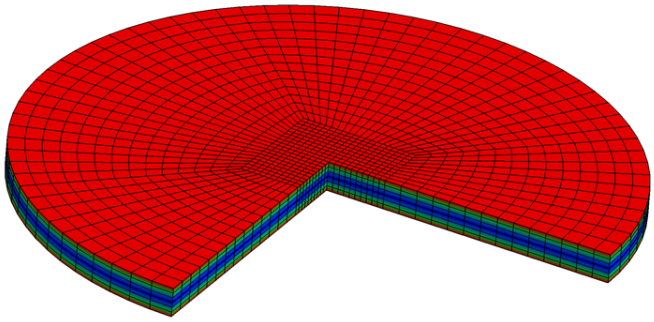

Fig. 2: Round adhesive substance sample as FE model during 1D diffusion calculation with LS-DYNA

To verify the parameters, the solution for the relative concentration equation (6) with the approaches for the water mass increase and the diffusion velocity equation (5) are presented together with the measured data at the different climatic conditions.

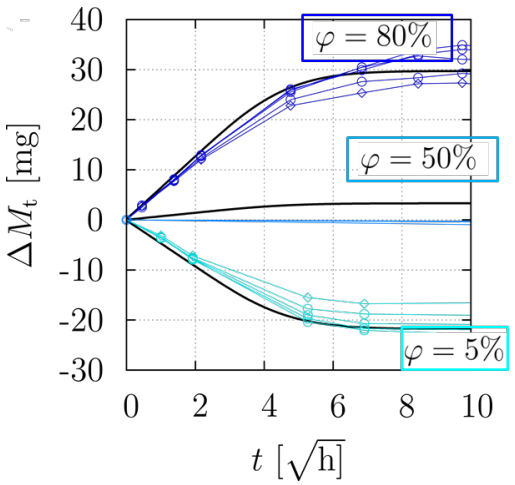

Fig. 3: Measurement data and simulation (black) of the water mass increase of the adhesive substance samples at room temperature and different relative humidities

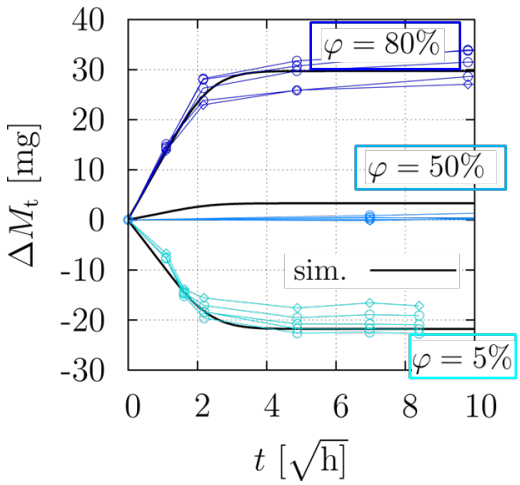

Fig. 4: Measurement data and simulation (black) of the water mass increase of the adhesive substance samples at room 70 °C and different relative humidities

The verification of the diffusion model and the parameters is carried out on the test data from the selected adhesive for the different climatic conditions. Figure 3 and figure 4 shows an overall good agreement between simulation (see figure 2) and test data [5] from an engineering point of view.

2 Finite viscoelastic model

The thick adhesive layer is modeled by a three-dimensional finite viscoelasticity model according [8] and [15].

$$\tilde{\mathbf{T}}(t) = \tilde{\mathbf{T}}^{\text{eq}}_{\text{vol}}(t) + \tilde{\mathbf{T}}^{\text{eq}}_{\text{iso}}(t) + \tilde{\mathbf{T}}^{\text{ov}}_{\text{iso}}(t, T, c) \tag{7}$$

$$\tilde{\mathbf{T}}^{\text{eq}}_{\text{vol}}(t) = J\frac{\mathrm{d}U}{\mathrm{d}J}\mathbf{C}^{-1}(t) \tag{8}$$

$$\tilde{\mathbf{T}}^{\text{eq}}_{\text{iso}}(t) = J^{-2/3}\left((c_{10} + c_{01}I_{\bar{\mathbf{C}}})\mathbf{1} - c_{01}\bar{\mathbf{C}} - \frac{1}{3}(c_{10}I_{\bar{\mathbf{C}}} + 2c_{01}I\!I_{\bar{\mathbf{C}}})\bar{\mathbf{C}}^{-1}\right) \tag{9}$$

$$\tilde{\mathbf{T}}^{\text{ov}}_{\text{iso}}(t, T, c) = \int_{-\infty}^{t}\left(\gamma_\infty + \sum_{i=1}^{N}\gamma_i \exp\left(-\frac{t-s}{a_{\text{res}}(T,c)\tau_i}\right)\right)\frac{\mathrm{d}}{\mathrm{d}s}\left(2\left(\frac{\mathrm{d}\bar{\mathbf{C}}}{\mathrm{d}\mathbf{C}}\right)^{\top} : \frac{\mathrm{d}\bar{W}^0(I_{\bar{\mathbf{C}}}, I\!I_{\bar{\mathbf{C}}})}{\mathrm{d}\bar{\mathbf{C}}}\right)\mathrm{d}s \tag{10}$$

The generalised MAXWELL model in the isochoric part consisting of N parallel chains with springs and dashpots is used with relaxation times $\tau_1, \ldots, \tau_{\text{N}}$ and stiffnesses $\gamma_1, \ldots, \gamma_{\text{N}}$.

The MAXWELL model can additionally account for the reversible effects of temperature and concentration using this approach.

The influences of temperature and humidity lead to the time-temperature and the time-concentration-shift model, respectively. The joint influences of temperature and humidity on the viscoelastic properties are taken into account by linking these two principles to the time-temperature-concentration-displacement concept, which results from the assumption of thermo- and hygrorheological simplicity.

By a_{res} is denoted the resulting displacement function, which is obtained from the individual displacements due to temperature and concentration as follows:

$$a_{\text{res}}(T, c) = a_{\text{T}}(T)\, a_{\text{c}}(c)\ . \tag{11}$$

For the time-temperature shift function a_{T}, above the temperature T_{aT}, the WILLIAMS, LANDEL, and FERRY (WLF) WILLIAMS and below temperature T_{aT} an ARRHENIUS-type approach is used, since these two approaches have proven successful for many polymers, cf. [14]:

$$\log_{10} a_{\text{T}} = \log a_{\text{T}} = \begin{cases} \dfrac{-p_{\text{aT1}}(T - T_{\text{aT}})}{p_{\text{aT2}} + T - T_{\text{aT}}} & , \quad T > T_{\text{aT}} \\ p_{\text{aT3}}\left(\dfrac{1}{T} - \dfrac{1}{\hat{T}_{\text{aT}}}\right) & , \quad T \leq T_{\text{aT}} \end{cases} . \tag{12}$$

In the WLF-ARRHENIUS approach, three parameters p_{aT1}, p_{aT2}, and p_{aT3} occur. The value T_{aT} is a reference temperature where between the two approaches. The parameter $\hat{T}_{\mathrm{aT}}$ is usually identical to the reference temperature: $\hat{T}_{\mathrm{aT}} = T_{\mathrm{aT}}$. It is only needed when the WLF approach is with already identified parameters with respect to a different reference temperature $\hat{T}_{\mathrm{aT}} \neq T_{\mathrm{aT}}$ is to be shifted. For the time-concentration shift function a_{c}, the expression analogous to the WLF equation is used in [10] and [11] to represent the influence of moisture on the viscoelastic properties for a polyester resin and epoxy resin, respectively.

$$\log a_{\mathrm{c}} = \begin{cases} \dfrac{-p_{\mathrm{ac1}}(c - c_{\mathrm{ac}})}{p_{\mathrm{ac2}} + c - c_{\mathrm{ac}}} & , \quad c > c_{\mathrm{ac}} \\ p_{\mathrm{ac3}} \left(\dfrac{1}{c} - \dfrac{1}{\hat{c}_{\mathrm{ac}}} \right) & , \quad c \leq c_{\mathrm{ac}} \end{cases} . \tag{13}$$

With this approach, reversible aging in the adhesive can be considered and simulated. for irreversible aging, a damage model is developed in the following. The sample saturation is followed by the creep tests. With the experimental results, a damage model [5] is conditioned and extended. The basic approach of the creep and hygro-damage differential equation (see equation 15) entails a large number of parameters that have to be identified.

3 Damage model and Simulation

The damage differential equation must be specified in order to represent the long-term failure due to mechanical, hygric and thermal loading. to be able to do so. Here it is assumed that the total damage evolution $\dot{D}_{\mathrm{ca}}$ consists of a creep $\dot{D}_{\mathrm{c}}$ and aging $\dot{D}_{\mathrm{a}}$ component. additively:

$$\dot{D}_{\mathrm{ca}}(\sigma, c, T) = \dot{D}_{\mathrm{c}}(\sigma, T) + \dot{D}_{\mathrm{a}}(c, T) \tag{14}$$

$$\dot{D}_{\mathrm{ca}} = \frac{1}{c_0} \left(\frac{\sigma}{\sigma_{\mathrm{ref}}(1 - D_{\mathrm{ca}})} \right)^{n} \exp\left(p_{\mathrm{c}} \left(\frac{1}{T_{\mathrm{refc}}} - \frac{1}{T} \right) \right) + B_{\mathrm{a}}(1 - D_{\mathrm{ca}}) \left(\frac{c}{c_{\infty,\mathrm{ref}}} \right)^{l} \exp\left(p_{\mathrm{a}} \left(\frac{1}{T_{\mathrm{refa}}} - \frac{1}{T} \right) \right) \tag{15}$$

σ_{ref} and n are two parameters to be identified. The quantity c_0 merely establishes consistent units. The creep damage evolution is extended with an ARRHENIUS-type approach to account for the influence of the temperature on the creep damage, cf. [12] and e.g. [13]: B_{ref}, l and $c_{\infty,\mathrm{ref}}$ are parameters to be identified for isothermal hygrodamage. The temperature influence is determined by identifying of the parameter p_{a}. The reference temperature T_{refa}, as well as T_{refc} in the creep damage approach, becomes is set to $T_{\mathrm{refa}} = T$ for the isothermal case and is otherwise fixed in the order of magnitude of the operating temperature of the adhesive layer. Eq. (15) shows that the influence of the temperature on the damage development is formally considered identically for both creep and hygro-damage. Both equations are potential approaches. The difference is in the exponent of the term $(1 - D_{\mathrm{ca}})$: In the creep damage evolution, it occurs as $(1 - D_{\mathrm{ca}})^{-n}$ and in the hygrodamage evolution as $(1 - D_{\mathrm{ca}})$, i.e., without exponents. For the calculation of the service life prediction, the damage differential equation, which takes into account the temperature, the moisture concentration and the mechanical load in a *usermat, is implemented to calculate the service life satisfactorily with LS-DYNA.

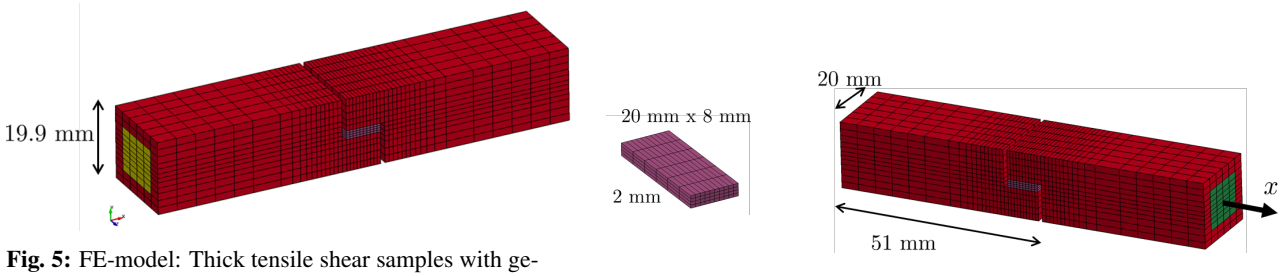

Fig. 5: FE-model: Thick tensile shear samples with geometric dimensions and adhesive layer in detail

Figure 5 shows the thick tensile shear specimen consisting of two steel parts and an adhesive layer. This joint specimen is used for the following FE calculations. The diffusion behaviour of the adhesive is already known from the previous investigations. Subsequently, the mechanical and temperature-dependent behaviour is investigated.

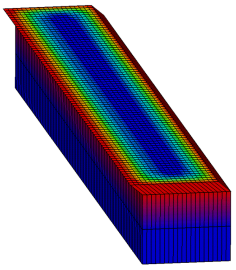

Fig. 6: Evaluation of the FE calculation with LS-PrePost: the damage progression is marked in colour

Figure 6 shows the thick tensile shear specimen from figure 5 in the LS-PrePost. The coloured representation from blue to red shows an increase in damage from the core to the edge. Blue elements have a damage value of zero and red elements are at approximately $D_{\mathrm{ca}} = 0.6$. The sample continues to be subjected to hygro-thermo-mechanical loading in the simulation until the first elements reach the damage value 1, fail and thus the adhesive layer fails. Depending on the influencing variables: mechanical load, temperature and concentration, different fracture times appear, as they are shown in the following calculations with measured data in the diagram.

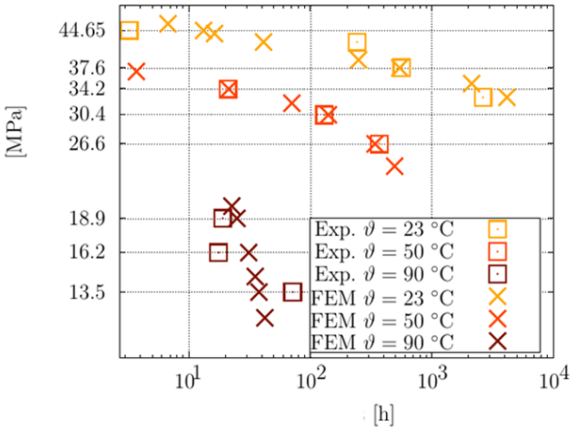

Fig. 7: Schematic process of moisture diffusion into the adhesive / Image based on [7]

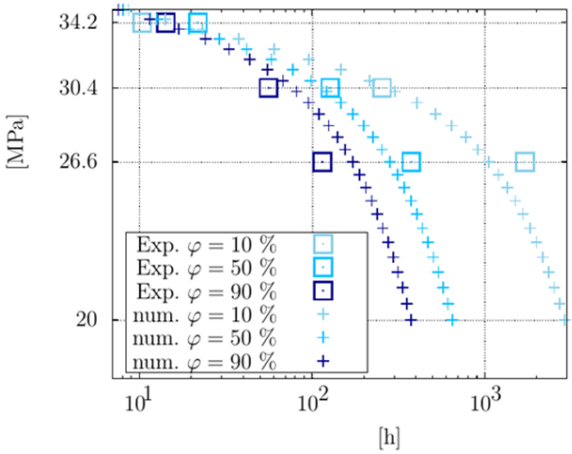

Fig. 8: Schematic process of moisture diffusion into the adhesive / Image based on [7]

The results show that the developed model is able to represent the long-term behavior and, in particular, the failure point due to hygro-thermo-mechanical creep stress very well.

The investigation of the diffusion of water in a PU adhesive and the development of an identification method for the diffusion coefficient and the saturation concentration, which is verified with FE calculation, is completed. The methods and investigations are focused on the evaluation of the measurement data. The results form the basis for further validations. Successful was the determination of the htm process parameters and their influence on the long-term behaviour and ageing resistance on the basis of the dimensioned basic tests. Based on the parameters, the test data can be reproduced with the htm damage approach and the FE calculations can be validated and improved. The aim is to predict the long-term behaviour of the bonded joint and to guarantee the ageing resistance for the operating period.

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