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IMPULSE ANNEALING AS POSSIBILITY OF MODIFICATION OF MAGNETIC PROPERTIES OF AMORPHOUS METALLIC ALLOYS

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This paper introduces the construction of the device used for application of Joule heating on amorphous melt-spun ribbons with a pulse length of the order of 0.1 s. The electric current of pulse is about 21 A at frequency 50 Hz. The samples of ribbons are annealed utilizing series of pulses with total duration of 80 up to 120 ms. The annealing is performed directly in solenoid of magnetometer, which is used for the subsequent measurements of magnetic properties of processed materials. The significant improvement in the magnetic properties after pulse heating was achieved on the amorphous ribbons of FINEMET type.

Keywords: FINEMET, pulse heating, induced anisotropy, Curie temperature, coercivity

1 INTRODUCTION

The utilization of soft magnetic materials in the form of amorphous melt-spun ribbons has an increasing tendency in the technical practice. Particularly interesting is the wide range of their properties after heat treatment. For improvement of the magnetic properties of these materials outside of doping and changing of composition the subsequent thermal processing is used [1]. The disadvantage of classical thermal treatment is irreversible change in the amorphous structure leading to the formation of atomic clusters, in extreme cases up to crystallization [2].

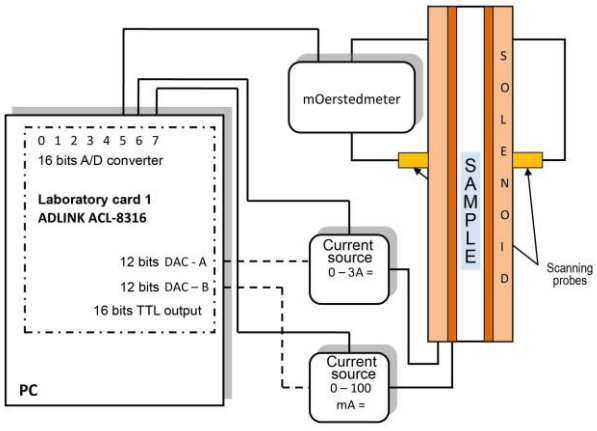

Fig. 1. Block diagram of the magnetometer

These changes are commonly associated with a significant increase in brittleness of the material. One possibility to avoid or minimize these changes is the utilization of a pulsed heating of the samples [3,4]. We have decided to use Joule heating with a pulse length of the order of 0.1 s. The diffusion of atoms occurs at short range is expected during the short pulses. The consequence is likely to cause a significant reduction in the internal stress introduced to the material in the production process, this results in reduced losses. We have possibility to apply external magnetic field or mechanical stress during thermal heating. This may introduce the induced anisotropy in to material, which can further improve its magnetic properties.

For demonstration of capabilities of our equipment the amorphous ribbons, made from known material such as FINEMET ($Fe_{73,5}Si_{13,5}B_9Nb_3Cu_1$), were used. This was done due to the possibility of adjustment of its magnetic and mechanical properties [5,6] by heat treatment. The ribbons were prepared by melt-spun technique with a thickness of 30 microns and a width of 10 mm.

EXPERIMENTAL

2.1 Magnetometer

The magnetic parameters of the samples were measured by a computer-controlled magnetometer using the magnetometric method. In this method, the magnetic polarization of the sample is evaluated by measuring the horizontal component of stray fields of sample. The advantage of this method is possibility to measure the magnetic polarization of the sample without changing of its magnetic state. The disadvantage is requirement of the open shape samples; hence the demagnetisation factor should be taken into account when processing the data.

Figure 1 shows schematic use of the magnetometer. Stray fields of sample are detected using of two fluxgate type probes of commercial milioerstedmeter. Sensing probes are set against each other at half of the height of the solenoid. The sample of length of 10 cm is positioned with one of the poles at the level of sensors. The reason for such an arrangement of the experiment is to achieve

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the highest sensitivity of the device. The probes are positioned on the outside from the solenoid; the solenoid is free and can be placed in it, *eg* the oven for heat treatment or temperature dependent measurements and/or, as in our case, a device for pulse heating.

2.2 Device for pulse heating

The device for the implementation of a pulsed heating of the sample was designed and constructed to allow placement in the measuring device. This configuration allows pulsed heating with different parameters without sample handling as well as measuring of the magnetic polarization of the sample in the process of pulsed heating.

The burst length can be controlled over a wide time range from 0.01 s to whole seconds, allowing the effective regulation of heating temperature. The sample is placed into a vertically oriented solenoid in the holder allowing pulsed annealing of the sample and simultaneous application of the magnetic field, or vertical mechanical tensile load.

The block diagram shown in Fig. 2 depicts the wiring apparatus for the pulsed heating and interconnection with apparatus for measuring of the magnetic properties.

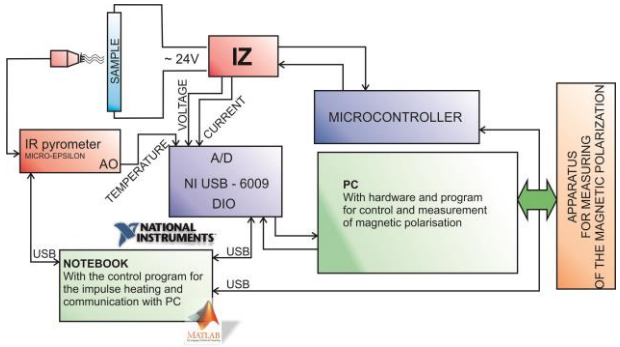

Fig. 2. Block diagram of the device for pulse heating with circuit for calibration of temperature

As shown in the figure, the sample is attached to a source of pulse alternating current (IZ). The current flows directly through the sample and the joule heating occurs during a pulse. The pulse source is controlled by a microcontroller enabling the current to be switched on once the sine wave of current crosses zero. The microcontroller counts the exact number of half periods of impulses by using the interrupt. The number of half periods is provided to microcontroller from the computer device. The computer also simultaneously performs measurement of physical parameters characterizing the pulse through the USB room card NI USB-6009. The data collection and management of the entire apparatus is programmed in MATLAB environment. The numerical calculations for determining the resistance of samples, performance and total energy (temperature reached) are performed based on the values of voltage on the sample and the current flowing through the sample collected during the pulse period. Results of these calculations are registered separately for every pulse in table and chart with the selected parameters (see Fig. 3, and Fig. 4). Subsequently, the data are sent to computer device for the management and measurement of magnetic polarization. The individual parameters characterizing the pulse as measured on a FINEMET sample with pulse duration of 100 ms are shown in Fig. 3 and Fig. 4.

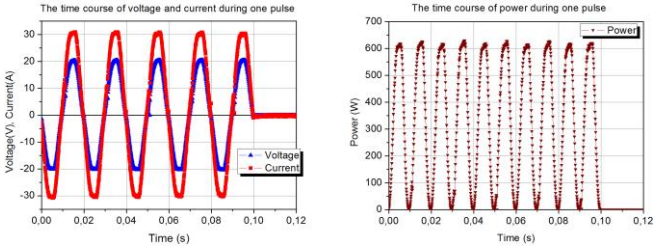

Fig. 3. The time dependency of selected parameters during the pulse of length of 100 ms

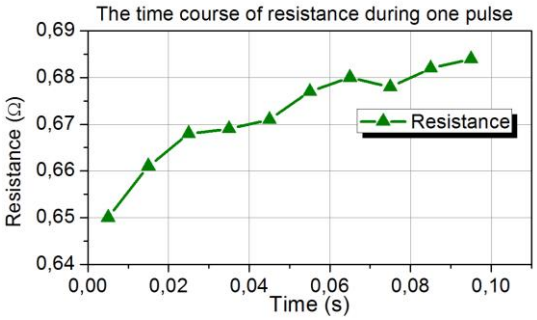

Fig. 4. The time dependency of electric resistance during the pulse of length of 100 ms

Measurement of the temperature initially presumed utilization of the contactless sensor directly during the heat processing, however no suitable nonferromagnetic sensor was found. Therefore temperature calibration was carried out of magnetometer. Temperature measurement was realized by miniature contactless infrared pyrometer (CTF-SF15-C3 MICRO-EPSILON). The reason for contactless temperature measurement is the low heat capacity of the sample and a large heat capacity of the sensors with small reaction times.

Every impulse, following the temperature calibration of this device, can be assigned not only with the maximum achieved temperature and energy (see. Tab. 1), however also with the timing dependency of the temperature. Examples of dependencies of temperature vs. time for the pulses of different length (and so for different energies) are presented in Fig. 5

Tab. 1. The peak temperature and energy assigned to pulse of different length

The pulse time (ms)	The peak temperature (°C)	Energy (J)
80	240	25,70
100	345	32,20
120	395	38,40

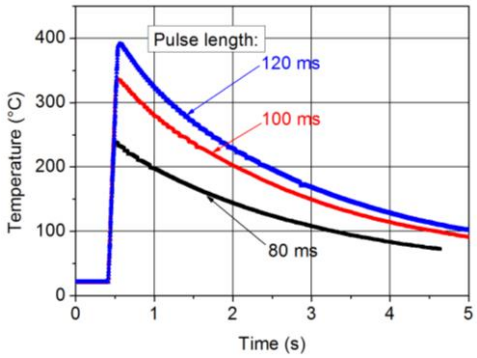

Fig. 5. The time dependencies of temperature for pulses of different length measured on sample FINEMET

In testing phase was used type of amorphous material called FINEMET, it was utilized for the temperature calibration of the device. At first, the Curie temperature (Tc) of this material was evaluated. This temperature was from the temperature dependency of the magnetization as measured on the vibrating sample magnetometer (VSM) (see Fig. 6). The value of T_c was estimated to 351°C. Subsequently, the ribbon of the same material was placed into our pulse device and some pulses of length 120 ms were applied on it. In Fig. 7 is presented the time dependency of the magnetic polarization of sample for different number of pulses together with dependency of temperature for pulse of length 120 ms.

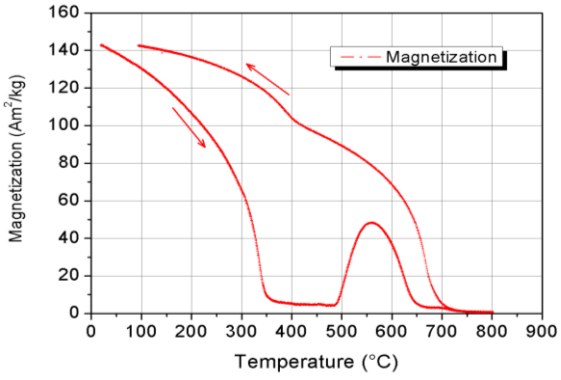

Fig. 6. The temperature dependency of magnetization for FINEMET sample

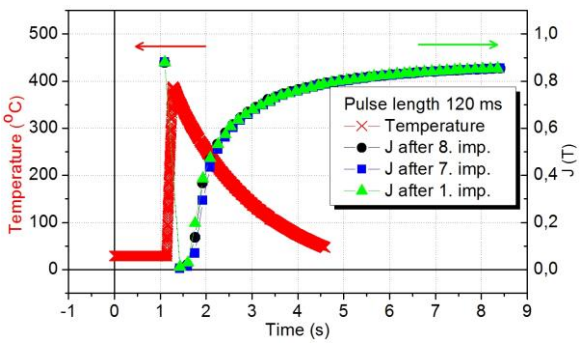

Fig. 7. The time dependency of temperature for pulse of length 120 ms (left) and the time dependency of the magnetic polarization of sample for different number of pulses (right)

From these data, the cube dependencies of magnetic polarization on the temperature were constructed (Fig. 8). Such dependency is of linear nature, thus one is able to accurately determine the intersection with the x-axis, *ie* the Curie temperature.

The results are three Curie temperatures for three different pulse counts. Increasing the Curie temperature as a function of the number of pulses can be explained by structural rearrangement of atoms and thus increase the value of the exchange integral. The relationship between the exchange integral and the Curie temperature is in Fig. 7.

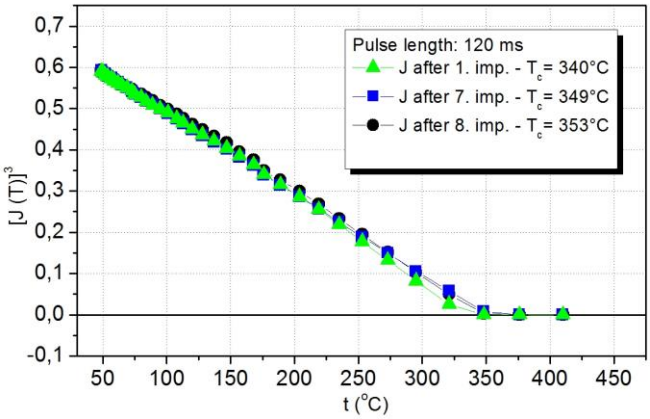

Fig. 8. The cube dependencies of magnetic polarization on the temperature

$$A = \frac{3kT_c}{2zS(S+1)},$$

where the k is the Boltzmann constant ($k = 1.38\times10^{-23}$ JK^{-1}), Tc is a Curie temperature of the sample, z the number of the next ferromagnetic atoms adjacent to ferromagnetic one and S the value of orientation of spins. By comparing the values of Tc obtained from VSM and from pulse heating we can see that their consistency is good, and thus the calibration of temperature can be considered very well.

3 RESULTS AND DISCUSSION

To verify the capabilities of the new equipment we selected once again the amorphous material FINEMET ($Fe_{73,5}Si_{13,5}B_9Nb_3Cu_1$), prepared by rapid cooling of the melt. The measurement was carried out as follows: First, the magnetization curves were measured on the sample in as-quench state. Then the heat pulse was applied and 10 minutes after the magnetization curves were again measured. This cycle was repeated 20 times.

In Fig. 9 hysteresis loops measured in both, as-quench state and after application of 20 pulses are shown, each with duration of 100 ms. One can see that pulse heating caused a reduction in coercivity and significantly increase the slope of the hysteresis loop. Hence increasing the slope of the hysteresis loop will imply also the reduction of the total magnetic anisotropy constant.

The dependency of coercivity of samples (H_c) on the number of applied pulses is plotted in Fig. 10. The picture shows three dependencies:

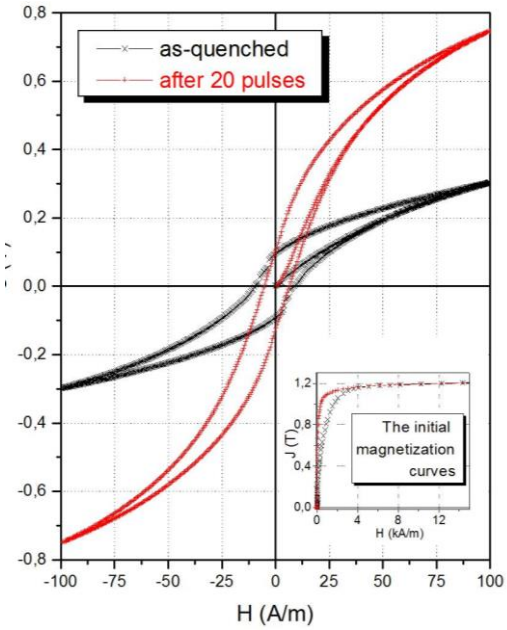

Fig. 9. The hysteresis loops of the FINEMET sample in as-quench state and after application of set of 20 pulses

Dependency on the pure pulse heating, dependency on the pulsed heating applied simultaneously with application of a magnetic field (H = 2000 A/m) in the direction of the axis of the sample, and lastly the dependency on the pulsed heating applied simultaneously with application of mechanical stress (σ = 2.27 MPa) in the axial direction of the sample.

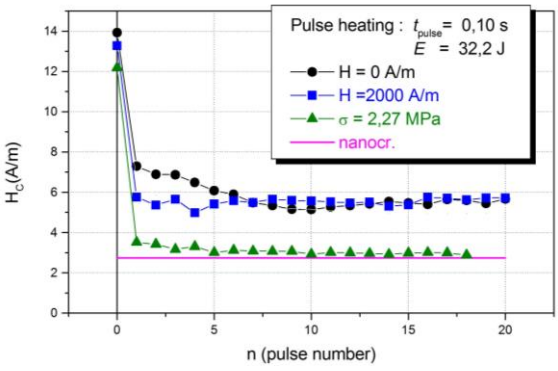

Fig. 10. The influence of pulse heating on the coercivity of FINEMET at the different conditions of processing during the manufacturing process

The sharp decreasing in coercivity after applying of the first pulse can be explained by a significant reduction in internal stresses which were introduced into the material

This effect is more significant for the case of the pulse heating with the simultaneous application of mechanical stress. It is believed that in addition to relieving occurs the rearrangement of atoms at a short distance what leads to an induced anisotropy, resulting in further reduction in the coercivity of the material.

4 CONCLUSION

The new equipment for pulsed Joule heating was designed and constructed. For every pulse was estimated not only the maximum temperature achieved but also the time course of the temperature during and after the pulse and energy the pulse induces.

Using pulse heating was achieved a significant improvement of the magnetic properties of the FINEMET material. Thus, the short heating time is sufficient to induce diffusion of atoms over short distances, leading to relieving internal stresses and also to elicit induced anisotropy. This short heating time is nevertheless insufficient for the diffusion of atoms over a long distance, leading to the formation of FeSi clusters or crystallites, causing embrittlement.

Acknowledgement

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REFERENCES

[1] NOVÁK, L. - POTOCKÝ, L. - LOVAS, A. - KISDI-KOSZÓ, É. - TAKÁCS, J. : J. Magn. Magn. Mat.. 19(1980), 149 - 151.
[2] POTOCKÝ, L. – KAREL, V. - NOVÁK, L. – LONGAUER, S. - KISDI-KOSZÓ, É.: acta phys. slov. 31,(1981) 2-3
[3] REED, R.C. - SHEN, Z. - AKBAY, T. - ROBINSON, J.M.: Materials Science and Engineering A232 (1997) 140- 149
[4] KISDI-KOSZÓ, É. – POTOCKÝ, L. – HRABČÁK, M. – NOVÁK, L. - LOVAS, A.: J. Mag. Mag. Mat. 92 (1990), 181 - 184
[5] LOVAS, A. - KISS L. F. - VARGA, B. - KAMASA, P. - BALOGH, I., - BAKONYI, I.: J. Phys. IV France, Vol.08. (1998) Pr2- 291 - Pr2- 29
[6] VARGA, L.K. - GERCSI, ZS. - KOVÁCS, GY. - KÁKAY, A. - MAZALEYRAT , F.: J. Magn. Magn. Mater. 254-255 (2003) 477-479
[7] CHIKAZUMI, S.: Physics of Ferromagnetism, Oxford University Press, 1997.

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