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Identification of the viscoelastic parameters of a polymer model by the aid of a MCMC method

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Abstract

A procedure to identify the viscoelastic material parameters of a solid amorphous polymer and to estimate their values is presented. Stress-strain material data is obtained for the polymer by a compression experiment. The material behavior of the polymer is modeled according to the generalized Maxwell model, which is fitted to the experimental data by the method of least squares to obtain a first approximation for the model parameters. The identification of the model parameters is completed by a Markov chain Monte Carlo (MCMC) method, which generates the probability distributions of the relevant parameters of the material. The utilized MCMC method enables us to determine a suitable complexity (i.e., the number of Maxwell elements) for the generalized Maxwell model, so that the model best fits the data and, simultaneously, leads to an identifiable set of parameters. The numerical results imply that the uniqueness of the solution is lost when the number of model parameters becomes redundant.

Keywords: polymer, generalized Maxwell model, compression experiment, parameter identification, MCMC

1. Introduction

Polymers are widely used engineering materials. While in many materials, for example, metals, deviations from perfect elasticity are negligible for small strains, in polymeric materials, by contrast, the mechanical behavior is dominated by viscoelastic phenomena. The difference is due to the fact that in the deformation of hard solids atoms are displaced from their equilibrium positions only locally, whereas, in polymers, the flexible threadlike molecules are rearranged on a local as well as long-range scale, giving rise to rapid as well as slow responses, respectively. This leads to viscoelastic behavior and to a wide range of time scales under external stress (Ferry, 1980; Ward and Sweeney, 2004).

When polymeric components are used in industrial applications, it is often necessary to know the structural parameters, such as stiffnesses and relaxation times, related to the polymers. These parameters can be estimated by several techniques that utilize the response of the material to a properly selected external excitation. It is commonplace that estimates for the parameters are

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found by a least squares fit to measurement data, see, for example, (Vuoristo et al., 2000; Sorvari and Malinen, 2007; Weick, 2009; Acton and Weick, 2011). A particular feature of mechanical analog models for viscoelastic materials, consisting of arrangements of springs and dashpots, is that the number of material parameters, that is, the number of springs and dashpots, is not fixed beforehands. A large number of parameters is likely to produce a good fit to the data, but in such a case the parameters might not be well-identified, that is to say, their values can be varied substantially without affecting the goodness of the fit too much. This raises the question: What is the optimal set of parameters that best fits the measurement data and is still unique at the same time? To this end, methods based on Bayesian statistics have recently emerged, which produce "all" the parameter combinations able to explain the measured data, not just those corresponding to the numerical minimum of the least squares cost function, providing novel means to include comprehensive statistical analysis into the model fitting procedure (Gilks et al., 1996; Robert and Casella, 2004).

In the case of the solid amorphous polymer studied in this work, the external excitation is provided by a simple compression test. A first approximation for the model parameters is obtained by fitting the generalized Maxwell model to the measured values by the method of least squares. Then, the material parameters are sampled using a Markov chain Monte Carlo (MCMC) method, and the expectation (mean) values as well as probability distributions for the material stiffnesses and relaxation times are obtained. By the aid of these, a suitable number of parameters for the model that best fits the data and simultaneously leads to an identifiable set of the parameters may be determined. A MCMC method has been used in a recent paper by Oates et al. (2013), who quantified the viscoelastic model uncertainty of a finite deformation, hyperelastic spring-dashpot model for *a fixed number* of model parameters and then incorporated the uncertainty into a refined stochastic-based model through homogenization. In light of this, the main novelty of our study is that the MCMC procedure in association with the method of least squares provides a way of determining, from a statistical point of view, *an optimal number*, not fixed a priori, of Maxwell elements to be included in the linear generalized Maxwell model when fitting the model to the stress-strain material data.

2. Mechanical model of viscoelastic behavior

Viscoelastic material models usually utilize a hereditary approach where stress is a function of the current strain as well as the past history of the strain in the material. For small strains, a linear mechanical model may be assumed. A popular constitutive model is the well-known classical generalized Maxwell model (Ferry, 1980; Ward and Sweeney, 2004). For example, in widely-used commercial finite element software (Simulia, 2010), the Prony series presentation of viscoelasticity corresponding to the classical generalized Maxwell model is typically used within the context of small strains (Park and Schapery, 1999; Park and Kim, 2001). In recent years, the fractional calculus versions of different viscoelastic models have also been of high interest, see, for example (Bagley and Torvik, 1986; Renaud et al., 2011). In this work, the viscoelastic material is a solid amorphous polymer which we model using the generalized Maxwell model shown in Fig. 1.

The material element contains $n+1$ springs with elastic moduli $E_\infty, E_1, \ldots, E_n$ and n dashpots with constants of viscosity $\eta_1, \ldots, \eta_n$. In the model, the spring associated with the long-term modulus E_∞ describes the restoring effect of the cross-links of the molecular polymer chains. The Maxwell elements in parallel represent the elastic deformations of the polymer chains and the damping due to friction between the chains (Cowie and Arrighi, 2008). Every Maxwell element

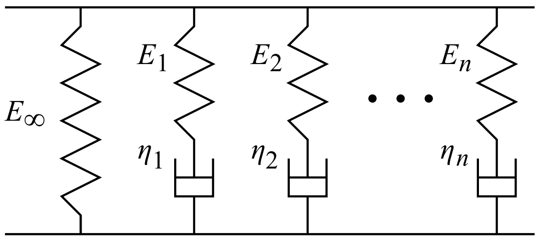

Figure 1: Generalized Maxwell model describing a viscoelastic material element. The number of the spring-dashpot pairs, that is, Maxwell elements, is n.

has a relaxation time

$$\tau_i = \frac{\eta_i}{E_i} \quad (i = 1, \ldots, n) \ . \tag{1}$$

Together the elements determine the relaxation spectrum of the polymer. Since in a compression test the inertial forces are small compared to the viscoelastic ones, the relations between stress and strain in the Maxwell model can be written as

$$\sigma_\infty = E_\infty \epsilon \tag{2}$$

$$\sigma_i = E_i(\epsilon - \epsilon_i) \tag{3}$$

$$\eta_i \dot{\epsilon}_i = E_i(\epsilon - \epsilon_i) \ , \tag{4}$$

where σ_∞ and σ_i are the stresses in the springs, ϵ is the strain of the whole material element, and ϵ_i is the strain of the ith dashpot ($i = 1, \ldots, n$). The total uniaxial stress (the total force divided by the contact area) of the material element is

$$\sigma = \sigma_\infty + \sum_{i=1}^{n} \sigma_i \ . \tag{5}$$

Applying Eqs. (2) and (3) in Eq. (5) yields

$$\sigma = E\epsilon - \sum_{i=1}^{n} E_i \epsilon_i \ , \tag{6}$$

where the total instantaneous stiffness of the element

$$E = E_\infty + \sum_{i=1}^{n} E_i \ , \tag{7}$$

has been used. In order to get a relation between the total stress and strain of the whole material element, the strains of the dashpots ϵ_i must be solved. Eqs. (1) and (4) give

$$\epsilon_i + \tau_i \dot{\epsilon}_i = \epsilon \quad (i = 1, \ldots, n) \ . \tag{8}$$

Eqs. (6) and (8) can be used to integrate the values of the strains $\epsilon(t), \epsilon_i(t)$ at a given time t, when the measured compressive stress history $\sigma(t')$ $(0 \leq t' \leq t)$ for the polymer test piece is given. Before the compression starts at $t = 0$, the material is at rest in equilibrium so that the initial conditions are $\epsilon(0) = 0$ and $\epsilon_i(0) = 0$ for all i. Eqs. (6) and (8) express the viscoelastic dynamic behaviour of the generalized Maxwell model under uniaxial loading.

3. Compression experiment

In this work, the data for the parameter estimation was created by a compression experiment of 31.9 seconds duration, where the strain of a block-like polymer piece was given a target value and the stress (via the applied force exerted on the block) was controlled so as to keep that target value. Samples of the measured stress and strain histories in the time interval (0,1) s are shown in Fig. 2(a) and the corresponding stress-strain relation in Fig. 2(b). The target value of strain was set to 3%. During the first 0.06 s the stress was increased from zero in such a way that the target value of strain was approximately achieved. After that the force control system regulated the stress so that the target value was sustained. As can be seen from Fig. 2(a), the strain remains quite close to the target value. There are, however, small fluctuations around this value due to external random disturbances and, possibly, due to small oscillations in the feedback control system. The relaxation of stress as a function of time can be clearly seen in Figs. 2(a) and 2(b). There are also fluctuations in the stress, although not as much as in the strain.

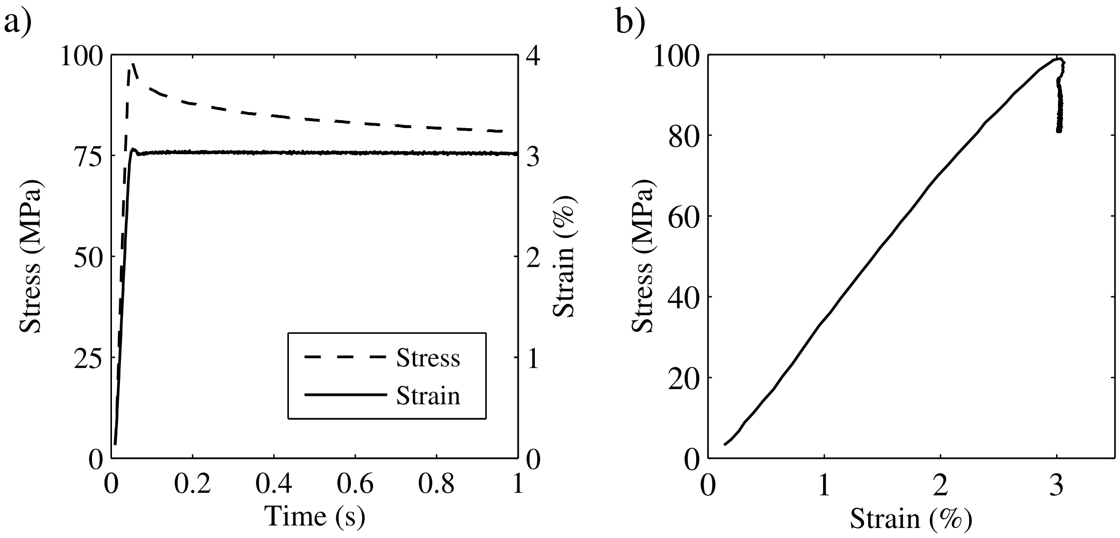

Figure 2: Samples of measured stress and strain histories in the compression test as a function of time, b) corresponding stress-strain relation.

In the following the extraction of the parameter values of the generalized Maxwell model from the measured data is done by using the method of least squares fitting (LSQ) and the Markov chain Monte Carlo (MCMC) method.

4. Parameter estimation

A good parameter estimation method should provide reliable estimates for the desired system parameters even in the presence of external perturbations and measurement errors. This can be achieved by a combination of the least squares fitting method together with the MCMC sampling approach.

A nonlinear curve fitting in the sense of least squares is utilized first. The least squares algorithm is known to be relatively robust due to its low-pass filtering properties. It consists of finding the

Table 1: Estimated values for the parameters E_∞, E_i and τ_i ($i = 1, \ldots, n$) of the generalized Maxwell model for different values of n.

n	2	3	4	5
E_∞ (GPa)	2.248	2.231	2.113	1.751
E_1	0.477	0.603	0.704	0.310
E_2	0.402	0.289	0.269	0.269
E_3		0.365	0.298	0.668
E_4			0.332	0.309
E_5				0.384
τ_1 (s)	0.431	0.058	0.024	0.411
τ_2	9.513	1.117	3.936	3.957
τ_3		12.05	0.407	0.025
τ_4			35.83	34.40
τ_5				1413

system parameters so that the squared 2-norm of the pointwise modeling error is minimized, that is

$$r(\boldsymbol{\theta}) = \sum_{j=1}^{m} \left[\epsilon_j(\hat{\boldsymbol{\sigma}}) - \hat{\epsilon}_j\right]^2 , \tag{9}$$

$$r = \min_{\boldsymbol{\theta}} r(\boldsymbol{\theta}) , \tag{10}$$

where r is the residual of the fit, m is the number of data points, $\boldsymbol{\theta} = (E_\infty, E_1, \tau_1, \ldots, E_n, \tau_n)^T$ is the vector of the system parameters to be determined, $\hat{\epsilon}_j$ and $\hat{\boldsymbol{\sigma}} = \{\hat{\sigma}_1, \ldots, \hat{\sigma}_j\}$ are the measured values of strain and stress, respectively, and $\epsilon_j(\hat{\boldsymbol{\sigma}})$ the strain calculated from Eqs. (6) and (8) for the measured stresses. The objective function (9) may be minimized by a suitable optimization algorithm. Here the basic Simplex method was used. The model was fitted to the data of the 31.9 s compression experiment for several values of the number of Maxwell elements in the model. The results for $n = 2, 3, 4$ and 5 are shown in Table 1. It can be seen from Table 1 that the spread of the estimated parameter values is quite large. Therefore, it is not obvious which is the optimal value for n and how unique the parameter values are. The measured and predicted (calculated) strain histories for $n = 4$, $n = 2$ and $n = 3$ using the values of Table 1 are shown in Figs. 3(a), 4(a) and 4(b), respectively. We can see that for $n = 3$ and $n = 4$ the predicted strain lies well within the limits set by the variations due to external noise and measurement error in the measured strain. Fig. 3(b) displays the histogram of the pointwise residuals of the fit for $n = 4$ (i.e., measured strain – predicted strain) together with the density function of the Gaussian normal distribution, whose average value ($\approx 1.9 \cdot 10^{-8}$) and standard deviation ($\approx 6.1 \cdot 10^{-5}$) are those calculated from the residual values. We can see that the residuals follow the normal distribution almost ideally well. We conclude that a fine goodness of the fit can be achieved for $n = 4$. However, the question of how unique the estimated parameter values are for different values of n, and how many Maxwell elements should be used in the model, remains yet open.

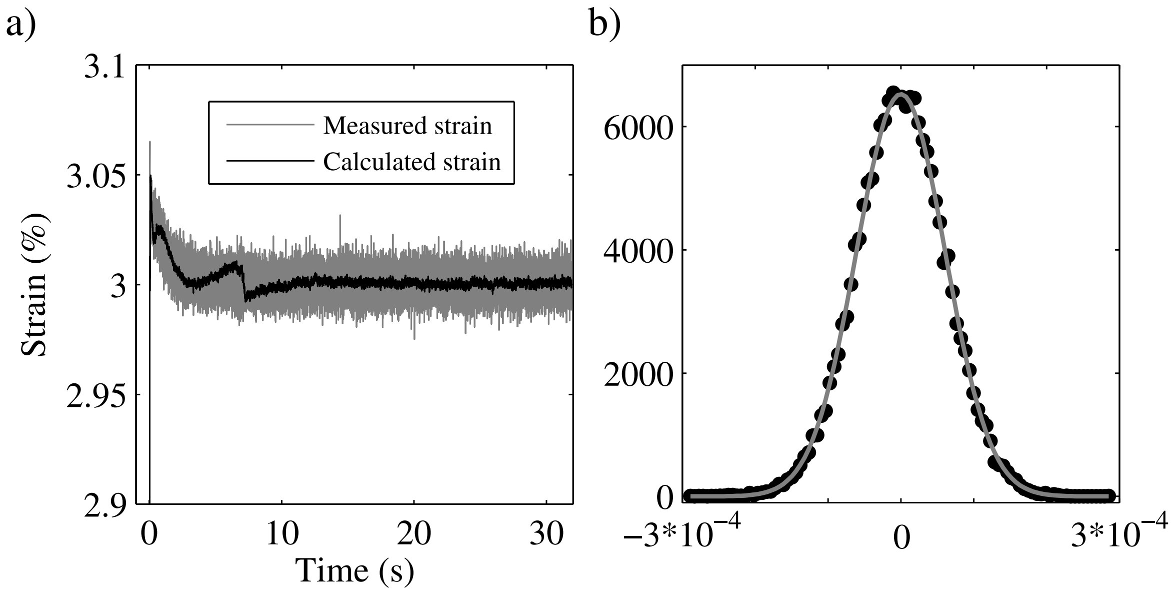

Figure 3: a) Measured and predicted strain histories for $n = 4$. The number of data points is $m = 31771$ and the separation of consecutive points is 1 ms. b) The histogram of the pointwise residuals of the fit for the 31771 points together with the corresponding normal density distribution.

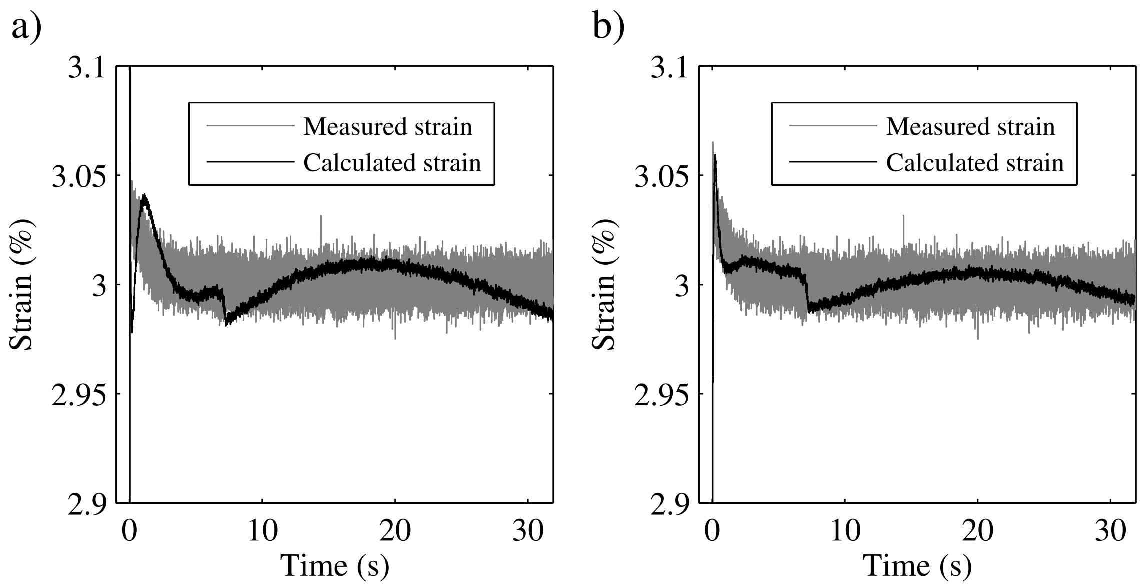

Figure 4: a) Measured and predicted strain histories for $n = 2$. b) Measured and predicted strain histories for $n = 3$.

5. Probability distribution of the parameters

The reliability of the estimated parameter values is analyzed using a MCMC method. MCMC methods are well-suited for the analysis of multiparametric models. When using a MCMC method, the estimation of the model parameters is performed according to the Bayesian paradigm, that is, all the uncertainties in the measurement data are treated as statistical distributions. The idea of MCMC algorithms is not to obtain the "best fit" for the model parameter values, but to determine (practically) all the parametrizations of the model that adequately fit the data. A multidimensional posterior distribution of the unknown parameters is generated using the available prior information and the statistical knowledge of the measurement data. Basically, MCMC methods are able to generate sample candidates in the parameter space, that are either accepted or rejected, by using a proposal distribution. The rule of acceptance favors the more probable parameter values – better fits to data – more than the unlikely ones. It can be proved that with an increasing sample size the histograms of the calculated parameter distributions approach the correct probability distribution of the parameters.

Here, for each value of n, the MCMC sampling algorithm to generate the posterior distribution is initialized by using the least square fit values as the first values for the sample chains, and the approximative covariance of the least squares estimate as the covariance of a Gaussian proposal distribution. For a more detailed summary of the basic concepts and algorithms of Bayesian parameter estimation and MCMC methods, see (Liu et al., 2013; Solonen and Haario, 2012). In this study, up-to-date adaptive computational schemes are employed in order to make the simulations as effective as possible (Haario et al., 2001). Furthermore, for the Adaptive Metropolis algorithm used here, see the readily available MCMC toolbox for Matlab, which provides the necessary MCMC tools for computational analysis, including several example problems (Laine, 2013).

The results of the MCMC analysis for the case $n = 3$ are shown in Fig. 5. The algorithm produces a sample from the full, seven-dimensional in the present case, parameter posterior distribution, from which one- and two-dimensional marginal distributions are presented in the figures. The sample size used was 30 000 samples, and the CPU time for creating such a sample was a few minutes on a usual computer. The results in Fig. 5 show that the probabilities are well centered around the most probable point, so that all the estimated parameters are rather well-identified in this case (cf. Table 1).

The same analysis was carried out for other values of n. The observed trend is clear: a low value of n leads to well-identified parameters, but to a less satisfactory goodness of the fit. A higher value of n usually improves the goodness of fit, but leads to poorly identified parameter values. For reliable predictions, in situations other than those used in the model fitting, however, the parameters should be well-identified. Fig. 6 presents one- and two-dimensional marginal posterior distributions of the parameters for $n = 4$. We can see that several parameter pairs are now clearly correlated (note the long cigar-type 2D distributions) indicating a weaker identification of those parameters.

The residual r of the fit and the indeterminacy e of the solution as a function of the number of Maxwell elements of the material model are shown in Fig. 7. The indeterminacy e, describing the non-uniqueness of the parameter identification, was evaluated by principal component projections of the sampled chains providing a measure of the total relative spread of the evaluated model parameters. We conclude that the optimal number of system parameters, on statistical grounds, is nine ($n = 4$) with the used measurement data. Above that the fit does not improve but the uniqueness of the estimated parameter values is gradually lost. It should also be noted that for

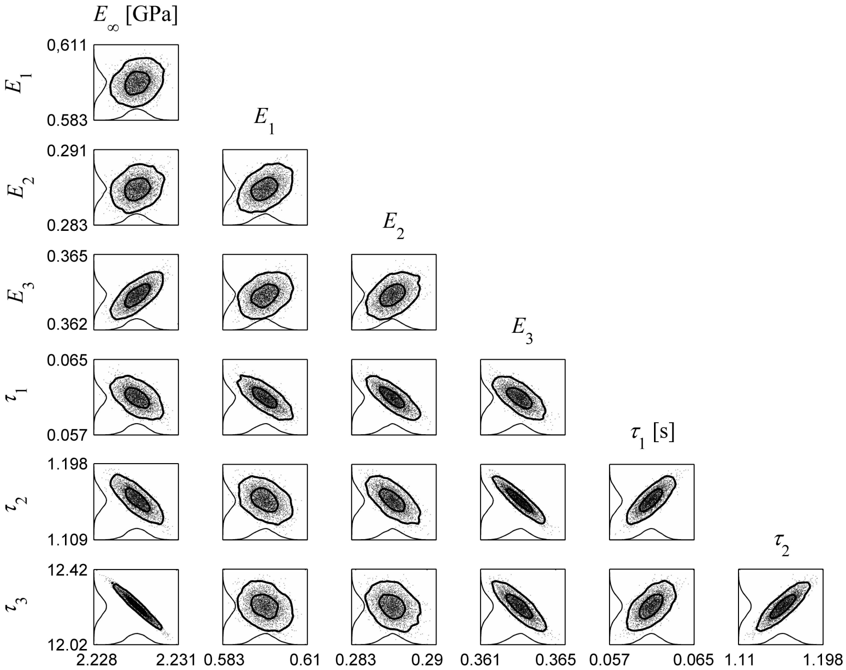

Figure 5: 2D and 1D marginal distributions of the estimated parameter values for $n = 3$. The ellipsoidal inner and outer lines represent the 50% and 95% confidence regions, respectively, and the separate curved lines are the corresponding one-dimensional density estimates.

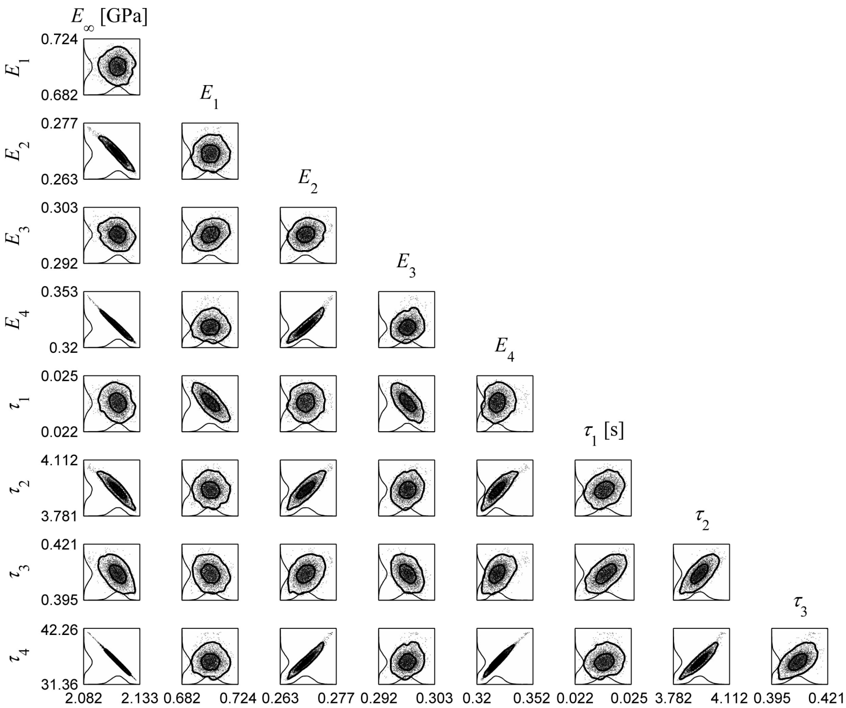

Figure 6: 2D and 1D marginal distributions of the estimated parameter values for $n = 4$. The ellipsoidal inner and outer lines represent the 50% and 95% confidence regions, respectively, and the separate curved lines are the corresponding one-dimensional density estimates. Note the thin distributions indicating a clear correlation between several parameters.

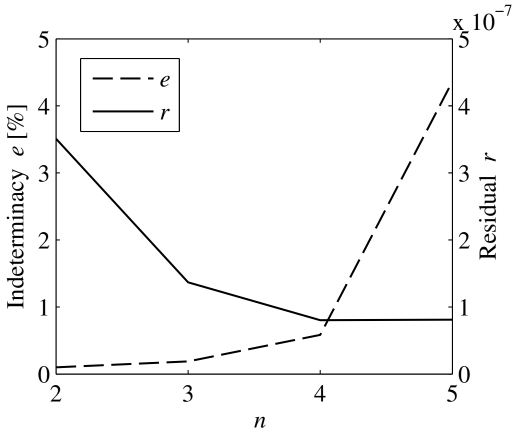

Figure 7: Residual of the fit r and non-uniqueness of the solution e as a function of the number of spring-dashpot pairs n in the Maxwell model.

$n = 5$ the relaxation time $\tau_5 = 1413$ s is completely outside the measurement data and, thus, unphysical. The limit of $n = 5$ is of interest because it cannot be known a priori, but there is no sense to use more than nine parameters ($n = 4$) in the generalized Maxwell model of the polymer studied in this work. A good choice is to use the mean values of the 1D marginal distributions of each parameter. These values for the model parameters are presented in Table 2.

Note that the values of Table 2 are very close to those of Table 1. For larger values of n, however, when the identifiability of the parameters is poorer ($n \geq 5$), the parameter values obtained from the LSQ fit can be considerably different from the corresponding mean values of the 1D marginal distributions obtained by the MCMC method. The reason for this is that inverse problems, in general, are ill-conditioned. This means that the minimum of the objective function (9) may be very flat at least in some directions of the parameter space. Therefore, the minimum search algorithm may end up quite randomly in a wide region of the parameter space. The mean values of the sampled MCMC chains, on the contrary, provide statistically reliable estimates for the model parameters.

Table 2: Mean values and ranges of the 95% confidence regions of the 1D marginal distributions of the generalized Maxwell model parameters for $n = 4$.

	mean value	95% confidence region
E_∞ (GPa)	2.113	2.095 – 2.125
E_1	0.702	0.689 – 0.718
E_2	0.269	0.265 – 0.274
E_3	0.298	0.295 – 0.301
E_4	0.331	0.322 – 0.343
τ_1 (s)	0.024	0.023 – 0.024
τ_2	3.938	3.820 – 4.052
τ_3	0.407	0.398 – 0.417
τ_4	35.76	32.24 – 39.52

6. Conclusions

In this communication, a system identification procedure, combining a least squares fitting method together with a Markov chain Monte Carlo sampling approach, for the evaluation of the material parameters of a viscoelastic polymer was presented. The purpose of the procedure is to determine reliable parameter values and a suitable complexity for the utilized material model, that is, an optimal number of material parameters for the model, when the number of parameters is not fixed a priori. In the present case, the generalized Maxwell model was used and the optimal number of the Maxwell elements was determined to be four, since for higher values of n, the goodness of the fit did not improve, but the uniqueness of the parameter values was lost. That is to say, numerical evidence suggests that the uniqueness of the solution is lost when the number of parameters becomes redundant, while, the goodness of the fit does not improve. Novel MCMC sampling methods provide an efficient way to perform statistical analysis and, thus, give tools for model selection, and their applicability extends to cases with strong nonlinear correlations between system parameters.

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