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RELATIONSHIP BETWEEN THE TENSILE AND STRESS RELAXATION BEHAVIOUR OF POLYPROPYLENE

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Abstract

The mechanical response of real polymers are basically of nonlinear character, therefore their behaviour patterns do not meet the idealized (linear) ones, especially in case of larger loads. Nevertheless, the linear viscoelasticity is often used successfully for describing the real behaviour in case of small or moderate loads. In this paper a way is sought and followed to estimate the real tensile load response by using the linear viscoelastic response given to the real relaxation stimulus and its relationship to the linear viscoelastic tensile load-strain curve and also by using some nonlinear transformations. Experiments were performed on PP as a test material and the stress relaxation behaviour, as well as the linear elastic and linear viscoelastic approximation of the tensile load-time curve were analysed. In order to demonstrate the applicability of this idea and to perform the numerical calculations a flexible function with three parameters was chosen to realize the nonlinear behaviour.

Keywords: stress relaxation, tensile test, polypropylene.

1. Introduction

The mechanical behaviour of polymeric materials is described as viscoelastic what means that the viscous type deformation is added to the elastic one and the latter may be of instantaneous or delayed type therefore the resultant deformation is time dependent [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. Consequently, depending on the temperature and other environmental parameters, some permanent deformation of viscous type can be experienced under relatively small load, as well. On the other hand, in case of constant load the so-called relaxation and creep processes can be observed. Nonlinear behaviour can make this picture even more complicated.

A number of theoretical and experimental studies and books have mainly dealt with the investigation of the relaxation phenomena and in some cases with the relationships between the results of different tests in order to estimate or predict the short and long term behaviour of polymers under different circumstances [2, 4, 5, 8, 9]. The stress relaxation behaviour of thermoplastics has been studied in several theoretical and experimental investigations such as those by URZUMTSEV and MAKSIMOV [4], RETTING [8], WORTMANN and SHULTZ [12], ANDREASSEN [11], AKINAY et. al. [13], as well as BAEURLE et. al. [14]. For describing the long term relaxation modulus or the components of the complex elastic modulus for small strains at different temperatures they used linear viscoelastic models and time-temperature correspondence [8, 11, 12, 13, 14] or power law equations and/or the so-called stretched exponential function forms [11, 14].

In accordance with the usual loading modes the most frequently used quasi-static mechanical tests are the tensile, the creep, and the stress relaxation ones. Among them the tensile test lasts for the shortest time and provides an increasing load up to the damage or fracture of the specimen. During the tensile test the specimen, so to say, lives its life to the very end in an accelerated way, consequently this test may give a lot of information about the material and its mechanical behaviour for various loading modes. Therefore, it may be expected that from a tensile test the behaviour for the medium or long term relaxation or creep and *vice versa* can be predicted. In [8] RETTING summarized his method and experimental results on the estimation of the real stress relaxation function as the product of the linear response and a strain dependent factor. The tensile load curve is estimated as the time integral of the stress relaxation curve in accordance with the linear viscoelastic theory.

This paper makes an attempt to investigate the relationships between the results of the tensile and the stress relaxation tests of polypropylene specimens, and to find a way to estimate the former from the latter based on the measurements and the theory of the linear viscoelasticity applied for real stimuli.

2. Theoretical Analysis Based on the Linear Viscoleasticity

2.1. A General Description of the Mechanical Tests

Fig. 1 shows the general scheme of mechanical tests where S is the specimen as a black box system, X is the input stimulus and Y is the response of the specimen. The two latter are time-functions. If S is an operator characterizing the material,

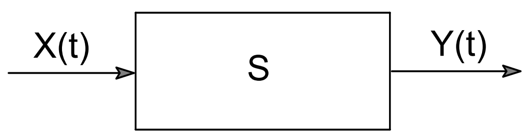


Fig. 1. General scheme of mechanical tests

that is the specimen, the response can be obtained as follows:

$$Y(t) = S[X](t) \tag{1}$$

The specimen, hence also operator S, can be characterized by all the possible pairs of X and Y. In case of mechanical tests the stimuli and responses are mechanical quantities such as the elongation of the strain and the load of the stress.

2.2. Basic Relationships in Case of Linear Viscoelasticity

In practice, operator S in *Eq.* (1) is often considered continuous and linear. In case of linear viscoelastic behaviour the so called Boltzmann's superposition principle is valid which leads to the following convolution integral [1, 5, 6, 8]:

$$Y(t) = \int_0^t W\,(t-u)\,\mathrm{d}X(u) \tag{2}$$

where W is a characteristic function of the material. The usual inputs are the strain (ε) or the stress (σ)

$$\varepsilon(t) = \frac{\Delta l(t)}{l_o} \tag{3}$$

$$\sigma(t) = \frac{F(t)}{A_o} \tag{4}$$

where Δl, l_0, A_0, and F are the elongation, the initial length, and the initial cross section area of the specimen tested, as well as the load, respectively. In case of these inputs the form of *Eq.* (2) is as follows [1, 4, 6, 8]:

$$X = \varepsilon \qquad \sigma(t) = \int_0^t E(t-u)\mathrm{d}\varepsilon(u) \tag{5}$$

$$X = \sigma \qquad \varepsilon(t) = \int_0^t J(t-u)\mathrm{d}\sigma(u) \tag{6}$$

where $E(t)$ is the relaxation modulus and $J(t)$ is the creep compliance. $E(t)$ and $J(t)$ are not independent. The relationship between them is given by [1]:

$$t = \int_0^t E(t-u)J(u)\mathrm{d}u \tag{7}$$

The most important features of the linear viscoelastic behaviour concerning the time range can appear as follows:

- Relaxation modulus $E(t)$ and creep compliance $J(t)$ do not depend on the load level (ε_0 and σ_0, respectively).
- Any two different responses of a linear viscoelastic material given to stimuli of different types can be calculated from each other.

However, the real polymers are basically of nonlinear behaviour, therefore the behaviour patterns described above do not meet the real one of materials, especially in case of larger loads. Nevertheless, the linear viscoelasticity often used successfully for describing the real behaviour in case of small or moderate loads.

2.2.1. Ideal Deformation Stimulus

The time function of the ideal strain stimulus with a load level ε_0 is as follows (*Fig. 2*(a)):

$$\varepsilon_1(t) = \varepsilon_o \cdot 1(t) \tag{8}$$

where $1(t)$ is the unit step function. The response, $\sigma_1(t)$, of a linear viscoelastic (LVE) polymeric material characterized by relaxation modulus $E(t)$ to stimulus $\varepsilon_1(t)$ can be calculated by *Eq.* (5) after a partial integration:

$$\sigma_1(t) = \int_0^t \varepsilon_1(t-z)\mathrm{d}E(z) = \varepsilon_0 \int_0^t \mathrm{d}E(z) = \varepsilon_0\left(E(t) - E(0)\right) = \varepsilon_0 E(t) \tag{9}$$

because $E(0) = 0$. *Eq.* (9) represents the result of a stress relaxation test.

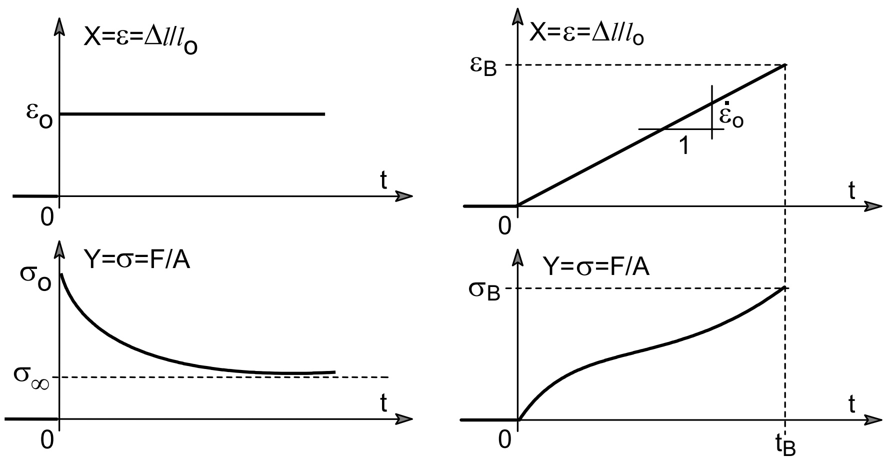


Fig. 2. Stimuli (X) and responses (Y) of linear viscoelastic materials in case of stress relaxation (a) and tensile tests (b)

The strain stimulus applied to standard tensile tests has got a constant rate ($\dot{\varepsilon}_o$) and is of speed step type (*Fig. 2*(b)):

$$\varepsilon_2(t) = \dot{\varepsilon}_o t \cdot 1(t) \tag{10}$$

The stress response, $\sigma_2(t)$, can be obtained by substituting $\varepsilon_2(t)$ into convolution integral (5) and after a partial integration we get:

$$\sigma_2(t) = \int_0^t \frac{\mathrm{d}E}{\mathrm{d}z}\varepsilon_2(z)\mathrm{d}z = \dot{\varepsilon}_0 \int_0^t \frac{\mathrm{d}E}{\mathrm{d}z} z\mathrm{d}z = \dot{\varepsilon}_0 \int_0^t E(u)\mathrm{d}u \tag{11}$$

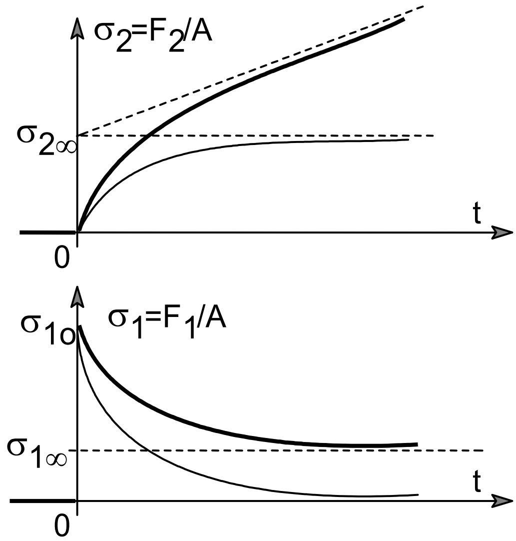


Fig. 3. Tensile load-strain curve types of LVE materials (top) as the integral of the stress-relaxation curves (bottom) according to *Eq.* (12) solid thick lines: $\sigma_{1\infty} \neq 0$; solid thin lines: $\sigma_{1\infty} = 0$; dotted lines: asymptotes

It can be seen that the left hand expression of *Eq.* (11) is proportional to the integral of *Eq.* (9):

$$\sigma_2(t) = \frac{\dot{\varepsilon}_0}{\varepsilon_0} \int_0^t \sigma_1(u)\mathrm{d}u \tag{12}$$

The measured load-strain curve is given in a parametric form, with value pairs $(\varepsilon_2(t),\sigma_2(t))$, however, taking into account that according to *Eq.* (10) the time (t) and the strain (ε) are proportional to each other for $t > 0$, therefore the usual form of the load-strain curve, $\sigma(t)$, can be obtained with a simple change of variables ($\varepsilon = \varepsilon_2$):

$$\sigma(\varepsilon) = \sigma_2\left(\frac{\varepsilon}{\dot{\varepsilon}_0}\right) \tag{13}$$

It can be established that in case of LVE materials the load-strain curve has got a form similar to the integral of the stress relaxation curve belonging to ideal stimulus and differs from that only in a scale constant. However, also the relaxation curves of real materials are created as a sum of a constant and a monotonously decreasing function (the bottom diagram in *Fig. 3*), consequently the integral of them is a monotonously increasing function starting from zero and asymptotically tending to a straight line with non-negative slope (*Fig. 3*).

The load-strain curves of real materials can be different from the ideal ones and the shape described above occurs only in a few cases. In general, the initial sections of the real load-strain curves, which are concave from below and significantly larger than what is approximated by the initial tangent, meet the requirements of the description above. The difference between the behaviours of the linear viscoelastic and real materials is observable especially well if the derivative of the tensile load-strain curve, the so-called tangent modulus curve, is compared with the measured relaxation curve. In case of linear viscoelasticity the tangent modulus curve is:

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\varepsilon}(\varepsilon) = \frac{1}{\dot{\varepsilon}_0}\frac{\mathrm{d}\sigma_2}{\mathrm{d}t}\left(\frac{\varepsilon}{\dot{\varepsilon}_0}\right) = \frac{1}{\dot{\varepsilon}_0}\frac{\dot{\varepsilon}_0}{\varepsilon_0}\sigma_1\left(\frac{\varepsilon}{\dot{\varepsilon}_0}\right) = E\left(\frac{\varepsilon}{\dot{\varepsilon}_0}\right) \tag{14}$$

which differs from the relaxation curve only in a scale constant.

2.2.2. *Real Deformation Stimulus*

Let $\sigma_i(t)$ (i=1, 2) be the stress response given by a polymeric specimen to deformation stimulus $\varepsilon_i(t)$. In case of tensile tests, however, usually force (F)-elongation (Δl) curves are recorded instead of stress-strain $\sigma(t) - \varepsilon(t)$ curves. The relationships between them are according to *Eqs.* (3) and (4) (i=1, 2):

$$F_i(t) = A_o\sigma_i(t) \tag{15}$$

$$\Delta l_i(t) = l_o\varepsilon_i(t) \tag{16}$$

where A_0 and l_0 are the cross section area and the gauge length of the unloaded specimen, respectively.

Here $F_1(t)$ is the measured relaxation curve as a response given to the following stimulus (*Fig. 4*):

$$\Delta l_1(t) = l_o\varepsilon_1(t) = l_o\dot{\varepsilon}_o t\,(1(t) - 1(t - t_o)) + l_o\varepsilon_o 1(t - t_o) \tag{17}$$

while $F_2(t)$ is the tensile force-time component of the measured tensile load-strain curve as a response given to the stimulus

$$\Delta l_2(t) = l_o\varepsilon_2(t) = l_o\dot{\varepsilon}_o t 1(t) \tag{18}$$

Taking into account that (*Fig. 4*) is:

$$\varepsilon_o = \dot{\varepsilon}_o t_o \tag{19}$$

the stimulus in *Eq.* (17) can be produced as the difference of stimuli according to (20):

$$\begin{aligned}\Delta l_1(t) = l_o\varepsilon_1(t) = l_o\dot{\varepsilon}_o t 1(t) - l_o\dot{\varepsilon}_o(t - t_o)1(t - t_o) = \\ = l_o\left(\varepsilon_2(t) - \varepsilon_2(t - t_o)\right)\end{aligned} \tag{20}$$

The convolution integral in *Eq.* (5) is a linear operator therefore the response given to a sum of stimuli can be produced as a sum of partial responses given to partial stimuli. Consequently, in case of LVE material behaviour, relaxation curve $F_1(t)$ as a response given to real stimulus (18), can be given with the aid of tensile load-time curve $F_2(t)$, as well (*Fig. 5*):

$$F_1(t) = F_2(t) - F_2(t - t_o) \tag{21}$$

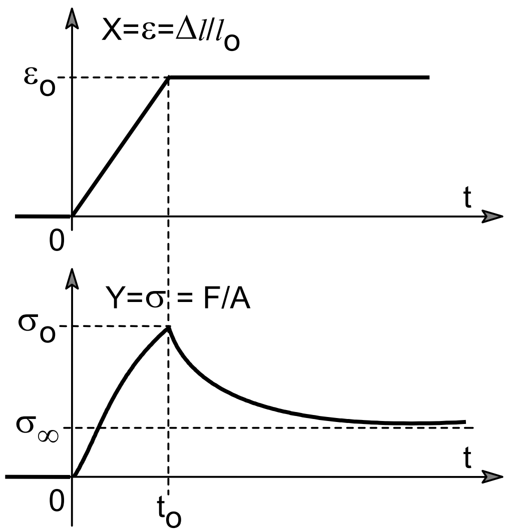


Fig. 4. Stress relaxation curve as a response to real deformation stimulus

2.3. *Estimating the Real Behaviour of Polymer Materials Using LVE Models*

On the basis of *Eq.* (21), if $F_1(t)$ is the response to the real relaxation stimulus given by a real material, that is the measured relaxation curve, the LVE estimation of the real tensile load-time curve of the given material can be calculated (*Fig. 5*):

$$F_{12}(t) = F_1(t) + F_{12}(t - t_0) \tag{22}$$

where $F_{12}(t)$ denotes the estimation of $F_2(t)$ calculated from $F_1(t)$ according to the following scheme:

$$F_1(t) \to F_{12}(t) \approx F_2(t) \tag{23}$$

For example, using sampling time points $t = it_0$ (i=1, 2, ...) *Eq.* (22) can be rewritten:

$$F_{12}(it_o) = F_1(it_o) + F_{12}\left((i-1)t_o\right) \tag{24}$$

Expanding the recursive relationship (24) we get a summing formula:

$$\begin{aligned}
i &= 1 \qquad F_{12}(t_o) = F_1(t_o) + F_{12}(0) = F_1(t_o) \\
i &= 2 \qquad F_{12}(2t_o) = F_1(2t_o) + F_{12}(t_o) = F_1(t_o) + F_1(2t_o) \\
i &= n \qquad F_{12}(nt_o) = F_1(t_o) + F_1(2t_o) + ... + F_1(nt_o)
\end{aligned} \tag{25}$$

where it was used that if $\varepsilon_2(-0) = 0$, then $F_2(0) = F_{12}(0) = 0$ and this is valid for the estimation, as well.

According to *Eq.* (25) the value of estimation F_{12} at a given time (nt_0) can be obtained by summing the values of the measured stress relaxation curve (F) measured at the sampling times (it_0)(*Fig. 6*). Decreasing t_0 the real stimulus tends to the ideal one and the sum according to *Eq.* (25) tends to an integral similar to *Eq.* (12).

Eq. (25) includes *Eq.* (12) essentially because if $t_0 \to 0$ the sum in *Eq.* (25) tends to the integral according to *Eq.* (12):

$$F_{12}(nt_o) = \sum_{i=1}^{n} F_1(it_o) = \frac{1}{t_o} \sum_{i=1}^{n} F_1(it_o)t_o \tag{26}$$

Fixing the strain rate $(\dot{\varepsilon}_o)$ for the tensile test yields:

$$\frac{1}{t_o} = \frac{\dot{\varepsilon}_o}{\varepsilon_o} \tag{27}$$

while for the relaxation tests the strain rate $(\dot{\varepsilon}_{oR})$ is increased; consequently the uploading time (t_0) is decreased over every bound:

$$t_o = \Delta t = \frac{\varepsilon_o}{\dot{\varepsilon}_{oR}} \underset{\dot{\varepsilon}_{oR} \to \infty}{\longrightarrow} 0 \tag{28}$$

and all that results in the integral mentioned above:

$$F_{12}(nt_o) = \frac{\dot{\varepsilon}_o}{\varepsilon_o} \sum_{i=1}^{n} F_1(i\Delta t)\Delta t \underset{\substack{n \to \infty \\ \Delta t \to 0}}{\longrightarrow} \frac{\dot{\varepsilon}_o}{\varepsilon_o} \int_0^t F_1(u)\mathrm{d}u \tag{29}$$

3. Experimental

3.1. Materials and Equipment

Isotactic polypropylene homopolymer (Tipplen H 543 F from TVK, Hungary) having a Melt Flow Index (MFI, 2.16 kg/230 °C) of 4 g/10 min was used for the tests. Dumbbell specimens were injection molded according to ISO 294-2 Standard on an

Arburg Allrounder 320 C 600-250 injection molding machine of specimen length 148 mm, width 10 mm and thickness 4 mm. Uniaxial tensile stress relaxation tests were performed on a Zwick Z005 tensile testing machine. The tensile force was measured by a 5 kN nominal capacity standard load cell.

3.2. Experimental Procedure

Tensile tests were performed on 5 test specimens with crosshead speed of 5 mm/min at room temperature. For the relaxation tests specimens were stretched up to 0.08, 0.16, 0.24, 0.50, 0.75, 1.00, 1.50, 2.00, 3.00, 5.00 and 7.00 % engineering strains (ε_0) with crosshead speed (v) of 5 mm/min. No necking of specimens was observed in the relaxation tests. Every test was performed on a new specimen. The engineering yield strain was calculated according to *Eq.* (16) from the crosshead displacement as $\varepsilon_o = \Delta l / l_o$ where l_0 is the gauge length at time 0 and $l_0 = 110$ mm. On the basis of these considerations the strain rate was $\dot{\varepsilon}_o = v / l_o = 0.0455/\text{min} = 4.55\%$.

3.3. Results of Tensile Tests

Fig. 7(a) shows the average of 5 measured tensile load-time curves up to the neck forming while *Fig. 7*(b) shows the initial part of the curve including the local maximum indicating the beginning of the yielding.

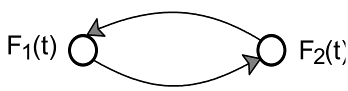


Fig. 5. Calculability of LVE characteristics. The relaxation curve $F_1(t)$, can be calculated from the load-time curve $F_2(t)$ and vice versa.

In *Fig. 7*(b) the significant nonlinearity of the initial part of the curve can be well observed. On the basis of Section 2.2.2 it is obvious that at most the initial concave part up to the peak can be approximated with LVE behaviour.

3.4. Results of Stress Relaxation Tests

The results of 11 stress relaxation tests are shown in *Fig. 8*(a). Each curve starts from the real initial point on the tensile load-time curve determined by the real stimulus at the given strain level ($t_o = \varepsilon_o / \dot{\varepsilon}_o$). For the sake of comparison two curves ($\varepsilon_0 = 5$and 7%) start after the peak of the tensile load-time curve. Their shape shows a steeper decrease in comparison to that of the others.

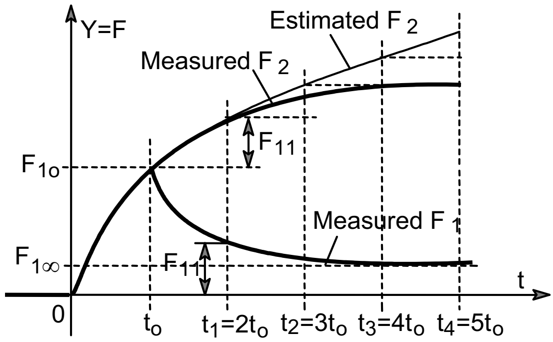


Fig. 6. Estimation of the tensile characteristic (F_2) from the measured stress relaxation curve (F_1)

In order to see better the variation of the curves measured at smaller strain levels some of the measurements are plotted in enlarged time scale in *Fig. 8*(b).

By depicting the values of the relaxation curves measured at $t = 0$ s and $t = 300$ s it is visible that their variations with the strain level (ε_0) are similar to each other (*Fig. 9*). Of course the $F_1(0)$ and ε values constitute the measured tensile load-strain curve.

This observation is confirmed by the high linear correlation between these force values in *Fig. 10*.

According to this linear trend the relaxation force at 300 s is approximately 73.3 % of the initial force independently of the strain level in the range $0 \leq \varepsilon_0 < 4\%$, consequently these relaxation processes of the PP tested are similar in a way because they take place at about the same rate.

3.5. Estimating the Tensile Characteristics with the Measured Relaxation Curve Using LVE Model

Based on the measured relaxation curves the LVE estimations of the tensile load-time curves were calculated using *Eq.* (22).

In *Figs 11* and *12* the measured tensile load-time curve (F_2) and its initial tangent as a linear elastic (LE) estimation as well as its linear viscoelastic (LVE) estimations, using relaxation curves (F_1) measured at a small (0.08 %) and a high (3 %) strain level, are depicted.

The LVE estimation in *Fig. 11* starts from a point close to zero and fits the tensile load-time curve along a section of about 4 s from the starting point. But the initial tangent that is the LE estimation fits only in a range of 1 s. By calculating with the on-set part of the relaxation curve as well, the fitting range of the LVE estimation is five times larger than that of the LE estimation.

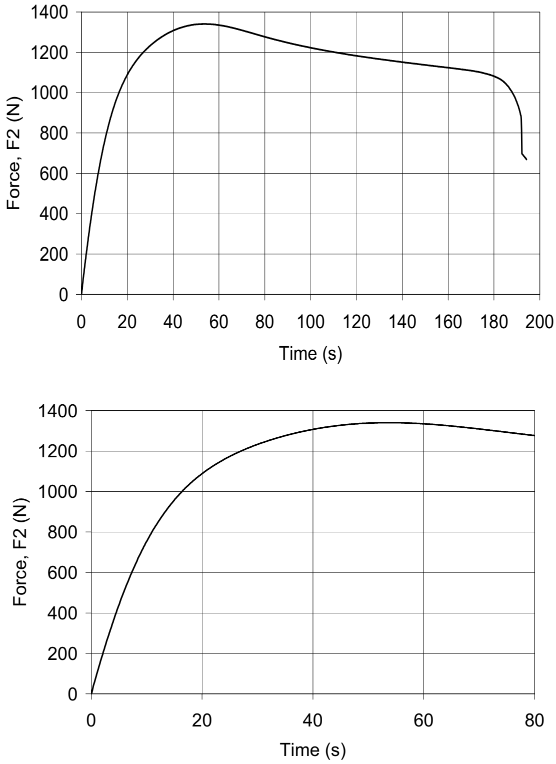


Fig. 7. Averaged tensile load-time curve (a): up to neck forming
(b): at the beginning of the test

The situation is different in *Fig. 12* where the strain level is high. Here there is no fitting except for the starting point, consequently the estimation should be modified in order to achieve an acceptable approximation.

3.6. A Measure of the Linear Elasticity and Linear Viscoelasticity

It can be seen for example in *Fig. 11* that the initial tangent (F_{2LE}) representing the linear elastic (LE) behaviour fits the measured load-time curve (F_2) only in the vicinity of the zero point and otherwise it goes above that. Mathematically it means that for $t \geq 0$:

$$F_{2LE}(t) \geq F_2(t) \tag{30}$$

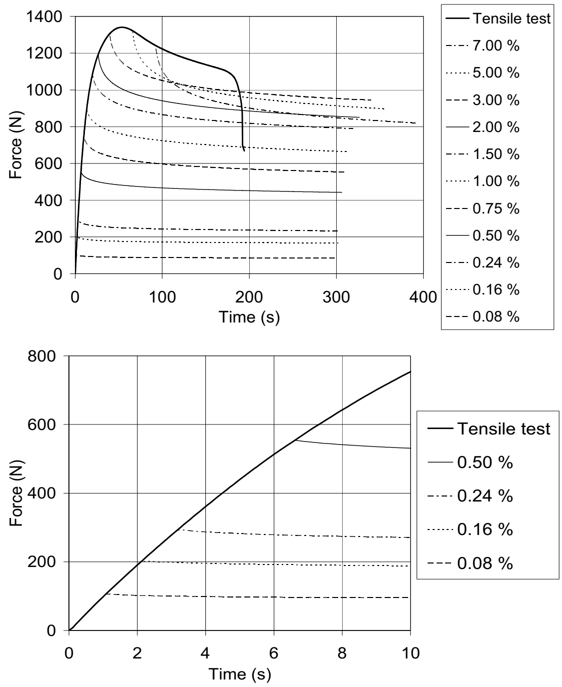


Fig. 8. Stress relaxation (a): at strain levels ($\varepsilon = \varepsilon_0$) up to 7 % and (b): at strain levels up to 0.5 %

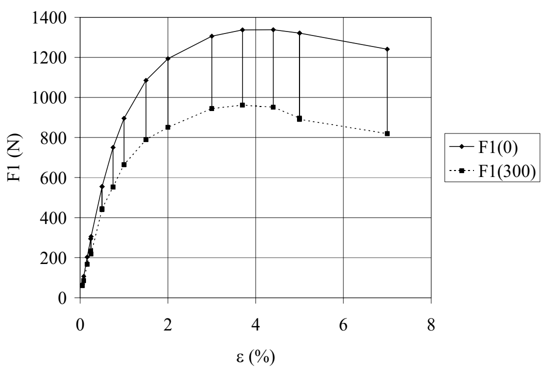


Fig. 9. Decrease of force (F_1) measured at 300 s and different strain levels ($\varepsilon = \varepsilon_0$) as a result of the relaxation

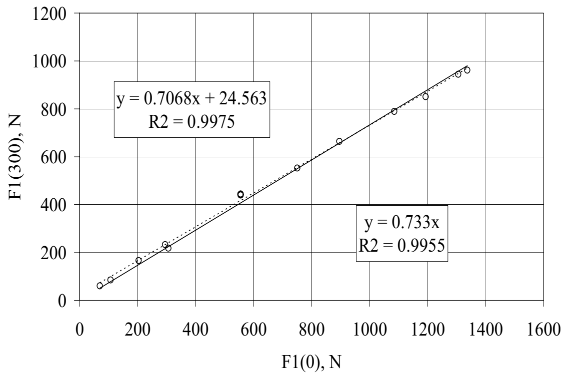


Fig. 10. Relationship between the initial load $F_1(0)$ and relaxation force $F_1(300)$ measured at $F_1(300)$s

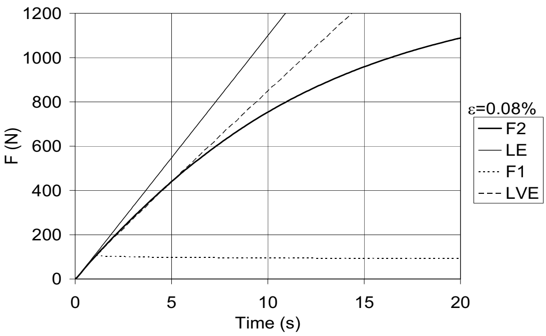


Fig. 11. The measured tensile load-time curve (F_2), its linear elastic (LE), and linear viscoelastic (LVE) estimations using the relaxation curve (F_1) measured at a small strain level ($\varepsilon = \varepsilon_0$)

On the basis of *Eq.* (30) a kind of measure of the linear elasticity (MLE) may be defined as follows:

$$0 < \varphi(t) = \frac{F_2(t)}{F_{2LE}(t)} \leq 1 \qquad (31)$$

This measure is equal to 1 where $F_2(t)$ is identical with F_{2LE}, otherwise F_2 represents nonlinear elastic (NLE) behaviour which is close to the LE if $\varphi \approx 1$.

Fig. 13 illustrates the behaviour of the PP material tested. The MLE drops very steeply and only the very initial part of the load-time curve below 1 s can be considered as of LE behaviour. The MLE is larger than 52% up to 20 s.

For the linear viscoelastic (LVE) estimation (F_{2LVE}) an inequality similar to

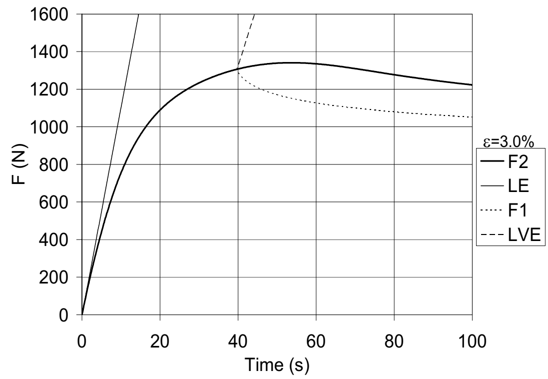


Fig. 12. The measured tensile load-time curve (F_2), its linear elastic (LE), and linear viscoelastic (LVE) estimations using the relaxation curve (F_1) measured at a high strain level ($\varepsilon = \varepsilon_0$)

Inequality (30) can be created as

$$F_{2LE}(t) \geq F_{2LVE}(t) \geq F_2(t) \tag{32}$$

Hence, a measure of the linear viscoelasticity (MLVE) may be defined on the basis of Inequality (32):

$$0 < \psi(t) = \frac{F_2(t)}{F_{2LVE}(t)} \leq 1 \tag{33}$$

In *Fig. 14* one can see the variation of the MLVE values for some LVE estimations belonging to different strain levels. The initial point corresponds to t_0 for each curve. For the LVE estimation with 0.05 % strain level the range of the LVE behaviour is about 6 s, while in case of 3 % it is less than 1 s. For the latter the MLVE is larger than 60 % up to 20 s.

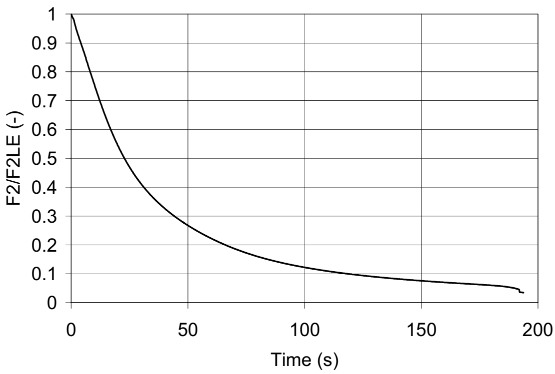
3.7. Nonlinear Estimation of the Real Tensile Characteristic

3.7.1. Methods of Nonlinear Estimations

Fig. 15 illustrates two different methods for estimating the tensile load-time (TLT) curve from a stress relaxation (SR) measurement, and *vice versa*, in indirect ways using the LVE estimations as an intermediate estimating operation.

The methods for the estimation of the real TLT curve (F_2) from the measured SR curve (F_1) can be described by the following sequences of transformation operations:

$$F_1(t) \to F_{12}(t) \to F_{122}(t) \approx F_2(t) \tag{34}$$



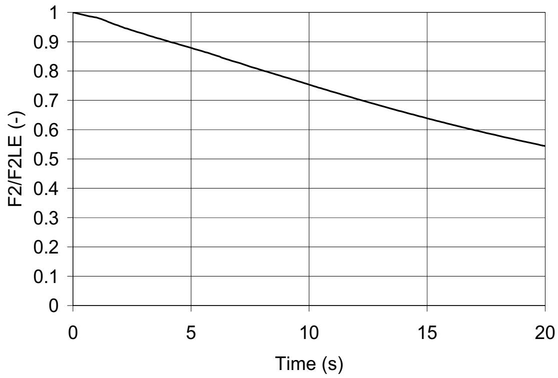


Fig. 13. Measure of the linear elasticity (MLE) in case of PP tested (a): up to neck forming, (b): at the beginning of the test

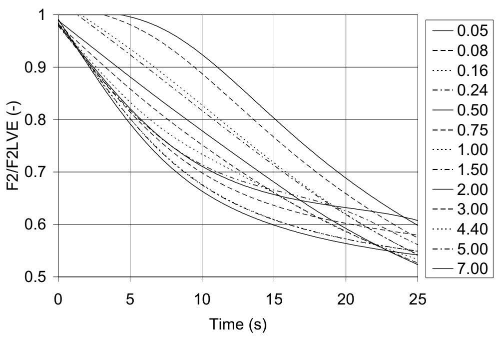


Fig. 14. The measure of the linear viscoelasticity (MLVE) for different strain levels (ε_0%)

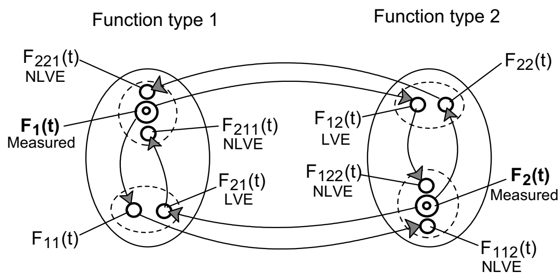


Fig. 15. Possible NLVE estimation ways of the real characteristics for types 1 and 2 using the LVE estimations. (Dashed line circles or ellipses denote the approximation vicinity of the estimated function.)

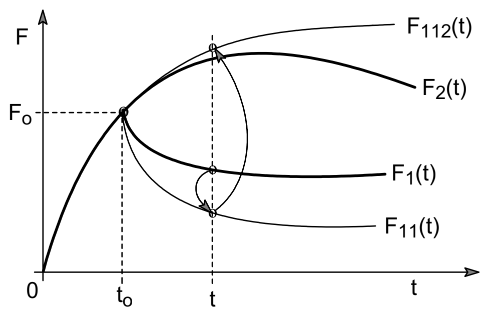


Fig. 16. The relation of the measured and the estimated curves

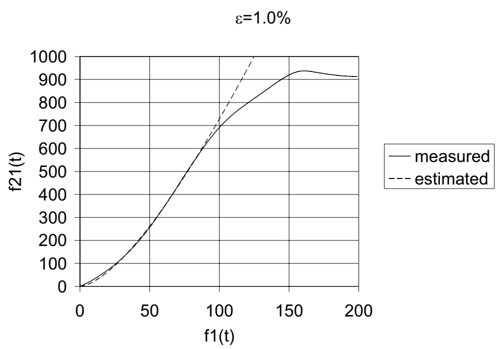


Fig. 17. Relationship between $f_{21}(t)$ and $f_1(t)$ ($\varepsilon = \varepsilon_0$)

$$F_1(t) \to F_{11}(t) \to F_{112}(t) \approx F_2(t) \tag{35}$$

In the inverse case the real SR curve (F_1) is estimated by the measured TLT curve (F_2) (*Fig. 15*):

$$F_2(t) \rightarrow F_{21}(t) \rightarrow F_{211}(t) \approx F_1(t) \tag{36}$$

$$F_2(t) \rightarrow F_{22}(t) \rightarrow F_{221}(t) \approx F_1(t) \tag{37}$$

Here, the operation in the first step in *Eqs.* (34) and (36) as well as in the second step in *Eqs.* (35) and (37) is an LVE transformation according to *Eq.* (22) and (21), respectively.

4. Nonlinear Estimation Based on the Relaxation Test with Ideal Stimulus

A method of estimating the TLT curve according to *Eq.* (34) was applied by RETTING (1992) [8], who assumed that for the nonlinear viscoelastic behaviour the left hand side of *Eq.* (9) could be used and the relaxation stress and modulus could be separated into two factors as follows:

$$\sigma(t, \varepsilon_o) = \varepsilon_o E_{NLVE}(t, \varepsilon_o) = \varepsilon_o a(\varepsilon_o) E(t) = g(\varepsilon_o) E(t) \tag{38}$$

where ε_0 is the strain level, $E_{NLVE}(t)$ and $E(t)$ are the NLVE and the LVE relaxation modulus, respectively, and $a(\varepsilon_0)$ and $g(\varepsilon_0)$ are functions to be determined for the given material. In case of linear viscoelasticity $g(\varepsilon_0) \equiv \varepsilon_0$ and $a(\varepsilon_0) \equiv 1$. For time-dependent strain the stress response can be obtained in a way similar to *Eq.* (5):

$$\begin{aligned} \sigma(t, \varepsilon) &= \int_0^t E(t-u) \mathrm{d}g(\varepsilon(u)) = \int_0^t E(t-u) \tfrac{\mathrm{d}g}{\mathrm{d}\varepsilon} \dot{\varepsilon}(u) \mathrm{d}u = \\ &= \int_0^t E(t-u) \left(a(\varepsilon) + \varepsilon \tfrac{\mathrm{d}a}{\mathrm{d}\varepsilon} \right) \dot{\varepsilon}(u) \mathrm{d}u \end{aligned} \tag{39}$$

where $g(\varepsilon(t))$ is considered as a stimulus. Also, RETTING assumed that the derivative does not depend on the time, hence

$$\sigma(t, \varepsilon) = \frac{dg}{\mathrm{d}\varepsilon} \int_0^t E(t-u) \dot{\varepsilon}(u) \mathrm{d}u. \tag{40}$$

In case of the tensile test $\dot{\varepsilon}$ is constant, therefore

$$\sigma(t, \varepsilon) = \frac{\mathrm{d}g}{\mathrm{d}\varepsilon} \dot{\varepsilon} \int_0^t E(u) \mathrm{d}u = G(\varepsilon) \sigma_{LVE}(t) \tag{41}$$

where $\sigma_{LVE}(t)$ is an LVE estimation of the tensile stress-strain curve identical with that according to *Eq.* (11). The estimation is valid for the ideal relaxation stimulus.

$$G(\varepsilon) = \frac{\mathrm{d}g}{\mathrm{d}\varepsilon} = a(\varepsilon) + \varepsilon \frac{\mathrm{d}a}{\mathrm{d}\varepsilon} \tag{42}$$

is a – usually nonlinear – so-called strain-function to be identified. This function is identically equal to 1 for LVE behaviour.

Rewriting *Eq.* (41) in order to get the tensile force by multiplying by the cross sectional area of the specimen (A), we obtain using the notations in *Fig. 15*:

$$F_2(t, \varepsilon) = G(\varepsilon) F_{12}(t). \tag{43}$$

According to Retting's examinations on PVC [8] the strain range of the linear viscoelastic behaviour revealed as about 0.5 % while applying the method shown above extended that up to the beginning of the yielding which was at 6.5 %. The strain range of the relaxation curves was 0.25 … 7 %, while the time range, however, was very short, only 3…50 s.

5. Estimation Based on the Relaxation Test with Real Stimulus

In this paper another way is sought and followed where we want to make use of *Eq.* (22) which is valid for the real relaxation stimulus. According to the other method applied here for estimating the TLT curve (see *Eq.* (35)), firstly curve $F_{11}(t)$, a kind of tangent modulus curve is calculated from fitting that to LVE estimation $F_{21}(t)$ and in the second step from $F_{11}(t)$ the tensile load-time curve $F_2(t)$ is estimated with $F_{112}(t)$ (*Fig. 15* and *Fig. 16*).

Rewriting *Eq.* (35) in a detailed form by means of operators we obtain:

$$\begin{aligned} F_1(t) = S[\Delta l_1](t) \rightarrow F_{11}(t) = S_1[F_1](t) \rightarrow F_{112}(t) = \\ = S_2[F_{11}](t) \approx F_2(t) = S[\Delta l_2](t) \end{aligned} \tag{44}$$

where S is the specimen operator, while S_1 and S_2 are NLVE and LVE estimating operators, respectively. The relations between the measured and the estimated curves are illustrated in *Fig. 16*.

It is advantageous to apply operator S_1 for the following difference (f_1) instead of $F_1(t)$:

$$f_1(t) = F_1(t_o) - F_1(t) \tag{45}$$

because in this case $f_1(0) = 0$. The resulting function can be obtained with operator S_1 in the following form:

$$F_{11}(t) = S_1[F_1](t) = F_{11}(t_o) - S_{11}[f_1(t)](t). \tag{46}$$

The mapping represented by operator S_2 is realized by *Eq.* (22) which is recursive, consequently operator S_2 is linear:

$$F_{112}(t) = F_{112}(t - t_0) + F_{11}(t) \tag{47}$$

Basically $t \geq t_0$ is meant here, however, it can be extended to $t \geq 0$ by joining the initial part of the tensile load-time curve in (0, $t \geq t_0$) with estimation $F_{112}(t)$.

The unknown operator S_1 or S_{11} is to be chosen so that estimation $F_{112}(t)$ provides an acceptable accuracy in a given time range.

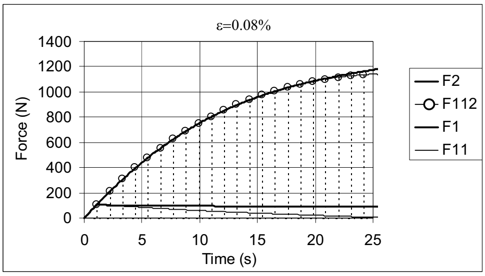


Fig. 18. NLVE estimation of the tensile load-time curve with the relaxation curve at strain level $\varepsilon = \varepsilon_0 = 0.08\%$ ($t_0 = \varepsilon_o/\dot{\varepsilon}_o = 1.06$ s)

Results of the Non-linear Viscoelastic Estimations

In order to demonstrate the applicability of our idea and to perform the numerical calculations we have chosen a flexible function with three parameters to realize nonlinear operator S_{11} according to *Eq.* (46) as follows:

$$F_{11}(t) = F_{11}(t_o) - a(\varepsilon)\,[f_1(t)]^{b(\varepsilon)}\,e^{c(\varepsilon)f_1(t)} \tag{48}$$

where parameters a, b, and c depend on the strain (ε). If $a = 1$, $b = 1$, and $c = 1$ then *Eq.* (48) gives the identity $F_{11}(t) = F_1(t)$ because $F_{11}(t_0) = F_1(t_0)$.

In order to determine parameters a, b, and c, function $F_{21}(t)$, the LVE estimation of the relaxation curve calculated from the measured tensile load-time curve (see *Fig. 15* and *Eq.* (36)), was used. The regression relation between differences $f_1(t)$ and $f_{21}(t)$

$$\begin{aligned} f_{21}(t) = F_{21}(t_o) - F_{21}(t) \approx f_{11}(t) = \\ = F_{11}(t_o) - F_{11}(t) = a(\varepsilon)\,[f_1(t)]^{b(\varepsilon)}\,e^{c(\varepsilon)f_1(t)} \end{aligned} \tag{49}$$

was calculated by maintaining good fitting mainly for the smaller values, that is the initial curve part convex from below (*Fig. 17*).

Figs 18, 19, and *20* represent the way how estimations $F_{112}(t)$ according to *Eq.* (47) approximate tensile load curve $F_2(t)$ at different initial strains as loads.

In case of a small strain level ($\varepsilon = \varepsilon_0 = 0.08\%$) the original approximation range of the LVE estimation (about 5 s) was enhanced to a four times larger one (>21 s is the total and about 20 s is the approximated range) (*Fig. 18*).

Increasing the strain level of the relaxation test to 0.5 % the time range of the good fitting for the NLVE estimation essentially remained at the same (>26 s is the total and about 20 s is the approximated range) (*Fig. 19*).

In case of a high strain level (3 %) the good fitting range for the NLVE estimation decreased (>22 s is the total and about 9 s is the approximated range) (*Fig. 20*).

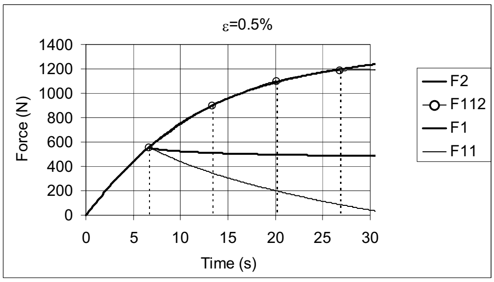


Fig. 19. NLVE estimation of the tensile load-time curve with the relaxation curve at strain level $\varepsilon = \varepsilon_0 = 0.5\%$ ($t_0 = 6.6$ s)

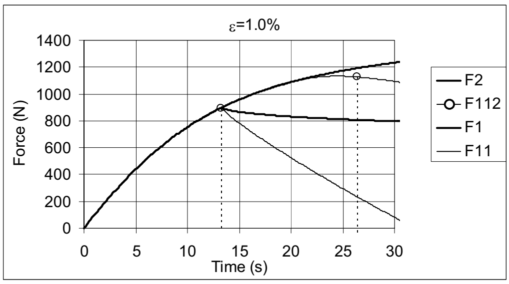


Fig. 20. NLVE estimation of the tensile load-time curve with the relaxation curve at strain level $\varepsilon = \varepsilon_0 = 1.0\%$ ($t_0 = 13.2$ s)

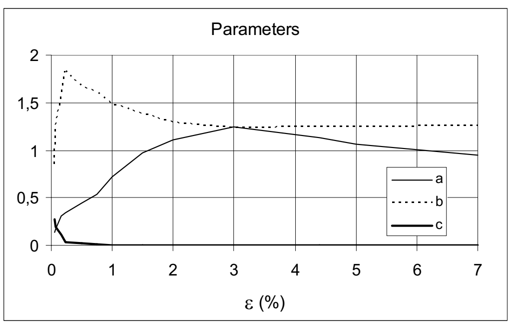


Fig. 21. Parameters a, b, and c versus strain level $\varepsilon = \varepsilon_0$, determined by fitting $F_{11}(t)$ to $F_{21}(t)$.

Of course parameters a, b, and c depend on the strain level ($\varepsilon = \varepsilon_0$) as *Fig. 21* shows their values determined by fitting $F_{11}(t)$ to $F_{21}(t)$, according to *Fig. 15*.

According to the measurements and fitting calculations as well as to theoretical considerations concerning the slope of the initial tangent of the curve in *Fig. 17*, it can be established that if εincreases then $b \to 1$ and $c \to c_0 \approx 0$.

Actually, b and c seem to tend to a constant value with the strain level and what is more so does a to a certain degree. Parameter b ranges from 1.23 to 1.27 with an average of 1.25 if $\varepsilon = \varepsilon_0 > 3\%$ in *Fig. 21*. The fluctuation of parameter c is smaller than that of b: -0.001 … +0.007 and the average is 0.003 if $\varepsilon = \varepsilon_0 > 0.75\%$. The variation of a is a bit more changeable, nevertheless a might tend to 1 if $\varepsilon = \varepsilon_0 > 4.4\%$ because it ranges from 0.94 to 1.14 with an average of 1.05 according to *Fig. 21*.

By choosing another suitable type of the possible mappings for operator S_{11} the range of good fitting can be increased. It would be important to make use of all the information included in the relaxation measurement even if it is carried out as a long term test.

6. Concluding Remarks

This paper made an attempt to investigate the relationships between the results of the tensile and the stress relaxation tests of polypropylene specimens and found a way to estimate the former from the latter based on the measurements and the theory of the linear viscoelasticity used for real stimuli.

The real polymers are basically of nonlinear character therefore the behaviour patterns do not meet the real one of materials, especially in case of larger loads. Nevertheless, the linear viscoelasticity often used successfully for describing the real behaviour in case of small or moderate loads.

Based on theoretical considerations a way was sought and followed where we made use of the linear viscoelastic response given to the real relaxation stimulus and its relation to the tensile load-strain curve.

Experiments were performed on PP and the stress relaxation behaviour, as well as the linear elastic and linear viscoelastic approximation of the tensile load-time curve was analysed.

In order to demonstrate the applicability of our idea for nonlinear approximation and to perform the numerical calculations we have chosen a flexible function with three parameters to realize the nonlinear behaviour. From the experimental and numerical analysis it was concluded that for the PP material tested the *linear elastic* range was about 1 s in time (what corresponds to 0.08 % in strain), while the *linear viscoelastic* approximation range calculated from the initial point was 4 s (0.3 %) at 0.08 % initial strain level and 1 s (0.08 %) at 3% initial strain level. Using the *nonlinear viscoelastic* approximation elaborated here the range of good fit resulted in 20 s (1.5 %) at 0.08 % initial strain level and 9 s (0.7 %) at 3% initial strain level.

By choosing another suitable type of the possible mappings for operator S_{11} the range of good fitting can be increased. It would be important to make use of all the information included inside the relaxation measurement even if it is carried out as a long term test.

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