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The Apparatus for Thermomechanical Analysis of Clay-based Ceramics

Tomáš Húlan*, Igor Štubňa, Omar Al-Shantir, Anton Trník

Constantine the Philosopher University, Department of Physics, A. Hlinku 1, 949 74 Nitra, Slovakia, thulan@ukf.sk

Abstract: A dynamic thermomechanical analysis (D-TMA) apparatus is described for measuring the resonant frequency of the flexural vibration and the internal damping of the sample using the impulse excitation technique (IET). Since the measurement is conducted at temperatures up to 1250 °C, an electromagnetic impulser is used for excitation. The free vibrations are registered by an electret microphone, stored and then converted into a frequency spectrum using the fast Fourier transform, from which the resonant frequency can be found. The furnace is built from refractory porous alumina bricks and alumina fiber pads. The heating elements are four SiC rods connected to a temperature controller. The temperature is measured with a Pt-PtRh10 thermocouple in close proximity to the sample.

Keywords: Impulse excitation technique, resonant frequency, internal damping

1. INTRODUCTION

Dynamic thermomechanical analysis (D-TMA) or modulated force thermomechanical analysis (mf-TMA), which investigates the elastic or damping mechanical properties in chosen temperature regimes (mostly linear heating/cooling or isothermal heating), is mostly based on resonant techniques. These important mechanical quantities are determined by the structure and microstructure of the material as well as by the external conditions [1]. Therefore, they can be used for indirect study of porosity, texture, intergranular phases, composition, cracking, phase changes, sintering, hardness, apparent density, mechanical strength, and others.

Several experimental techniques have been developed to determine Young's modulus (YM) of solids. Methods that meet the requirements of TMA use a resonant frequency [2]-[5] or sound velocity [2] as the basis for determining YM. The resonant methods are usually based on the flexural vibration of the sample. This vibration can be easily excited, has a large amplitude of deflection, and is reliable at high temperatures. The samples have a simple geometry, most often a cylinder or a prism with a uniform cross-section.

Two techniques are used to determine the resonant frequency. Both of them use the same theoretical basis for flexural vibration and give the same results of YM [6], [7]. The measured quantities are the dimensions and mass of the sample and the resonant frequency.

The first technique, called the sonic resonance method, is based on forced vibration of the sample at a known frequency [8]. This method is superseded by the impulse excitation technique (IET), which is most commonly used today due to advances in computer technology. The IET is based on the free vibrations of the sample after being exposed to a mechanical impulse [3], [9]-[11]. The initial response decreases according to the time exponential function from which the coefficient of internal damping can be determined. The free vibrations are registered by the sensor, stored, and then analyzed using the fast Fourier transform. Its result is a frequency spectrum of the sample vibrations, where the resonant frequency of the fundamental mode of the vibrations can be found. The mechanical impulse can be performed manually with a small hammer if the measurement is conducted at room temperature. If the measurement is conducted at higher temperatures, steel or ceramic balls dropped on the sample or electromagnetic impulser are used to excite the flexural vibration.

This paper aims to describe the apparatus used for D-TMA of clay-based ceramics at the working temperature of 1250 °C.

2. THEORETICAL BASE OF D-TMA

YM can only be measured indirectly. To determine its value, three physical quantities have to be measured and substituted into a relevant formula. These are the mass and dimensions of the sample and its resonant frequency. For the flexural vibration, YM can be calculated by the formula [2]-[4].

$$E = \left(K\frac{l^2 f_0}{d}\right)^2 \rho T, \quad (1)$$

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*Corresponding author: thulan@ukf.sk (T. Hulan)

where E is YM, f_0 is the resonant frequency of the fundamental mode, ρ is the bulk density, l is the length and d is the diameter or thickness of the sample. The values of the constant K are:

K = 1.12334 for a circular cross-section and the fundamental resonant frequency,

K = 0.97288 for a rectangular cross-section and the fundamental resonant frequency.

The correction coefficient $T = 1$ if $l/d \geq 20$. If not, correction coefficient T must be calculated from formulae given in [3], [4] or it can be found in tables in [2]. To obtain T, Poisson's ratio of the measured material and dimensions of the sample must be known.

The logarithmic decrement δ can be determined simultaneously with the resonant frequency from the relation [12].

$$\delta = \frac{\pi \Delta f}{\sqrt{3} f_0}, \tag{2}$$

where Δf is the width of the resonant peak in its half height.

3. Apparatus for D-TMA

Two apparatuses – one for a room-temperature measurement and one for a high-temperature measurement (i.e., for D-TMA) – were designed and built based on the IET. The supports of the sample for the room-temperature measurements are made of soft foam pads and vibration is excited by a blow of the small hammer (steel ball of Ø4 mm glued on a thin plastic/wooden stick). The sample is placed on supports 0.224 l away from both ends of the sample, i.e., in the nodal points of the fundamental mode of the flexural vibration. This simple apparatus is used to determine the fundamental resonant frequency of the sample when new material is investigated.

High-temperature measurements require a special design. The first solution of the apparatus for D-TMA made in the authors' laboratory was described in [13], [14]. A principal schematic of the new D-TMA apparatus, which measures the resonant frequency of the flexural vibration of the sample, is shown in Fig. 1. Almost all parts of the original apparatus were changed: sample holder, impulser, temperature controller, and operating software.

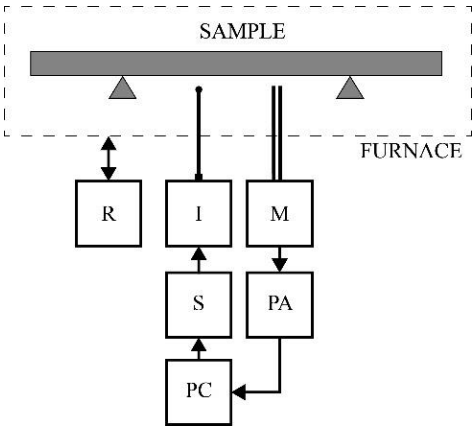

Fig. 1. The schematic of the high-temperature apparatus for D-TMA: I – impulser, M – microphone, PA – preamplifier, PC – computer, R – temperature controller, S – programmable power supply.

A. Impulser

To excite D-TMA to vibrate at higher temperatures , the sample must be hit with an impulser in the area between the nodal points, i.e., between the supports, as shown in Fig. 1. We used an electromagnetic impulser shown in Fig. 2. It consists of an alumina tube with an inner diameter of 6 mm, in which a ∅3 mm alumina rod is moved upward in the vertical direction by an electromagnet. Its coil is wound on the outer side of the tube. The parameters of the coil are as follows: number of turns = 500; coil diameter = 8 mm; coil height = 50 mm; wire diameter = 0.84 mm; number of layers = 9; total resistance = 0.54 Ω. A permanent neodymium magnet ∅5 × 11 mm is glued to the bottom end of the alumina rod and represents a movable core of the electromagnet. The second permanent magnet is located at the bottom of the alumina tube. The magnets are aligned with the same poles to each other, creating a repulsive force that allows a soft, contactless, and noiseless settling of the rod to its initial position.

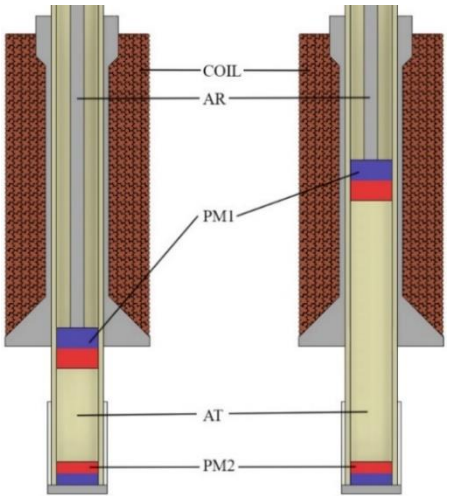

Fig. 2. The schematic of the impulser with the current of the coil off (left) and on (right): AR – alumina rod, AT – alumina tube, PM1 – permanent magnet, the core of the electromagnet, PM2 – permanent magnet, COIL – the coil of the electromagnet.

The impulser is controlled by a computer program and the mechanical impact is executed every $T = t_1 + t_2$ seconds. The program sends a command to the AxioMet AX 3005PQ programmable power supply (5 A, 40 V), which operates in a constant current mode. The coil current is zero for the period t_1, i.e., the rod is in its initial low position (left image in Fig. 2). The rectangular current impulse (from 1 A to 3 A) keeps the impulser ~5 mm below the sample for the period t_2. When the impulse is switched on, the rod moves upward and passes through this equilibrium position due to inertia, hitting the sample only once. During the period t_2, the vibration (sound) of the sample is recorded. Then the coil current is set to zero, returning the impulser to its initial position.

The program adjusts the current impulse to achieve the lowest energy of hit needed to detect the vibrations of the sample. When the output of the preamplifier is below the selected threshold, the current impulse is slightly increased. The period T is usually 10, 15 or 30 seconds, depending on the heating rate. The lower limit of T is 3 s. The timing of all

operations is crucial. The program starts recording the sound signal a few milliseconds before the impulse and stops recording after the preset time, usually 0.5, 1 or 2 seconds. The duration of the recording depends on the material of the sample. If the damping is strong, a short time interval can be used.

The current design of the impulser solves the problem with excess friction between the alumina tube and the impulser rod, which arose when the rod and the guide aperture in the furnace were not coaxial. This sometimes happened as a result of thermal deformations of the furnace.

B. Sample holder

It is desirable to have the sample supported at the nodal points of the fundamental mode of the flexural vibrations. Since the mechanical impulse is directed vertically upwards, the sample must be reliably fixed with wires. This is done with Kanthal A1 wire ∅0.3 mm. The best results were obtained when the length of the sample was between 100 mm and 120 mm. The sample is placed in a horizontal position on the wires in its nodal points and two other wires fix the sample to avoid vertical movement (Fig. 3). Two heatproof steel cylindrical weights (∅20 × 90 mm) provide sufficient force to fix the sample. The sample holder allows more flexible fixation of the sample, reducing external damping of the sample vibration.

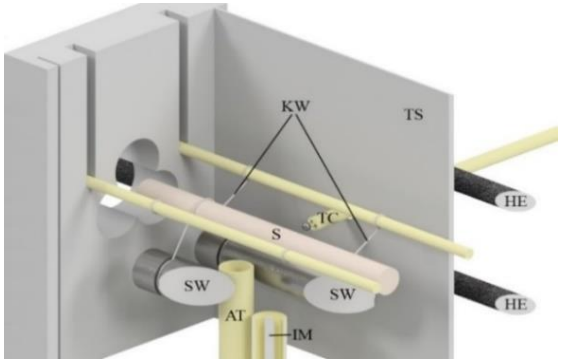

Fig. 3. Interior of the furnace: TS – radiation sheets, HE – heating elements, KW – Kanthal wires, SW – steel weights, AT – alumina tube (sound guide), IM – impulser, TC – thermocouple.

C. Furnace and temperature control

The furnace is constructed of refractory porous alumina bricks and alumina fiber pads. The working volume is 120 × 120 × 250 mm^3. The heating elements are four SiC rods connected to a temperature controller. The temperature is measured with a Pt-PtRh10 thermocouple in the close proximity to the sample. To improve the homogeneity of the temperature field, two radiation shields (alumina plates) are located between the sample and the heating elements.

The apparatus can operate in various temperature regimes, including heating and cooling at rates up to 20 °C/min or isothermal heating at selected temperatures up to 1250 °C. Temperature control is performed by the temperature controller Clasic Clare (Czech Republic) without the communication with PC. The actual temperature during measurement is recorded by Keysight DAQ970A, which features cold junction compensation and voltage-to-temperature conversion for Pt-PtRh10 thermocouples.

D. Sensor and preamplifier

The electret microphone is used to record the sound response of the sample after impulse excitation. Since the microphone must be located outside the furnace, an alumina tube ∅6 × 230 mm is used as a waveguide between the microphone and the sample (Fig. 1 and Fig. 3). There is a gap of ~5 mm between the alumina tube and the sample so that the measurement of the sample vibrations is contactless. A voltage signal from the microphone is weak, so the microphone is connected to an input of the CanaKit UK 151 preamplifier, which was designed based on the LM386 amplifier integrated circuit. The output of the preamplifier is connected to the microphone input of the computer.

The computer sound card (16-bit) is used for data recording. The sampling frequency can be set by the user, but normally 44.1 kHz or 96 kHz is used. The measured signal is stored in a time domain (upper image in Fig. 4) and displayed. In the next step, the signal is transformed into a frequency domain using the fast Fourier transform implemented in MATLAB® (see lower image in Fig. 4). The frequency spectrum is then analyzed to determine the resonant frequency and the logarithmic decrement of the vibration (as a measure of the internal damping).

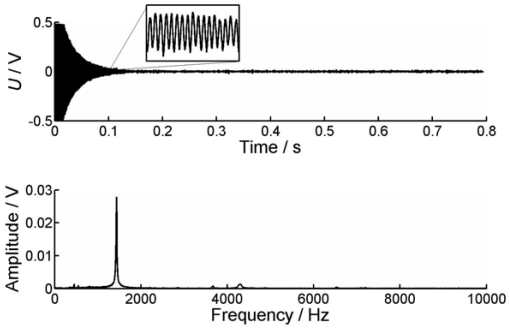

Fig. 4. A typical response of the sample to the mechanical impulse, where the output voltage of the microphone is visible in the upper picture and its frequency spectrum is visible in the lower picture.

E. Signal processing

The most important requirement for accurate determination of the resonant frequency of the flexural vibration f_0 and the logarithmic decrement δ is to excite the sample to vibrate in such a way that only the first mode of its out-of-plane flexural vibration is excited (Fig. 5(a)). In addition, external noise (for example, from the movement of the impulser) must be as low as possible. In practice, and especially at higher temperatures, other modes of sample vibration are often excited and the noise from the supporting system and the impulser movements cannot be neglected. However, probably the most significant factors for the correct determination of the resonant frequency are the inherent properties of the sample, particularly its internal damping. The higher the internal damping, the more difficult it is to obtain a reliable sound signal from the vibration of the sample. In extreme cases of high damping, only a few oscillations are registered before the damping drops below the noise level (Fig. 5(b)). Despite the low-quality signal, reliable f_0 and δ can be obtained if appropriate processing of the signal is used. We have found these approaches to be useful:

Determining δ from the width of the resonant peak rather than local maxima on the sound signal. This is much more reliable for highly damped signals (Fig. 5(b)).

Using only a part of the signal that corresponds to the actual vibration of the sample (green lines in Fig. 5) to evaluate the spectrum will improve the spectrum, especially in case of high damping.

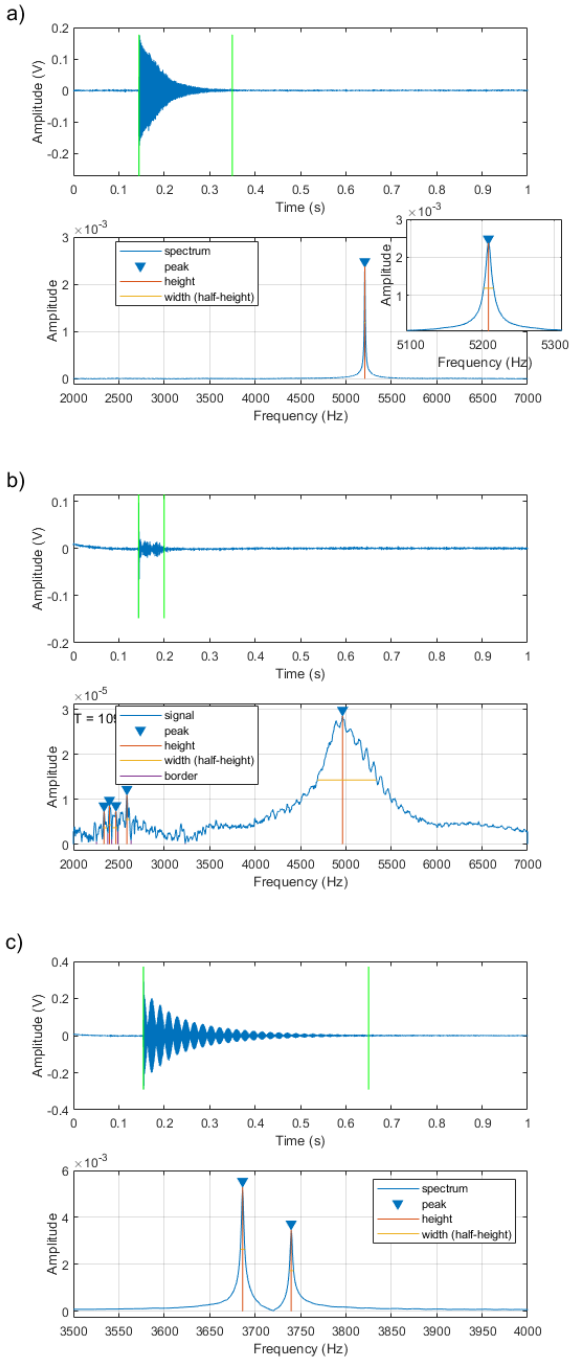

Fig. 5. The examples of an acquired sound signal and its frequency spectrum: (a) a good signal with low damping and a single peak on the frequency spectrum; (b) a highly damped signal showing multiple peaks on the frequency spectrum (small peaks did not satisfy the condition for minimal prominence); (c) a signal with beats of vibration which results in two close peaks on the frequency spectrum (their relative height may change during measurement).

Increase the minimum peak prominence [15] to avoid identifying noise as a peak (Fig. 5(b)).

To select the peak closest to that of the previous impulse instead of selecting the highest peak, sometimes a "false" peak with higher amplitude appears in the spectrum. This improves the automatic selection of the correct resonant peak in case of a bad spectrum or if there are "double" peaks due to the sample geometry (Fig. 5(c)).

F. Program interface

The measurement is controlled by a computer program with a graphical interface written in MATLAB®. Before the measurement, it allows the user to set the sample information. A screenshot of the program is shown in Fig. 6. The program has several features to improve the evaluation of recorded sound signals, which were discussed in the previous section. Moreover, the user can decide whether the measured sound signals are stored for post-processing.

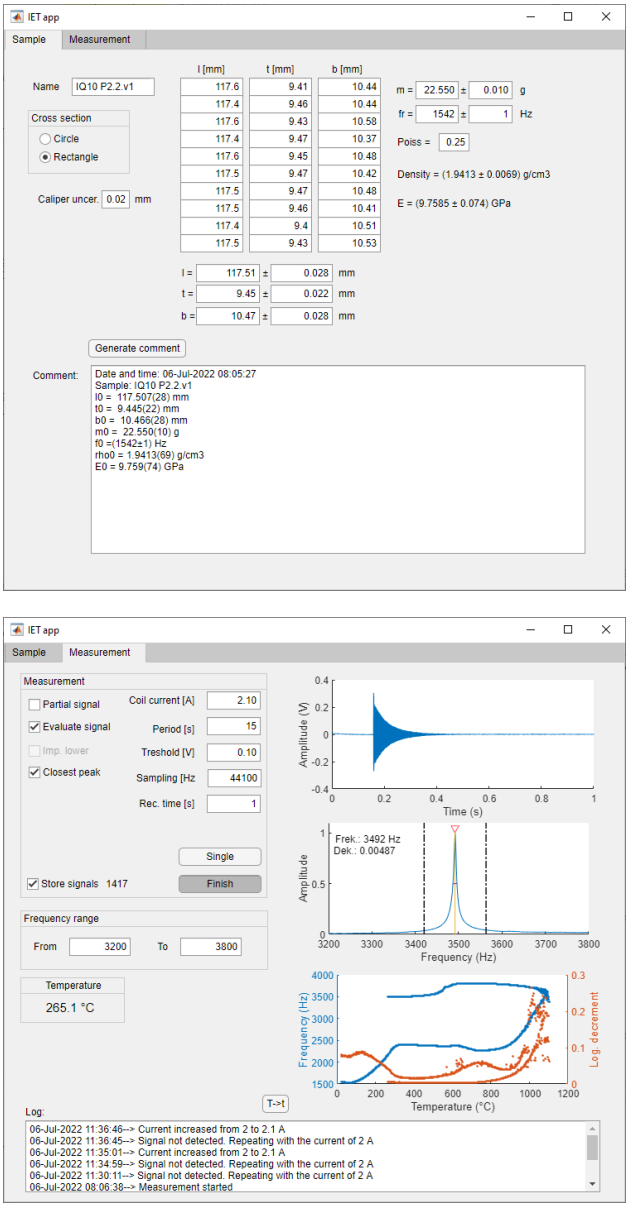

Fig. 6. The screenshot of the program.

4. MEASUREMENT EXAMPLE

An example of the measurement can be found in Fig. 7. The sample was made from a mixture of illitic clay with 20 mass% of quartz by plastic extrusion. After free drying, the sample was cut and abraded to final dimensions of ~99.0 × 9.5 × 10.5 mm^3. The resonant frequency f_0 and logarithmic decrement δ were measured at 15 s intervals from room temperature to 1100 °C and back to room temperature at heating and cooling rates of 5 °C/min. The data in Fig. 7 is obtained from automated measurement without further post-processing.

Up to ~300 °C, both f_0 and δ sensitively reflect the removal of residual moisture from the material. A few scattered points are visible on the δ curve. After a linear decrease, f_0 increased at ~573 °C due to quartz modification transition. The further decrease of f_0 and the increase of δ are attributed to the dehydroxylation of illite [16]. At 733 °C, f_0 reached its local minima and δ its local maxima. From this point, f_0 started to increase and δ to decrease as a result of solid-state sintering. The start of viscous flow sintering can be assigned to the temperature of 900 °C, above which δ increased rapidly. A local decrease in δ was registered at about 1000 °C, possibly due to an as yet unknown process. δ reached its global maximum at 1055 °C, which correlated with a decrease in the rate of f_0 change. After reaching the maximum temperature, δ started to decrease immediately due to an increase in the viscosity of the glassy phase. On the other hand, f_0 continued to increase for the same reason. At 573 °C, a deteriorating effect of quartz was manifested during cooling. Due to the extraordinary temperature dependence of the elastic properties of quartz during its modification transition [17] and because of the rapid change in its thermal expansion coefficient, the course of f_0 showed its typical V-shaped minima followed by a substantial decrease [16]. The modification transition of quartz was also reflected in δ. Upon further cooling to room temperature, f_0 continued to decrease because of microcrack formation, which correlated with an increase in δ. δ had a local maximum at ~275 °C during cooling, but the origin of this maximum is still unclear.

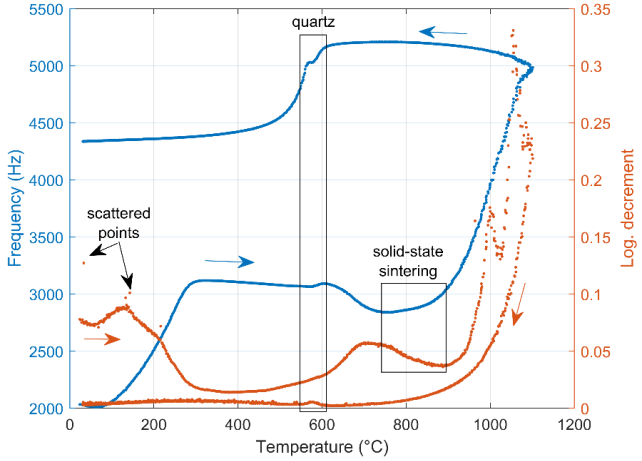

Fig. 7. The example of measurement. Sample: illitic clay + 20 mass% of quartz; heating and cooling rate was 5 °C/min.

From this example, it can be seen that resonant frequency and internal damping are very suitable material properties that can be used to study the firing process. In some cases, both reflect the process equally well, but the sensitivity to some processes differs between the two, making them very useful when used in a complementary manner. The constructed apparatus has high sensitivity to both f_0 and δ, with low scatter of the measured points, which is achieved by the construction of the impulser and supporting system together with algorithms for automatic signal evaluation.

5. CONCLUSIONS

The apparatus for dynamic thermomechanical analysis (D-TMA) designed and constructed by the authors is described. The apparatus measures the resonant frequency of the flexural vibration of the sample using the impulse excitation technique (IET), which is based on the free vibrations following a mechanical impulse. Since the measurement is performed at temperatures up to 1250 °C, an electromagnetic alumina impulser is used for excitation. The free vibrations are registered with the electret microphone, stored and then analyzed with the fast Fourier transform. The result is a frequency spectrum, that can be used to determine the resonant frequency of the fundamental mode of the flexural vibrations. The furnace is built from refractory porous alumina bricks and alumina fiber pads. The heating elements are four SiC rods connected to a temperature controller. The temperature is measured with a Pt-PtRh10 thermocouple in close proximity to the sample. The example of measurement has shown that the apparatus can reliably determine the resonant frequency and logarithmic decrement of the samples with relatively high internal damping.

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