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Evaluation of relaxation properties of digital materials obtained by means of PolyJet Matrix technology

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Abstract. The paper presents the results of research on the stress relaxation of selected digital materials obtained by means of additive technology. A 5-parameter Maxwell-Wiechert model was used to describe the stress relaxation curves, allowing to obtain a very close fit. Anisotropy of properties was found due to the direction of sample printouts.

Key words: additive technology, digital materials, stress relaxation, Maxwell-Wiechert model.

1. Introduction

Polymers have found widespread use in additive manufacturing technologies, also known as 3D printing technologies. Their dynamic development has been observed recently. As a result of additive shaping, the starting materials – most often liquids or powders – provide a composite (solid material) with a pre-designed shape. During the additive manufacturing process, properties of the material obtained are also shaped. Those include e.g. mechanical or rheological properties, which are the subject of research [1–4]. Additive technologies, including PolyJet, and digital materials (http://www.stratasys.com/materials/polyjet/digital-materials) are used in this technology. The concept and principle of building digital materials can be found in paper [5]. Digital materials are created as a result of mixing two or three base materials (photocurable resins) during the dosing process on the machine's working platform. Dispensing resin droplets using appropriate heads is digitally controlled, hence the name of "digital materials". The dispensed resins are cured using UV light emitted by lamps mounted onto the dosing head. The basic line of the PolyJet Matrix technology is formed by base materials with the commercial names of TangoBlackPlus and VeroWhitePlus, which when mixed together in different proportions allow to obtain new materials with different Shore hardness. The material properties given in the catalogs are indicative. Additively shaped materials show anisotropy depending on the setting on the machine's working platform, i.e. on the print direction. More information on the anisotropy of mechanical properties of additively shaped materials can be found, for example, in papers [6–10].

The polymers exhibit viscoelastic properties, i.e. to phrase it more simply, they deform elastically under the influence of external stresses, taking the time factor into account. We the flow of material is considered, two aspects need to be distinguished: creep is the increase in strain over time under a constant level of stress, and relaxation is the decrease in stress over time with constant deformation. Stress relaxation is determined by conducting the so-called stress relaxation test, recording a decrease in stress over time at a predetermined pre-set strain. In real bodies, deformation processes are highly complex and depend on factors such as type of stress, loading speed or load duration. Different models are used for mathematical description. They simulate viscoelastic systems, and include e.g. the simple Hooke, Newton or Maxwell models (Hooke, Newton, Maxwell), or more complex ones, constitute a combination of simple models. The Wiechert model [11] is one of those. In experimental practice, the frequently obtained curve is approximated by the relation between stress and strain appropriate for the adopted rheological model. In some cases, description using ordinary differential equations proves not enough and you can use fractional differential calculus [12, 13].

The goal of this work is to examine the stress relaxation of materials obtained by means of the PolyJet Matrix technology, and to describe the obtained stress relaxation characteristics by using a second-order Maxwell-Wiechert model. It has been demonstrated that the model assumed describes stress relaxation in tested digital materials in a reliable manner.

2. Rheological model

Once we obtain the result of the test in the form of a curve that constitutes the subject of our research and that characterizes a given phenomenon or property, we ask ourselves the following: what mathematical formula should be used to describe it and how shall we find an equation that will give us the result with satisfactory accuracy. The Maxwell-Wiechert model was used to describe stress relaxation in this paper, but it did not happen immediately. Initially, simpler models were tried, but they proved lacking in accuracy.

The Maxwell model is used frequently to describe the stress relaxation curve. Examples of that are to be found in papers [14–17]. The classic stress relaxation curve described

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by the basic Maxwell model, which is a serial connection of the Newton model and the Hooke model, is expressed by the following equation:

$$\sigma(t) = \sigma_0 e^{\frac{-t}{t_1}}, \tag{1}$$

where: σ_0 – initial value of stress for time $t = 0$,
t_1 – relaxation time.

Relaxation time is defined as the ratio of the material properties of both simple center components, i.e. the Newton and Hooke bodies, and is calculated as follows:

$$t_1 = \frac{\mu_1}{E_1}, \tag{2}$$

where: μ_1 – coefficient of viscosity,
E_1 – elastic modulus.

This model can be slightly improved by using a fractional Maxwell model, as described in papers [12, 18, 19]. The Maxwell model is also used in parallel with the Hooke model [11] described by the following equation:

$$\sigma(t) = \sigma_0 + \sigma_1 e^{\frac{-t}{t_1}}. \tag{3}$$

Equation (2), describing the basic Maxwell model, along with equation (3), describing the combined Maxwell and Hooke models, are not always sufficient to approximate the stress relaxation curve obtained by means of experiments. Therefore, a more complex model is used, e.g. the general Maxwell model, often referred to in the literature [1] as the Wiechert model. The Maxwell-Wiechert model is shown in Fig. 1, in graphical form. This model consists of n basic Maxwell models connected in parallel and of a Hooke's body model. E_0, E_1, E_2 ... E_n stand for the elastic modulus and m_1, m_2, ... m_n are the coefficients of viscosity of basic models. The general differential equation describing this model is as follows:

$$\begin{aligned} & a_0\sigma(t) + a_1\frac{d\sigma}{dt} + a_2\frac{d^2\sigma}{dt^2} + \dots + a_n\frac{d^n\sigma}{dt^n} = \\ & = b_0\varepsilon(t) + b_1\frac{d\varepsilon}{dt} + b_2\frac{d^2\varepsilon}{dt^2} + \dots + b_n\frac{d^n\varepsilon}{dt^n}. \end{aligned} \tag{4}$$

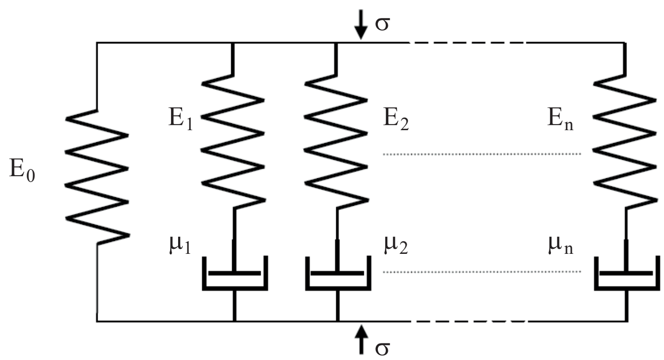

Fig. 1. General Maxwell-Wiechert model

In the case of stress relaxation with $\varepsilon = \varepsilon_0$, i.e. constituting a constant value, the solution of equation (4) is [1]:

$$\sigma(t) = \varepsilon_0 \left(\sum_{i=1}^{n} E_i e^{\frac{-t}{t_1}} + E_0 \right), \tag{5}$$

where: ε_0 – set displacement,
n – number of basic models,
i – marking the number of the next model,
t_i – relaxation time of the Maxwell's i-th model,
which is:

$$t_i = \frac{\mu_i}{E_i}, \tag{6}$$

where: μ_i – viscosity coefficient of the i-th model,
E_i – elastic modulus of the i-th model.

Formula (5) is a practical engineering formula because it can be applied after the construction of any multi-parameter Maxwell-Wiechert model in order to estimate the value of its individual elements. The user does not have to build and solve respective differential equations every time.

In this paper, the Maxwell-Wiechert model of the second order was used, i.e. one consisting of two basic Maxwell models and the Hooke model connected in parallel. The second-order Maxwell-Wiechert model is shown graphically in Fig. 2.

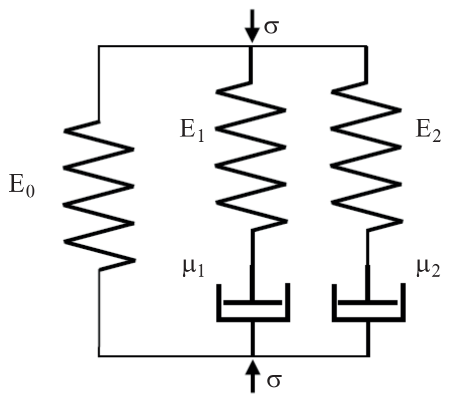

Fig. 2. Second-order Maxwell-Wiechert model

Due to the fact that the second-order model contains two basic Maxwell models and the Hooke model connected in parallel, formula (5) for $n = 2$ is as follows:

$$\sigma(t) = \varepsilon_0 \left(E_1 e^{\frac{-t}{t_1}} + E_2 e^{\frac{-t}{t_2}} + E_0 \right), \tag{7}$$

which after transformations gives:

$$\sigma(t) = \sigma_0 + \sigma_1 e^{\frac{-t}{t_1}} + \sigma_2 e^{\frac{-t}{t_2}}. \tag{8}$$

Equation (8) was used to describe stress relaxation in the tested materials.

3. Experimental phase and results

3.1. Samples and tests. Samples for testing were made of digital materials (photocurable resins) with the trade names of DM_8515_Grey35 and DM_9895_Shore95. They constitute a mixture of base materials Vero White and Tango Black plus. Cylindrical samples of the following dimensions: diameter D = 10 mm and height H = 15 mm, were made using the PolyJet additive technology (http://www.stratasys.com/3d-printers/technologies/polyjet-technology) in an Objet Connex 350 printer (now Stratasys). The solid model of the sample was drawn using the CAD 3D program and was then saved in a digital file with the extension *.stl*, using triangulation parameters in the export options: resolution – adjusted, deviation – tolerance 0.016 mm, angle – tolerance 5°. ext. Using the Objet Studio program, the sample models were placed (virtually) on the Connex 350 machine's working platform in a vertical and horizontal position. The arrangement of samples made from the DM_8515_Grey35 material during printing is shown in Fig. 3, with the samples positioned vertically and horizontally. Samples made from the DM_9895_Shore95 material were arranged in the same manner.

The samples were made in glossy mode to obtain a smooth surface. Ten samples were prepared for each material. After printing, the sample was removed from the machine's working platform and then the support material was removed and prepared for the stress relaxation test. The stress relaxation tests were performed using the Inspect mini testing machine. The test parameters were set in the Labmaster program, which is equipped with the Inspect mini machine, using block programming. For compressing the cylindrical samples, flat plates were used, with the lower plate oscillating, i.e. the flat part of the plate was mounted onto the spherical joint. The use of swinging discs to compress cylindrical samples provides for even loading of both flat surfaces of the sample. A displacement value of 1 mm was set and the value of this displacement was maintained for a time of 600 s for the DM_8515_Grey35 material and for 120 s for the DM_9895_Shore95 material. During this time, a decrease in the value of the sample compression force was recorded.

The detailed procedure for printing samples and the procedure for conducting stress relaxation tests are both described in paper [17]. The stress relaxation curves for individual samples were recorded using the Labmaster program in the form of graphs and raw ASCII data. The ASCII code (text entry) is extremely useful in laboratory practice. In this case, it enabled the separation of the stress relaxation curve and the use of data in the procedure of fitting the theoretical curve to the experimental curve.

3.2. Results. The collective results of the stress relaxation tests of the tested digital materials are presented in Figs. 4–7.

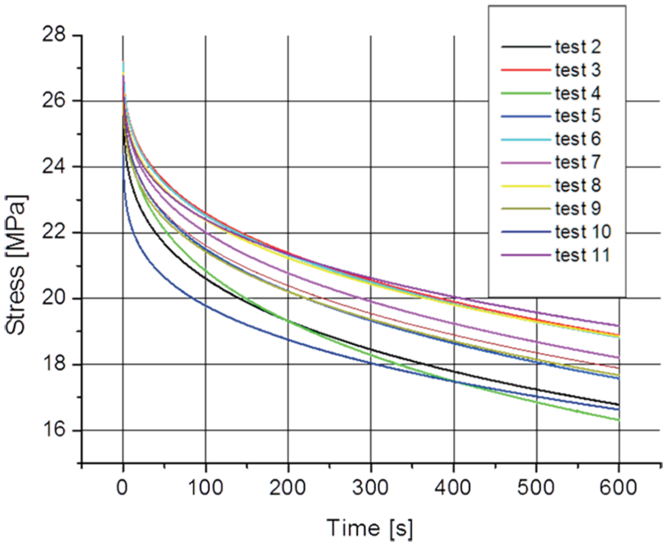

Fig. 4. Experimental stress relaxation curves of DM_8515_Grey35 digital material for vertically printed samples

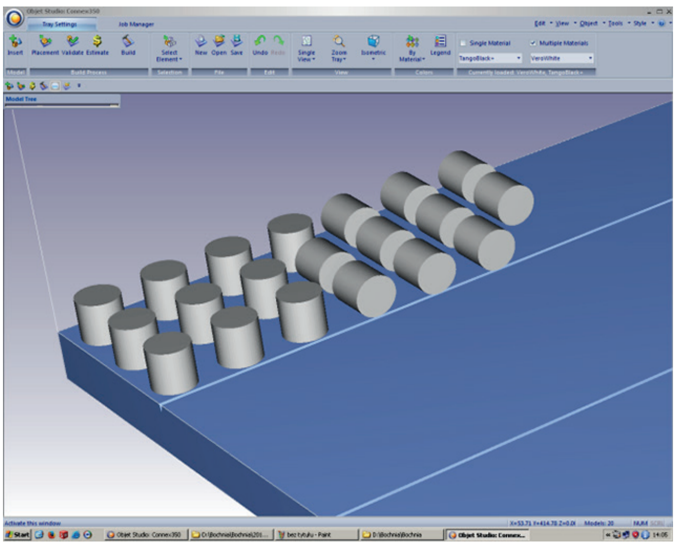

Fig. 3. Placement of samples on the working platform of the Connex 350 printing machine

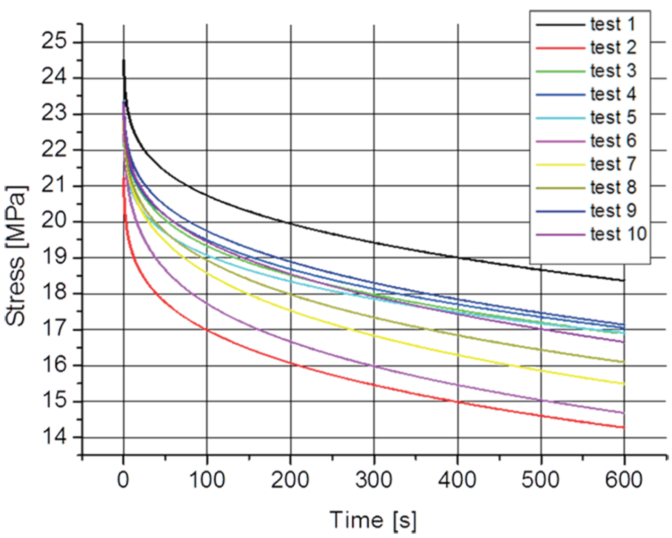

Fig. 5. Experimental stress relaxation curves of DM_8515_Grey35 digital material for horizontally printed samples

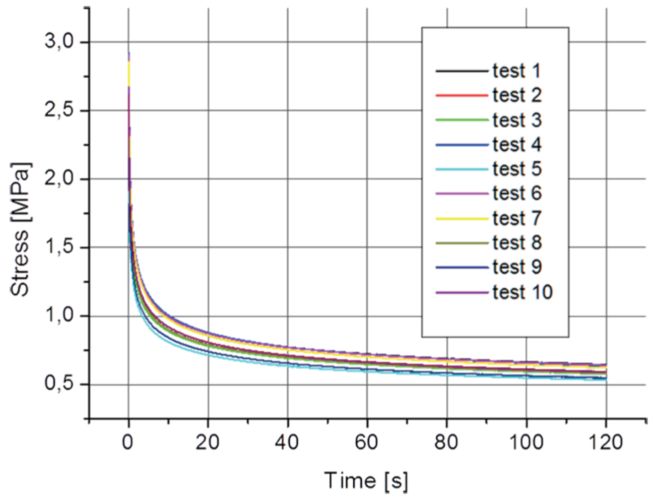

Fig. 6. Experimental stress relaxation curves of DM_9895_Shore95 digital material for vertically printed samples

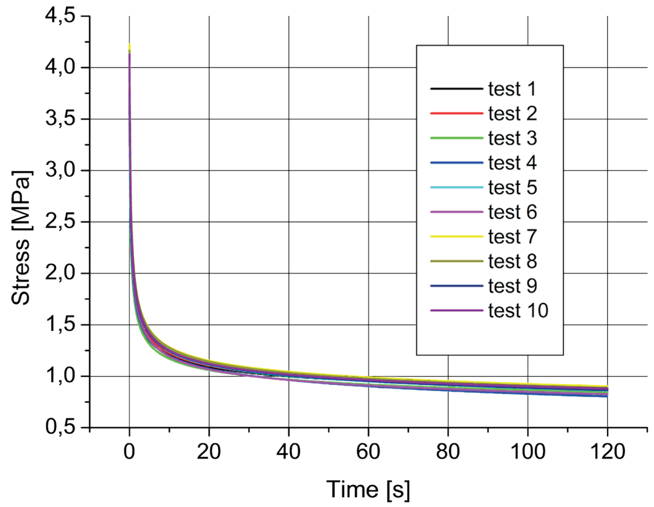

Fig. 7. Experimental stress relaxation curves of DM_9895_Shore95 digital material for horizontally printed samples

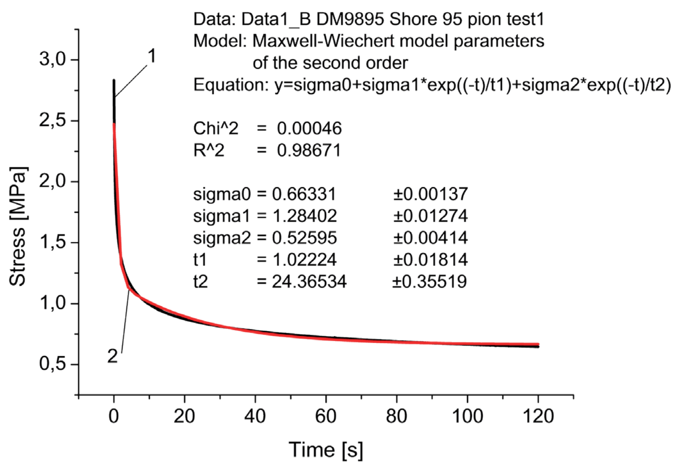

Fig. 8. Experimental curve compared with the approximation curve. 1 – experimental curve for samples made of DM_9895_Shore95 (vertical), 2 – approximation curve obtained using the second-order Maxwell-Wiechert model

Even a preliminary qualitative analysis of the stress relaxation curves obtained indicates different relaxation properties and different dispersion of the results arrived at. For quantification, an approximation was made using (8) for each experimental curve. The σ_0, σ_1, σ_2, t_1, t_2 parameter values were estimated for each curve. The fitting was carried out using the Levenberg-Marquardt method. The matching program simultaneously estimated the Chi^2 compatibility test values and R2 correlation coefficients for each match. Figure 8 shows an example together with estimated parameters.

The fit shown in Fig. 8 was made for each curve obtained. The results obtained are summarized in Tables 1, 2, 3 and 4, on

Table 1
Parameters of the stress relaxation curves obtained for material DM_8515_Grey35, samples printed vertically

Test No.	σ_0 [MPa]	σ_1 [MPa]	σ_2 [MPa]	t_1 [s]	t_2 [s]
1	15.646	2.628	6.551	21.8	352.3
2	17.558	2.325	6.447	23.4	390.3
3	14.974	2.915	7.734	23.0	352.1
4	16.243	2.528	6.813	22.3	377.5
5	17.482	2.357	6.468	22.7	391.1
6	16.877	2.384	6.619	22.6	383.5
7	17.621	2.274	6.137	22.1	383.8
8	16.345	2.427	6.558	22.0	385.6
9	15.626	2.329	5.447	20.4	366.2
10	18.075	2.240	5.602	21.7	379.0
$-\bar{X}$	**16.645**	**2.441**	**6.438**	**22.2**	**376.1**

Table 2
Parameters of the stress relaxation curves obtained for material DM_8515_Grey35, samples printed horizontally

Test No.	σ_0 [MPa]	σ_1 [MPa]	σ_2 [MPa]	t_1 [s]	t_2 [s]
1	17.670	1.966	4.060	19.2	353.1
2	13.538	2.135	4.622	19.4	338.4
3	16.179	2.021	4.196	20.0	354.3
4	16.302	1.794	4.207	20.6	358.3
5	16.323	1.762	3.683	19.2	338.8
6	13.933	2.693	5.125	21.3	324.1
7	14.633	2.194	5.246	21.2	342.7
8	15.271	2.007	4.846	20.7	349.7
9	16.291	1.793	4.506	21.1	370.0
10	15.644	1.934	4.874	21.3	392.7
$-\bar{X}$	**15.578**	**2.030**	**4.537**	**20.4**	**352.2**

Table 3
Parameters of the stress relaxation curves obtained for material DM_9895_Shore95, samples printed vertically

Test No.	σ_0 [MPa]	σ_1 [MPa]	σ_2 [MPa]	t_1 [s]	t_2 [s]
1	0.663	1.284	0.526	1.0	24.4
2	0.600	1.264	0.469	1.0	24.5
3	0.594	1.126	0.469	1.0	24.4
4	0.657	1.343	0.555	1.0	24.3
5	0.547	0.989	0.421	1.0	24.3
6	0.660	1.345	0.543	1.0	23.8
7	0.643	1.317	0.537	1.0	23.9
8	0.599	1.190	0.486	1.0	24.0
9	0.563	1.052	0.445	1.0	24.2
10	0.608	1.222	0.501	1.0	24.1
$-\bar{X}$	**0.614**	**1.213**	**0.495**	**1.0**	**24.2**

Table 4
Parameters of the stress relaxation curves obtained for material DM_9895_Shore95, samples printed vertically

Test No.	σ_0 [MPa]	σ_1 [MPa]	σ_2 [MPa]	t_1 [s]	t_2 [s]
1	0.901	1.800	0.532	0.7	21.5
2	0.900	1.915	0.002	0.6	22.0
3	0.869	1.753	0.525	0.7	22.1
4	0.826	1.900	0.590	0.8	25.9
5	0.889	2.083	0.616	0.8	21.9
6	0.854	2.149	0.605	0.7	21.2
7	0.932	2.207	0.630	0.7	21.0
8	0.917	2.158	0.631	0.8	21.7
9	0.893	2.082	0.613	0.8	22.0
10	0.909	2.155	0.619	0.7	20.7
$-\bar{X}$	**0.889**	**2.020**	**0.536**	**0.7**	**22.0**

the basis of which the uncertainty of the estimated parameters was calculated.

Having determined the average values of parameters σ_0, σ_1, σ_2, t_1, t_2, one can save (8) in a detailed form for individual materials. For DM_8515_Grey35 vertically printed samples, it is as follows:

$$\sigma(t) = 16.645 + 2.411e^{\frac{-t}{22.2}} + 6.438e^{\frac{-t}{376.1}}. \tag{9}$$

For DM_8515_Grey35 horizontally printed samples, it takes the form below:

$$\sigma(t) = 15.578 + 2.03e^{\frac{-t}{20.4}} + 4.537e^{\frac{-t}{352.2}}. \tag{10}$$

For DM_9895_Shore95 vertically printed samples, it is as follows:

$$\sigma(t) = 0.614 + 1.213e^{-t} + 0.495e^{\frac{-t}{20.4}}. \tag{11}$$

And for DM_9895_Shore95 horizontally printed samples, it takes the form below:

$$\sigma(t) = 0.889 + 2.02e^{\frac{-t}{0.7}} + 0.536e^{\frac{-t}{24.2}}. \tag{12}$$

Figures 9 and 10 display the chart of function (9, 10, 11 and 12).

Standard uncertainty for parameters σ_0, σ_1, σ_2, t_1 and t_2 in individual series of measurements, calculated by means of the A type method, is as follows:

$$u_A = \sqrt{\frac{1}{n(n-1)}\sum_{i=1}^{n}(x_i - \bar{x})^2}, \tag{13}$$

where: n – number of measurements (number of samples made of individual materials),
x_i – individual result in the series,
$\bar{x}$ – arithmetic average of individual parameters obtained as a result of approximation.

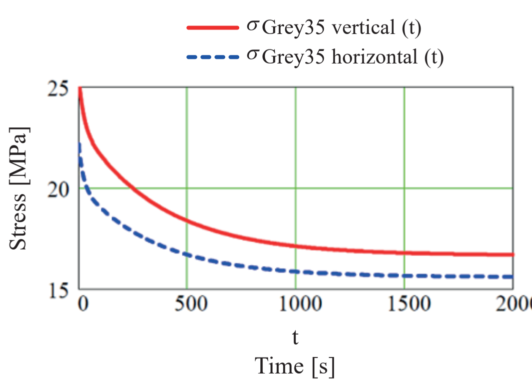

Fig. 9. Stress relaxation curves of digital material fitting on the basis of (9) and (10) for DM_8515_Grey35

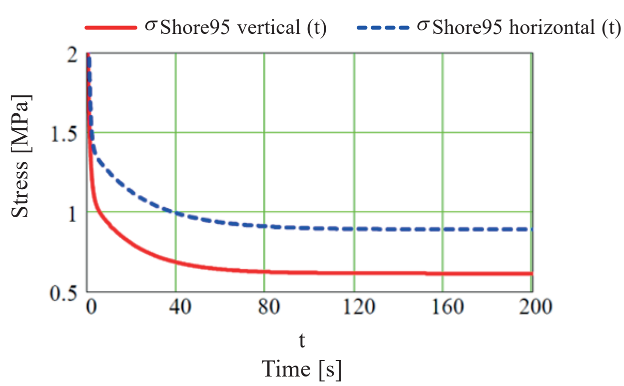

Fig. 10. Stress relaxation curves of digital material fitting on the basis of (11) and (12) for DM_9895_Shore95

Due to the fact that the sample size is less than thirty (n = 10), expanded uncertainty was estimated using the Student's distribution:

$$u_{CA} = k_p u_A, \tag{14}$$

where: k_p – an expansion factor that assumes a value of 2 for the confidence level $p = 0.95$.

The results of calculations of standard deviation and uncertainty of measurements are presented in Table 5.

Table 5
Results of calculations of uncertainty of approximation

Type of material	Uncertainty of approximation u_A				
	$u\sigma_0$ [MPa]	$u\sigma_1$ [MPa]	$u\sigma_2$ [MPa]	ut_1 [s]	ut_2 [s]
DM_8515_Grey35 vertical	0.328	0.064	0.201	0.3	4.6
DM_8515_Grey35 horizontal	0.397	0.087	0.158	0.3	6.0
DM_9895_Shore95 vertical	0.013	0.039	0.014	0.0	0.1
DM_9895_Shore95 horizontal	0.010	0.052	0.061	0.02	0.5

The values of uncertainty of approximation contained in Table 5 show that a good fit of equation (8) to the experimental curves has been achieved. Small uncertainty values calculated for individual parameters of relaxation curves result from the fact that these parameters, as shown by formulas (5) and (6), depend on material properties such as elastic modules and viscosity coefficients represented in equation (8) by parameters σ_1, σ_2, t_1 and t_2.

4. Conclusion

A very good fit of the adopted five-parameter Maxwell-Wiechert model to the experimentally obtained stress relaxation curves of selected digital materials obtained with additive technology was achieved. This is evidenced by the small values of the estimated uncertainty of approximation of individual model parameters.

Having average values of σ_0, σ_1, σ_2, t_1 and t_2 parameters calculated with high accuracy, one can use the equations describing individual materials. The description of experimental stress relaxation curves using the adopted model is of great importance due to the physical nature of the parameters obtained. It also extends the possibilities of modeling materials, especially digital ones, for the relaxation properties assumed, or, more precisely, for the assumed parameters of the adopted rheological model describing a given material.

The research presented herein also showed the anisotropy of the properties due to the direction of the printout, i.e. setting the object on the bild tray platform of the printing machine. One can see the difference in the mean values obtained for model parameters in the case of samples printed vertically and those printed horizontally.

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