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Test Method

Determination of the viscoelastic properties of elastomeric materials by the dynamic indentation method

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Abstract

In this paper the dynamic indentation test method, which is not often used, is discussed. The goal of the paper is to consider the possibility of applying a dynamic indentation test method to investigate rubber materials. The basic equations for the determination of the viscoelastic characteristics of a material are presented and the experimental set-up is described. The Kelvin–Voigt model is used to describe the characteristics of a material. Experimental and theoretical curves for velocity, force and penetration in the indentation process are compared for rubbers with different hardness. A semi-empirical relationship between the Shore hardness and the rigidity *c* was derived.

Keywords: Indentation; Impact testing; Kelvin–Voight model; Rubber; Hardness; Rigidity

1. Introduction

Viscoelastic materials like rubber, latex and composites, can be applied in a wide range of situations with variable pressures and deformations. In this context the behavioural study of these materials in such conditions is of great interest. The most obvious and convenient way of obtaining data on material properties is by finding dependencies between the load and strain of a material. These dependencies, corresponding to various kinds of stress levels, reflect the ability of a material to resist straining. Furthermore, by predicting possible behaviour in use, these dependencies can be used for quality control and development of new materials. Moreover, they define advantages and disadvantages of a material in comparison with previously created materials.

One of the test methods for a viscoelastic material, which simulates conditions of variable pressures and deformation, is indentation [1,2,3]. This method enables one to study the resistance to straining of a material under an active contact force. The load in the indentation method can be dynamic or static, which allows the method to establish a response of a material in a wide range of strain rates.

An estimate of the properties of materials is obtained by determining the hardness of a material using static indentation [4,5]. This measurement is the simplest method to define the elastic properties and the ability to resist the impression under the action of a contact force. Methods can differ from each other in the value of and the way of applying the load and by the form of the tip of the indenter (sphere or cone). Because of the historical importance of the Shore hardness measurement, it is widespread in industry [6]. Nevertheless, the Shore method has some disadvantages: special samples (at least 6 mm thick) are needed, the properties of these samples may differ from those of finished products and the method only provides information about the hardness of a sample.

Dynamic indentation has a number of advantages over static indentation. In conditions in which the velocity of the indenter cannot be neglected, elastomers not only

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show elastic behaviour but also viscous behaviour. The determination of these properties gives more complete information about the behaviour of a material under various modes of loading. Moreover, dynamic indentation is a non-destructive method, which gives an opportunity to carry out measurements with low loading and to test thin layers of the product without utilisation of special samples. However, there are some difficulties in using dynamic methods for determining the viscoelastic properties of polymers. The response of the material to pulse loading is determined by the physical nature of the material and by the real history of the loading process. Universal equations, which allow us to describe the rheological and time-dependent behaviour of a viscoelastic half space under impact loading, do not exist and therefore simplified models or empirical equations are usually used to compute the response of materials in these conditions. The main consideration for using possible models is how well they characterise material behaviour in specific conditions. Utilisation of such models is a compromise between simplicity and reality in describing the straining process. Another interesting question, which is unfortunately not yet solved, is if a relation exists between the dynamic and static characteristics of a material, for example for hardness.

2. Theory

Usually the viscoelastic properties are conveniently described by a phenomenological model of masses, springs and dampers. The elastic properties of a material are simulated by a spring, which has a storage capacity and a perfect memory and for which stress σ and strain ε are proportional in accordance with Hooke's law. The viscous properties are represented by a damper, for which the stress σ is proportional to the strain rate $\dot{\varepsilon}$. A damper has no storage capacity, i.e. it dissipates all energy, and no memory at all. The basic viscoelastic model, applicable to our area of interest, is the Kelvin–Voigt model for viscoelastic solids. This model describes the dynamic influence of a rigid body on a material when the strain recovers in time. The constitutive relation for a Kelvin–Voigt model (represented as a spring and damper in parallel) is given by

$$\sigma = E\varepsilon + \gamma\dot{\varepsilon}, \tag{1}$$

where E is the elastic modulus and γ is the viscosity of the material.

Let us consider a case in which the indenter and the material can be represented by a Kelvin–Voigt model with only one degree of freedom as shown in Fig. 1.

The behaviour of the model during the period of contact is described by the linear differential equation:

$$m\ddot{\alpha} + \eta\dot{\alpha} + c\alpha = 0, \tag{2}$$

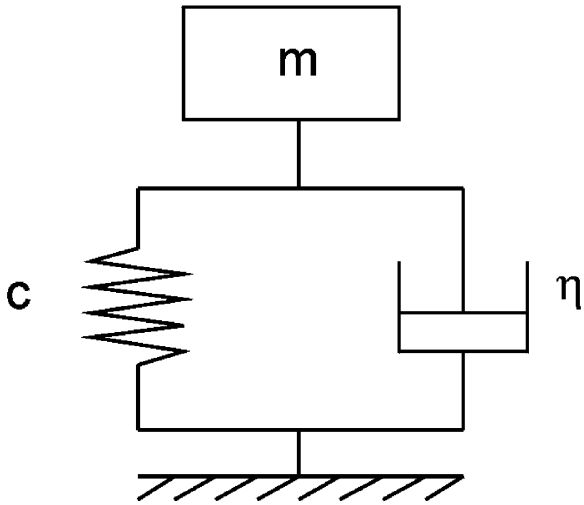

Fig. 1. The mechanical (Kelvin–Voigt) model of the indentation of the rigid indenter into the viscoelastic material.

where m is the mass of the indenter, η is the viscosity, c is the rigidity of the spring and $\alpha(t)$ is the penetration of the indenter into the tested material.

The first term of the equation describes the effects of inertia, the second term describes the effects of viscosity, which are proportional to the indenter's velocity, and the last term describes the effects of elasticity. Both η and c will depend on the geometrical form of the indenter tip.

From the moment the indenter touches the surface of the tested sample, the kinetic energy of the indenter is transferred into potential energy. During this stage, which will be called the active stage, the energy is conserved by the sample until the velocity of the indenter is equal to zero. During the passive stage, in which the rebound of the indenter occurs, the reserved energy is transferred into kinetic energy of the indenter and, in the absence of energy losses, the rebound velocity will be equal to the initial speed of the indenter before the impact.

For a periodic movement during impact, the angular velocity ω can be expressed as:

$$\omega = 2\pi \cdot f_{\text{cycle}} = \frac{2\pi}{T_{\text{cycle}}} = \frac{\pi}{2 \cdot t_{\text{active}}}, \tag{3}$$

where t_{active} is the duration of the active stage of the impact.

For a system with low damping $\left(\frac{c}{m} > \frac{\eta^2}{4 \cdot m^2}\right)$, the general solution of the equation of motion will be

$$\alpha(t) = C_1 \cdot e^{-p \cdot t} \cdot \cos(\omega \cdot t) + C_2 \cdot e^{-p \cdot t} \cdot \sin(\omega \cdot t), \tag{4}$$

where $p = \frac{\eta}{2 \cdot m}$ and $\omega = \sqrt{\frac{c}{m} - \left(\frac{\eta}{2 \cdot m}\right)^2}$.

Using the initial conditions

$$\alpha_{t=0} = 0, \frac{d\alpha}{dt}\bigg|_{t=0} = V_0,$$

the constants in the equation can be derived:

$$C_1 = 0, C_2 = \frac{V_0}{\omega}.$$

In accordance with Eq. (4), the penetration and the velocity of the indenter and the force exerted by the indenter can be written as

$$\alpha(t) = \frac{V_0}{\omega} \cdot^{-p \cdot t} \cdot \sin(\omega \cdot t), \tag{5}$$

$$V(t) = \frac{d\alpha}{dt} = -\frac{V_0 \cdot p}{\omega} \cdot e^{-p \cdot t} \cdot \sin(\omega \cdot t) \tag{6}$$

$$+ V_0 \cdot e^{-p \cdot t} \cdot \cos(\omega \cdot t),$$

$$F(t) = -m \cdot \int \alpha(t) dt \tag{7}$$

$$= \frac{m \cdot V_0}{\omega} \cdot \frac{e^{-p \cdot t} \cdot [p \cdot \sin(\omega \cdot t) + \omega \cdot \cos(\omega \cdot t)]}{p^2 + \omega^2}.$$

For a system with low damping, the angular velocity ω can be approximated by the natural frequency ω_0 of the system, determined from the equation of motion

$$\omega \approx \omega_0 = \sqrt{\frac{c}{m}}. \tag{8}$$

Now, using Eqs. (3) and (8), an equation can be extracted for the rigidity c expressed in the mass m and the active-time t_{active} :

$$c = \frac{m\pi^2}{4t_{\text{active}}^2} = m\omega^2. \tag{9}$$

To determine the other constant of the applied model η, the logarithmic decrement method was used. The logarithmic decrement was calculated from the natural logarithm of the ratio of the amplitudes, i.e. two adjacent peak values of the velocity, of any two oscillations

$$\Delta = \ln\left(\frac{V}{V_0}\right). \tag{10}$$

For a system with low damping the logarithmic decrement for a Kelvin–Voigt model is

$$\Delta = \pi \cdot \frac{\eta \cdot \omega}{c}. \tag{11}$$

After substitution and rewriting of the equation for the angular velocity, an equation can be derived for viscosity η expressed in the logarithmic decrement Δ, the mass m and the angular velocity ω

$$\eta = \frac{\Delta \cdot c}{\pi \cdot \omega} = \frac{m \cdot \omega \cdot \Delta}{\pi}. \tag{12}$$

3. Experimental set-up

For measuring the dynamic characteristics, the IVUS-device was used which was developed at the Academy of Sciences of the Institute of Applied Physics in Minsk, Belarus. A schematic drawing of the experimental set-up of the dynamic indentation test is shown in Fig. 2.

The apparatus consists of an impact device with indenter (1) with spherical tip attached to a rotating lever (3) with a permanent magnet (2) mounted on top of the indenter. The characteristics are measured during the impact stage by a stationary inductive coil (4), in which the permanent magnet (2) induces an electromagnetic force (EMF) $U = f(t)$. This EMF is directly proportional to the velocity of the indenter *V(t)*. The electrical parts of the device (5) represent a circuit for A/D conversion of the signal. Further processing of the signal is conducted with the use of specially developed algorithms and with the help of a computer (6).

The calibration factor of the signal induced in the coil is easily determined from the data at the moment of first contact between the indenter and the test sample

$$K_v = \frac{U_{\max}}{V_0}, \tag{13}$$

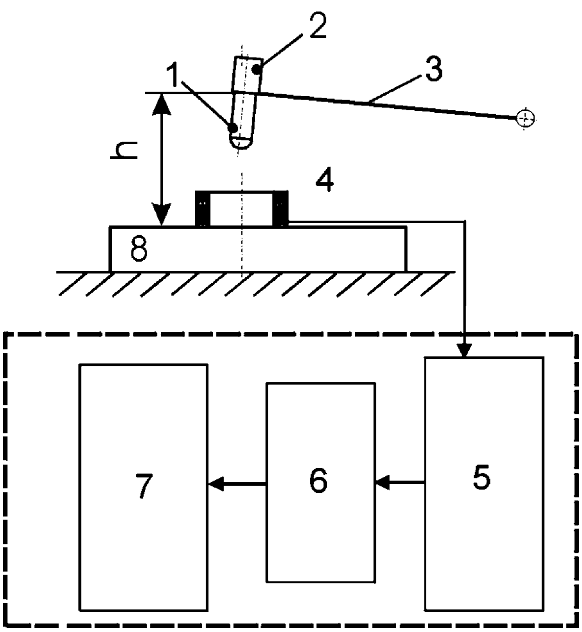

Fig. 2. Schematic arrangements of the used experimental components. 1—indenter, 2—magnet, 3—lever, 4—inductive coil, 5—analog to digital converter, 6—computer, 7—printer, 8—sample.

where U_{max} is the EMF before impact.

The initial velocity is determined from the energy conservation law: $V_0 = \sqrt{2gh}$, where h is the initial fall height (Fig. 2).

The velocity–time dependence is now described as a relation between a calibration factor and the measured voltage-signal as:

$$V_i(t) = \frac{U_i(t)}{K_v}, \quad (14)$$

where i is the index of the discrete signal.

The contact force $P(t)$, exerted on the material during impact, can be determined by numerical differentiation of the velocity signal $V(t)$, because of momentum conservation (with m the mass of the indenter):

$$P(t) = m\frac{\Delta U_i(t)}{K_v \Delta t_i}. \quad (15)$$

The indenter displacement is found by numerical integration of the velocity signal $V(t)$:

$$\alpha(t) = \frac{\sum U_i(t)\Delta t_i}{K_v}. \quad (16)$$

The IVUS-device has certain advantages for determination of parameters over similar devices (instrumented impact test). Only the velocity of the movement must be measured, the application of a displacement gauge or an accelerometer is not necessary. Furthermore, the indenter can be replaced quite easily, which allows one to carry out a large number of experiments with various shapes and weights of the indenter to test thin polymer samples. In our case the experiments were carried out with an indenter with a mass of $m = 3.5$ g, an initial velocity of $V_0 = 1$ m/s and a tip radius of $R = 0.77$ mm.

A range of natural isoprene, butadiene-nitrile and some other synthetic rubbers with different viscous and elastic properties were tested. The compounds contained different amounts and type of plasticizers and extenders but the manufacturer did not provide more details of the compositions. Some of these rubbers showed nearly elastic behaviour, others had a significant time-dependent character.

For the purpose of the tests, about 75 rubber samples were tested with Shore hardness from 32–95 HS and different rebound characteristics. The shape of the samples was a standard disk with a diameter of 60 mm and a thickness of 6 mm.

4. Results and discussion

Fig. 3 shows typical curves obtained with these rubber samples. These curves clearly show the viscoelastic character of deformation. As can be seen from Fig. 3, there is a shift θ between force and penetration in the impact process. Also, the sample has a residual deformation α_{res}, which is recovered in a certain period of time after impact.

The correctness of the application of the Kelvin–Voigt model for the description of the indentation process has been considered. The parameters of the model (c,η) were determined by using Eqs. (9) and (12) in order to check the correlation between the theoretical model and the experimental results. For this, seven samples with different characteristics have been selected as shown in Table 1. These samples were chosen because of their different values of hardness, creep and elastic properties. Correlation between the theoretical and experimental values for other intermediate samples had a similar character.

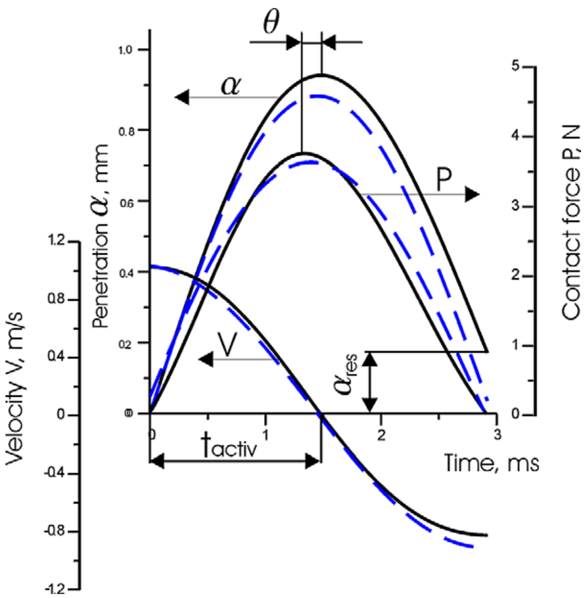

Fig. 3. Curves for velocity, penetration and force obtained on rubber (sample no. 1, Table 1). The dotted lines are theoretical predictions, the solid lines are experimental observations.

Table 1
Shore characteristics for seven different samples

Sample	Shore hardness HS		
	1 s	3 s	15 s
1	35	35	35
2	52	48	40
3	60	57	54
4	58	58	58
5	72	70	70
6	87	85	85
7	95	95	95

The theoretical and experimental characteristics for velocity, force and indentation are plotted in Fig. 3 (sample no. 1 of Table 1) and Fig. 4 (sample nos 2–7 of Table 1).

The correspondence between the experimental results (solid line) and the theoretical prediction (dotted line) for synthetic and natural rubbers is shown. It can be concluded from Fig. 3 and Fig. 4 that the best correspondence between the theoretical and experimental data is obtained for rubber samples with low hardness. The experiments which were carried out have shown that the lower the hardness of the rubber the better the convergence between the theoretical and experimental values. On the other hand, if the samples show pronounced creep (Table 1) this convergence is worse. This is valid for the whole range of hardness and especially between the theoretical and experimental curves for penetration.

As can be seen from Fig. 4, the theoretical equation does not describe residual deformation well. Especially for hard samples, possible plastic deformation is not described in a correct way and results in a large derivation from the theoretical prediction. A second disadvantage of the Kelvin–Voigt model is the fact that at $t = 0$ the force is not equal to zero.

Another interesting question in our investigation is the derivation of a relation between the parameters of the Kelvin–Voigt model and standard characteristics used in industry, like Shore hardness. This hardness was related to the elastic properties of materials in a number of papers [7,8]. An acceptable model parameter (2), which describes elastic properties, is the rigidity c, which is directly obtained from the equation of the indenter motion [9].

Careful consideration of the experimental results has shown that the measured and calculated rigidity has a correlation with the Shore hardness with a satisfactory correlation coefficient. By least squares estimation, a regression equation can be found with a negative, or inverse, relationship between the Shore hardness and the square of the rigidity. This semi-empirical correlation can be given by:

$$\mathrm{HS} = 99{,}43 - \frac{4150}{C^{0.5}}. \tag{17}$$

This dependence is shown in Fig. 5. After examination of the confidence interval, we conclude this is too large to determine the Shore hardness accurately (the confidence interval is about 10 HS, residuals can even reach 40%). One of the reasons for this observation is the fact that this graph was plotted including all tested samples. After removal of the samples with large creep character-

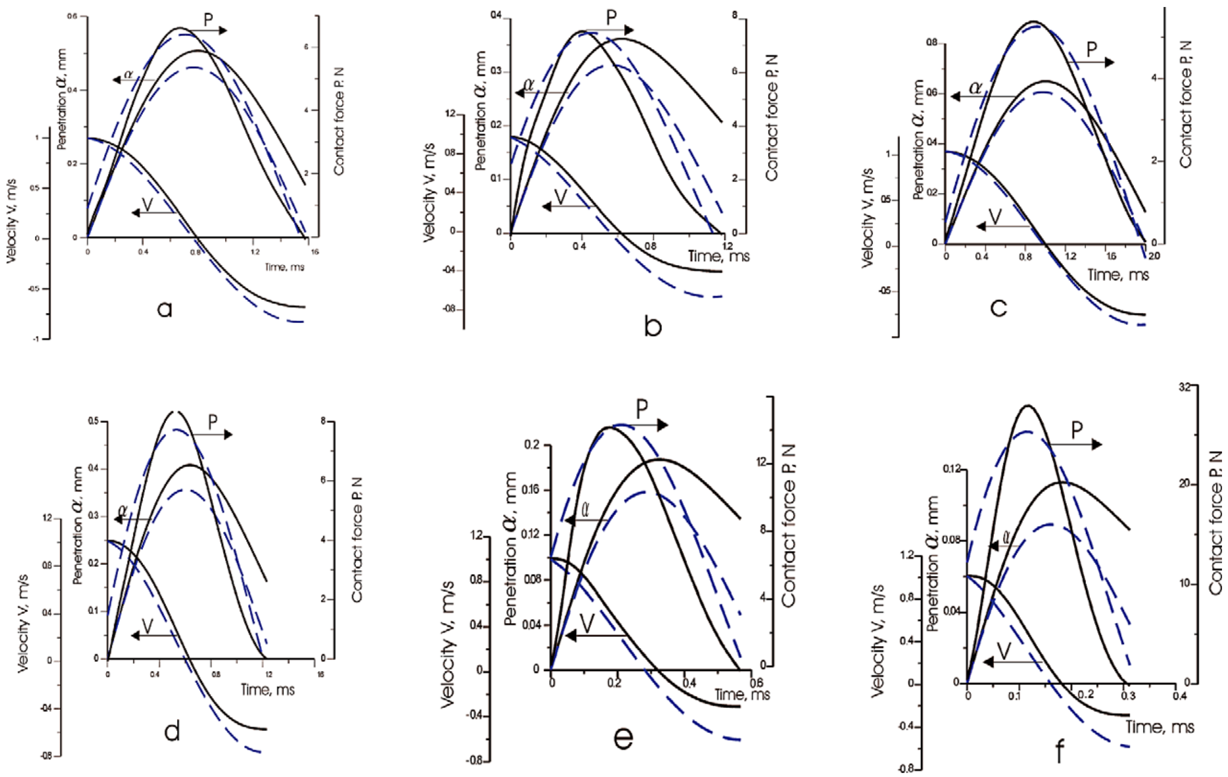

Fig. 4. Typical curves for velocity, penetration and force for rubber samples with different hardness and creep characteristics. Graphs a–f represent respectively samples 2–7 in Table 1.

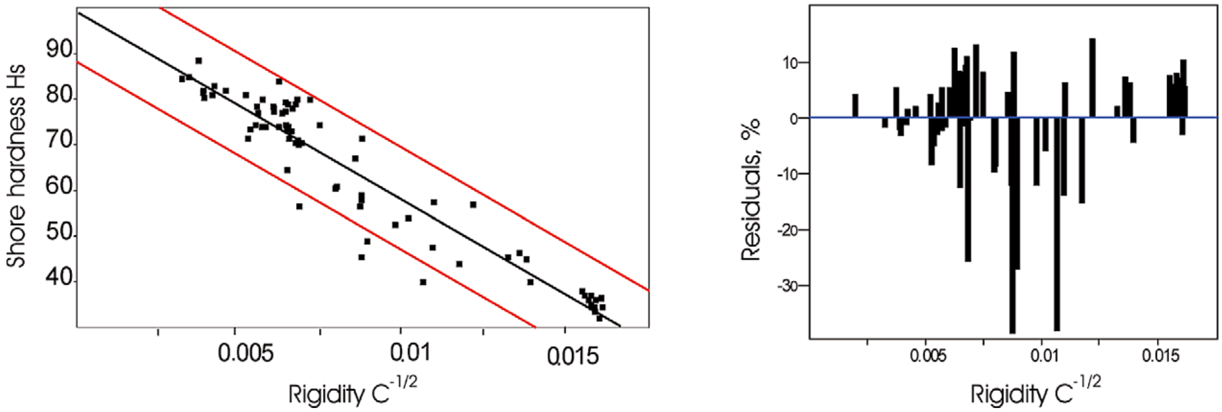

Fig. 5. Dependence Shore hardness—rigidity and residuals—rigidity for all samples.

istics, the confidence interval is better and the residuals are only about 10% of the ideal value according to the correlation shown in Fig. 6. For these samples, we derive another empirical relation for HS with a good regression correlation coefficient, $R^2 = 0.968$

$$HS = 102,17 - \frac{4200}{C^{0.5}}. \tag{18}$$

This relation can be used as a first approximation to a Shore hardness determination. For the remaining samples, we need to find a more appropriate model which will give proper weight to the creep factor.

5. Conclusion

Dynamic indentation is a robust method that can be used to measure viscoelastic properties of polymeric materials. The method does not demand application of a displacement gauge or an accelerometer. At the same time, with the help of an algorithm that was developed, it allows one to determine not only the velocity of the indenter, but also the contact force on and the resulting displacement of the sample. The relative simplicity of the device allows one to use it not only in laboratories, but also in a practical environment with only a notebook computer at ones disposal.

From the results obtained one can conclude that the dynamic indentation method can be applied to evaluate both the elastic and viscous properties of a material. It is shown that the Kelvin–Voigt model describes the response of rubber materials under impact indentation in engineering applications. For low hardness rubbers without significant creep properties (up to 60 HS), the model describes the process with a satisfactory reliability. However, for higher values of the Shore hardness and for more extensive creep, it is shown that there is less convergence between the theoretical and experimental results.

The experiments which were carried out have shown that the method can be applied for the Shore hardness determination for vulcanised natural and synthetic rubber, without significant creep properties. A semi-empirical dependence is derived which relates dynamic rigidity and Shore hardness.

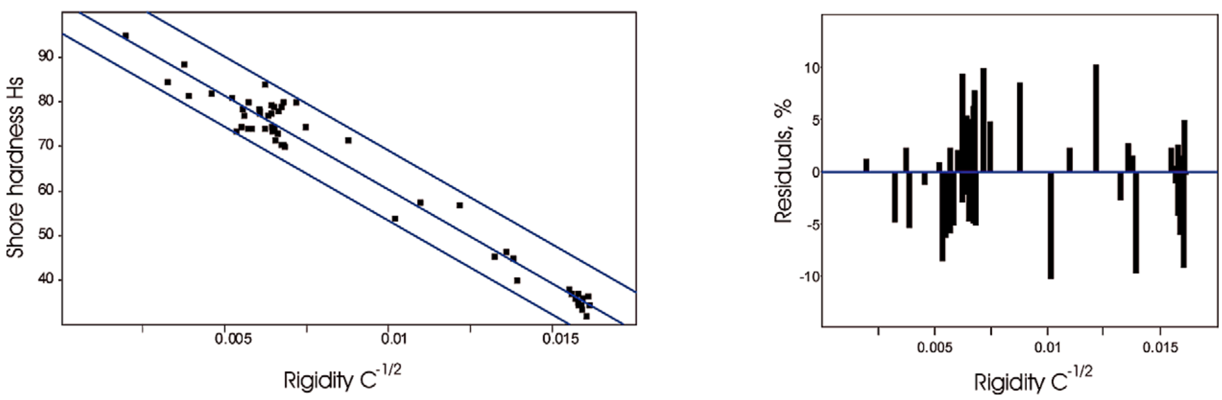

Fig. 6. Dependence Shore hardness—rigidity and residuals—rigidity for selection of samples.

Further research has to be carried out to determine a satisfactory relationship for the samples with a reasonable amount of creep. The theoretical model should be adjusted to take into account this time-dependent effect. Also, it would be interesting to investigate the relationship between the rigidity *c* and the elastic modulus *E* from measurement of modulus data on the same samples.

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