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# DEVICE FOR IN-SITU DETECTION OF MICROSTRUCTURAL TRANSFORMATIONS

# UREĐAJ ZA IN-SITU DETEKCIJU MIKROSTRUKTURNIH TRANSFORMACIJA

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## ABSTRACT

*Innovative idea of the device for detecting microstructure changes, concept and construction was carried out and controlled by authors of this article in cooperation with Slovenian industry.*
*Since the electrical resistance is one of the most structurally sensitive properties of the materials, we used its measurement to detect microstructural changes caused by phase transformations or chemical reactions.*
*Until now, we analyzed the phase transitions in rapidly solidified alloys of copper and aluminum, nickel and nickel alloys, and several types of steel made by Slovenian steel producers.*

**Keywords:** microstructural transformations, in-situ measurement of electrical resistivity measurement, material research

## SAŽETAK

*Inovativna ideja za uređaj za otkrivanje promjena mikrostrukture, koncept i izrada provedeni su i kontrolirani od strane autora ovog članka u suradnji sa slovenskom industrijom.*
*Budući da je električni otpor jedno od najvažnijih svojstava koje je osjtljivo na mikrostrukturne promjene u materijalu, ova mjerenja su korištena za otkrivanje mikrostrukturnih promjene uzrokovanih faznim transformacija ili kemijskim reakcijama.*
*Do sad su analizirane fazne transformacije u brzo očvrslim legurama bakra i aluminija, nikla i legura nikla i nekoliko vrsta čelika iz slovenskih željezara.*

**Ključne riječi:** mikrostrukturne transformacije, in-situ mjerenja električne otpornosti, istraživanje materijala

## 1. INTRODUCTION

Innovative idea, concept and construction of the device for the in-situ detection of microstructural transformations during heating was conducted at the Chair of Engineering Materials, Department of Materials and Metallurgy, Faculty of Natural Sciences, University of Ljubljana in cooperation with Slovenian companies YDRIA Motors, National Instruments, Vacutech, Induktio and Kolektor. This device is intended for both, research laboratories and industry. The beginning of its construction dates back to 2008, and the first measurements have been successfully implemented in early 2011. To date, we have analyzed the phase transformations in rapidly solidified copper and aluminum alloys, nickel and nickel alloys, and several types of steel for Slovenian steel producers. A measuring system for in-situ microstructural transformation detection is an important acquisition for education in the field of materials, metallurgy, mechanical engineering and manufacturing technologies for graduate and postgraduate study courses [1].

## 2. MEASURING METHOD

Several methods are available in order to monitor microstructural transformations, and can be classified into discontinuous ”ex-situ” and continuous ”in-situ” measurement methods. In-situ measurement methods enable real-time monitoring of microstructural transformations and study of transformation kinetics. For microstructural transformations to occur, thermodynamic and kinetic conditions must be satisfied.

Evaluation of the kinetic conditions requires data acquisition, which is obtained by the measurement of certain physical property. Since the electrical resistivity is one of the most sensitive structural properties of the materials, we used it for the detection of microstructural changes created by phase transformation or chemical reactions in the material.

Microstructural transformations in pure metals or alloys are perceived on the basis of alteration of electrical resistance of the sample. Because alterations in the electrical resistance, due to microstructural transformations are in the range of micro-ohms, reliable measurements of such small electrical resistance deviations are only possible with an appropriate measuring method and special measuring equipment. The entire measurement system has to be designed to compensate the errors caused by the disturbing effects, such as:

- contact electrical resistance
- measuring cables electrical resistance
- voltage induced by the thermoelectric effects
- undesired heating of the sample by measuring current
- electromagnetic disturbances from the surroundings.

The measurement of electrical resistance is carried out through measurement of voltage change at a known direct current to the sample. For accurate measurement of the sample electrical resistance, a four-point U-I method has been used, which eliminates the influence of measuring wires on measured electrical resistance. The undesired heating of the sample by measuring current is minimized by changing the polarity of the power source, and the electromagnetic disturbances are minimized by electromagnetic shields [2].

## 3. MEASURING SYSTEM

Device for following the microstructural transformations by measuring the electrical resistance is composed of three basic parts:

- heating system
- vacuum system
- monitor/control unit.

The scheme of the measurement system is shown in Figure 1. Heating system consists of a vertical electrical resistance furnace with water cooled outer chamber and inner ceramic retort, acting as a radiation shield,which prevent direct incident radiation from the heater o the sample. Sample can be heated with a constant heating velocity, annealed at a constant temperature or more complex temperature regimes. Maximal operational temperature is 1200°C. Temperature in the furnace is controlled by PID regulator and S-type contact thermocouple attached to the sample.

The vacuum assembly combines a rotary and turbomolecular pump, which enables high vacuum in a furnace chamber. Pressure is continuously monitored by two Pirani gauges in low and middle vacuum range, and by Penning high vacuum gauge. Measurements can be carried out in a vacuum (< $10^{-5}$mbar), or reactive ad inert gas atmosphere, which can be induced into the system through capillary tube and a precision metering valve from the gas bottle. The partial pressure of the inert gas is measured by membrane gauge. Maximal pressure in the furnace chamber is 1bar. The monitoring/control unit allows measurement of electrical resistance during annealing in the real-time, and consists of measuring cell with a precision current source KEITHLEY 6220, nano-voltmeter KEITHLEY 2102A, data acquisition card National Instruments, PC-computer and the LabVIEW program for visualization an control. Direct measuring currents used in measurements, are due to the risk of heating of the sample very small, of the order of 10 mA. Accordingly, measured voltage alteration is also extremely small, and has to be perceived by the sensitive nano-voltmeter.

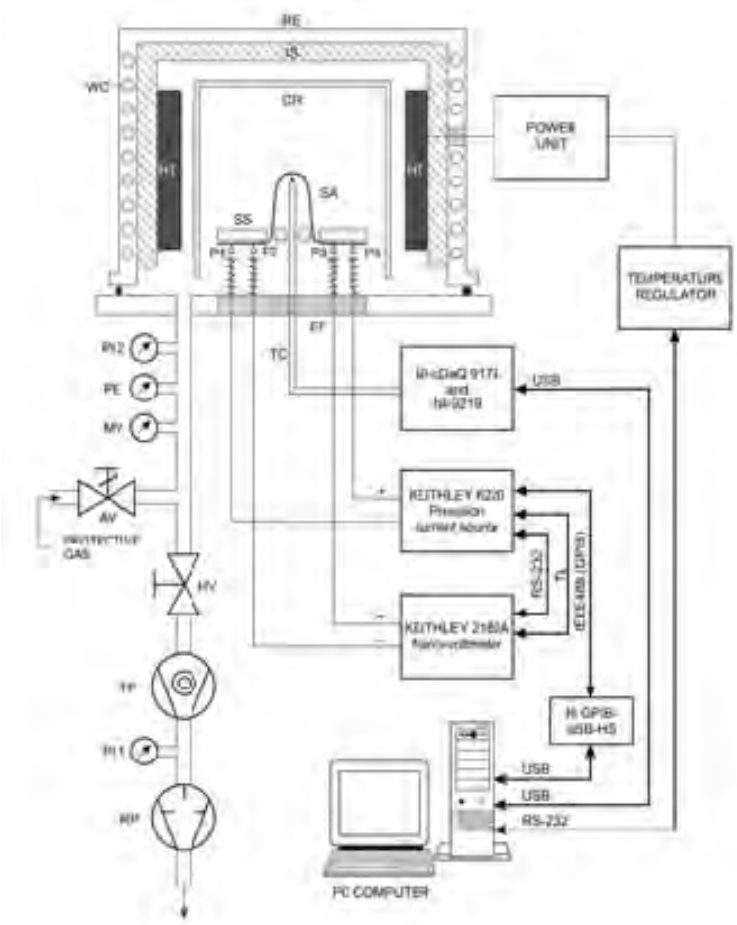



*Figure 1. A scheme of a measuring system for in-situ electrical resistance measurement (RE- Recipient, IS-Isolation, CR-Ceramic retort, WC-Water cooling, HT- Heater, SA-Sample, SS-Sample support, P1, P2, P3, P4-Spring loaded measuring contacts, EF-Electric (cable) feed-trough, TC-Thermocouple, RP-Rotary vane pump, TP-Turbomolecular pump, HV-High vacuum valve, AV-Argon dosing valve, PI 1-Pirani vacuum gauge, PI 2-Pirani vacuum gauge, PE-Penning vacuum gauge, MV-Membrane vacuum gauge).*

Metals and their alloys have relatively low electrical resistivity with positive (linear or polynomial) temperature coefficient. To increase electrical resistance of the sample and reduce background measuring "noise", samples have to be prepared in the form of thin ribbons of plates. Standard procedure for sample preparation consist of cutting or precision sawing of thin foil from the bulk material and grinding to uniform thickness of a few tens of microns. The sample is then mounted on a special ceramic holder (Figure 2) and attached to four contact pins, which are spring supported to ensure reliable and stable electrical contact. Contact pins are made from platinum, due to its linear electrical resistivity temperature dependency, and connected to the measuring units by cables of known electrical resistance.

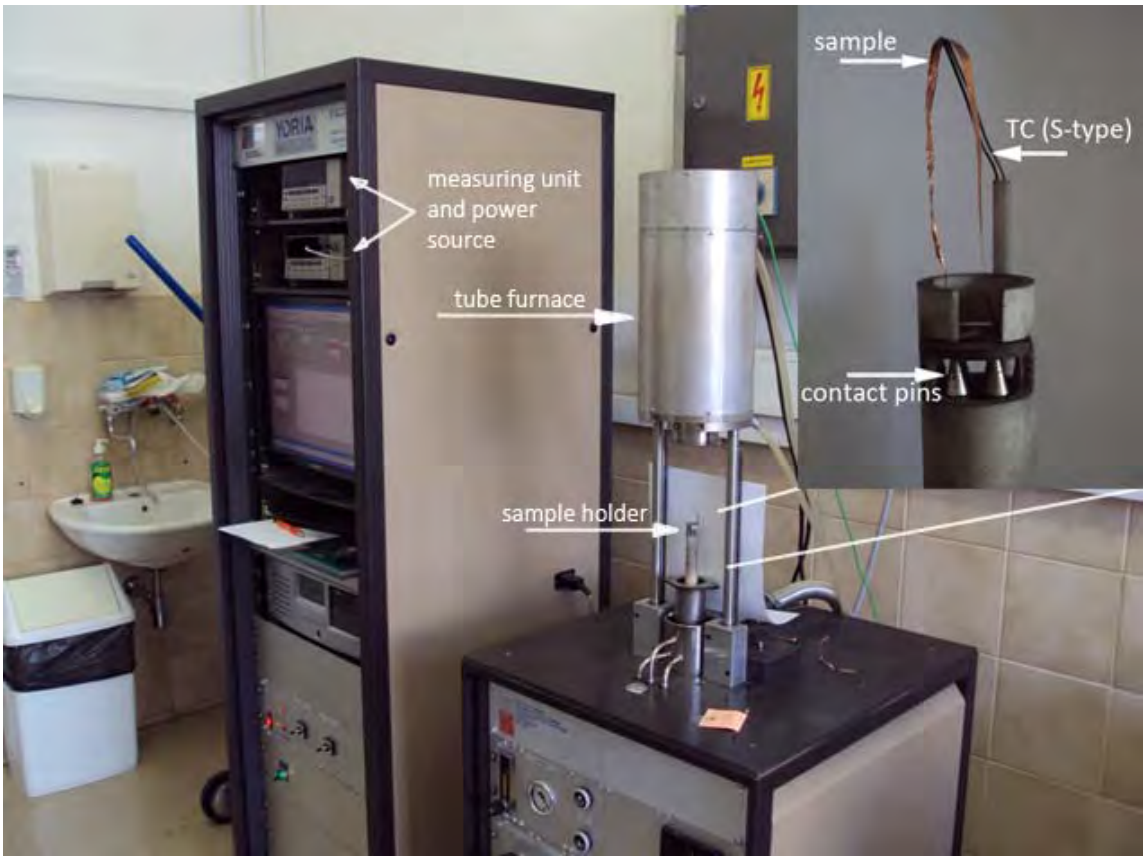



*Figure 2. A device for microstructural transformations detection. On the upper right corner is the detail of a sample holder.*

The measuring system is controlled via the virtual control panel on the computer screen (Figure 3). The control panel allows easy setting of measurement parameters, data acquisition, graphical real–time presentation and exporting the data in various file formats (CSV, TXT, JPEG…). Results are graphically presented in the form of curves showing the dependency of the electrical resistance as a function of temperature or time. The virtual command plate is programmed in LabWiev a program language.

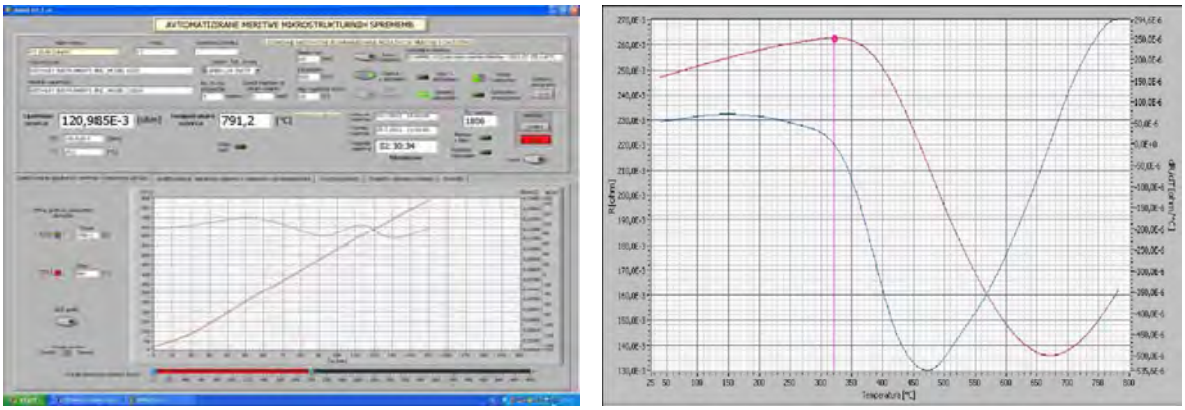


*Figure 3. User interface-virtual command panel(left) and example of electrical resistance and its first temperature derivative curve of rapidly solidified Cu-Cr alloy(right).*

## 4. ELECTRO-THERMOMETRY

Results of electro-thermometry are presented in the form of curves showing the dependency of the electrical resistance as a function of temperature or time, and can be used for the interpretation of various phenomena in physical metallurgy, such as temperature of precipitation from saturated solid solutions, phase transformation start or finish, Currie temperature determination as a function of chemical composition, activation energy calculation for phase transformations, study of reaction kinetics in solids, etc. An automated system allows the study of thermally activated processes in a variety of thin layers of metallic glasses, magnetic and electrical materials [3,4], quasi-crystalline alloys, rapidly solidified alloys,shape memory alloys [5,6] and other metallic and non-metallic materials in the crystalline or amorphous state.

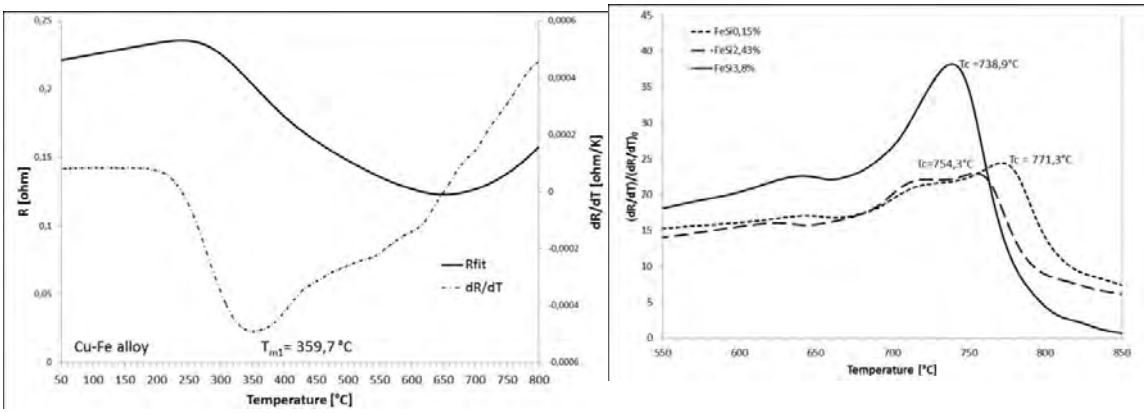



*Figure 4. Currie temperature determination as a function of chemical composition of Fe-Si electrical steel (left), resistance and its derivative curve as a function of temperature determinate precipitation temperature of Fe from Cu-Fe supersaturated alloy (right).*

Figure 5 shows a comparison of the results obtained by DTA, DSC and electrical resistivity measurements during constant heating rate of 5 K/min for AlFe7Si2,3Zr rapidly solidified alloy. As can be seen, the temperature of the exothermal peak on DTA and DSC curves coincide well with the temperature of the first minima on the electrical resistivity derivation curve. The difference between $T_{m1}$ electrical resistivity minima and DSC and DTA exothermal peaks temperature is only 1.5 °C. On the contrary, DTA and DSC measurements didn't detect any heat generation at the temperature $T_{m2}$ where the second electrical resistivity

minima was recorded. By electron scanning microscopy it was proved that first electrical resistivity minima corresponds to precipitation of $Al_xFe_ySi_z$ spherical particles and the second to precipitation of $Al_3Zr$ globular particles.

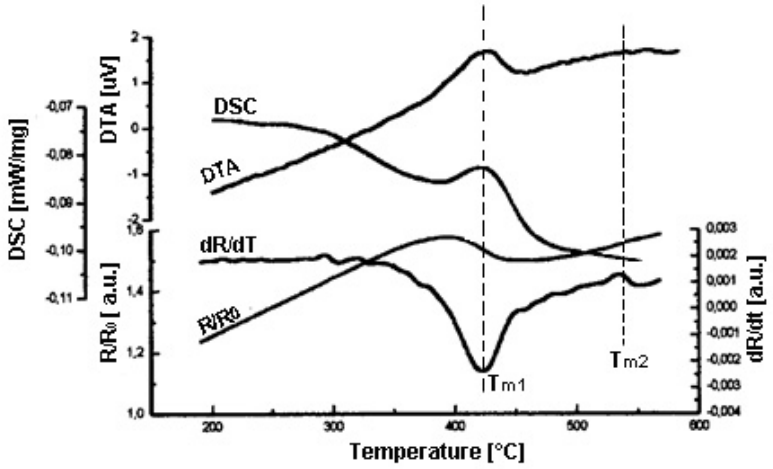
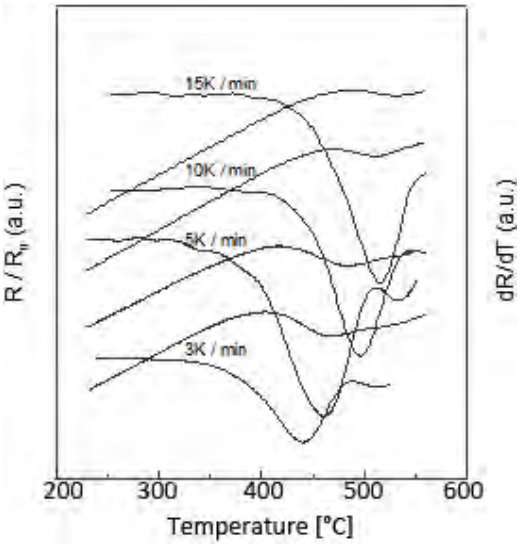



*Figure 5. Comparison between electrical resistance, DTA and DSC curves recorded at a constant heating rate of 5 K/min for rapidly solidified Al-Fe7-Si2,3-Zr1 alloy [7].*

To calculate activation energy, measurements at various heating rates have to be performed. As can be seen from the figure 6, minimums on the first temperature derivative curves shift to higher temperatures as the heating rate increases, which is a consequence of a shorter time available for the reaction inside the temperature interval. These $T_m$ temperature rise can be used for the calculation of activation energies by Ozawa or Kissinger methods.




Equations for activation energy calculation.

Ozawa method [8]:

$$\log \beta = -0.4567 \cdot \left( \frac{E_a}{R \cdot T_m} \right) + A$$

Kissinger method [9]:

$$ln\left( \frac{T_m^2}{\beta} \right) = \left( \frac{E_a}{R \cdot T_m} \right) + B$$

*Figure 6. Electrical resistance curves and their first derivative curves as a function of heating rate of Al-Fe alloy[10].*

## 5.CONCLUSIONS

Device for following microstructural transformations during heating by in-situ electrical resistance measuring has broad applicability and a number of advantages over other methods and is a useful instrument, from both a theoretical and practical point of view. In some cases, the electrical resistivity measuring method is even more sensitive than other thermal

analytical methods and can in combination with microscopy represents a very powerful tool in the field of material science.

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