In situ annealing experiments combined with orientation mapping technique in SEM and TEM

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1. Introduction

Additional equipment in TEM and SEM as in-situ holders and stages give an opportunity for investigation of materials and obtained complex information about materials behaviour during e.g. mechanical tests, heating. In-situ devices can be applied for many material studies but what is most important thanks to them scientists can obtain unique data about phenomena changing in time.
History of in situ annealing experiments in electron microscopy is long but controversial. People considered how difficult conditions of sample preparation and observation can influence on the beahaviour of the material and how close it can be to the real behaviour of bulk samples. Many research which were done convinced that with taking into account many factors which can influence in situ measurements, they can be successful source of interesting observation. In order to avoid artificial results and confirm observation of the in situ annealing supplementary calorimetric study should be performed.
New benefits from in situ experiments came with development of Orientation Microscopy in TEM and SEM. Combined in situ study and acquisition of crystallographic data from the chosen areas of the sample together with calorimetric study can improve the quality and reliability of investigations (fig. 1).

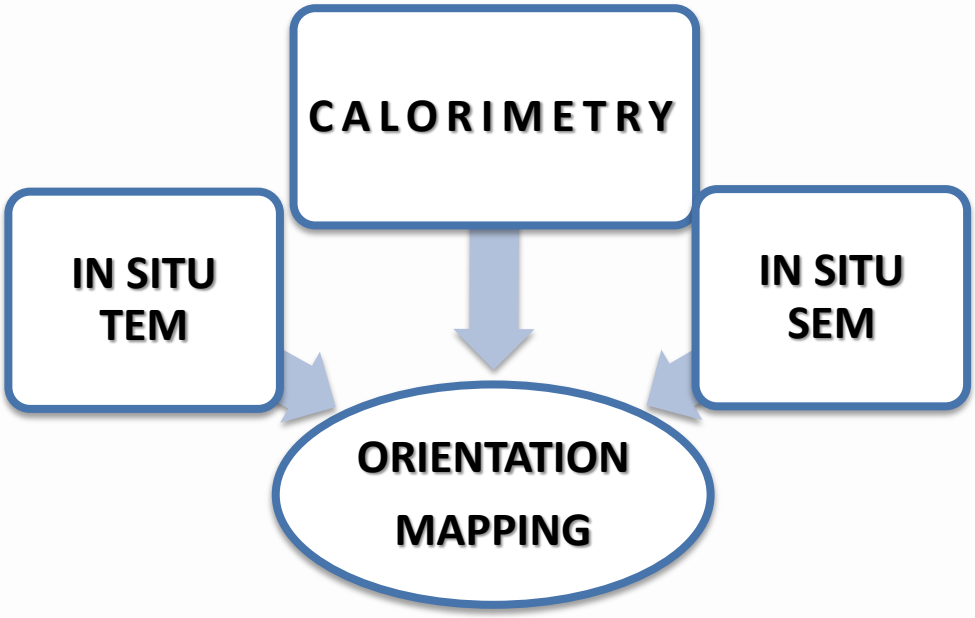
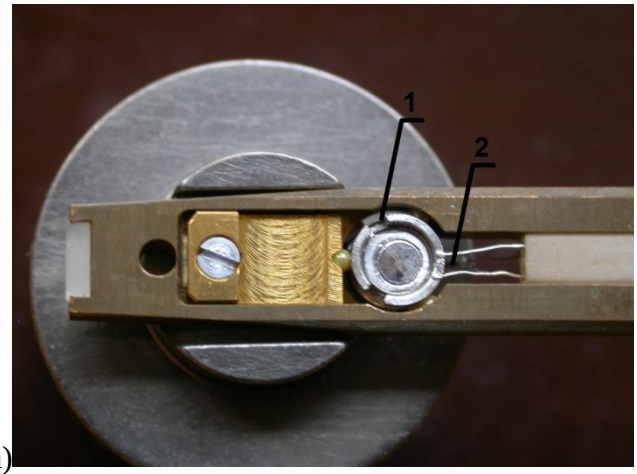


Fig. 1. Schema of in-situ investigations.

2. In situ investigations in TEM

Involving heating device in the study of materials enables additional information about behaviour during annealing. Using TEM, these phenomena can be studied in nano and micro scale. Fig. 2a shows heating holder, which resembles a standard holder for transmission electron microscopy, but has an extra heating element. The sample is mounted in the so called furnace, which is isolated from the handle by zirconium oxide rings. The heater consists of a attached small heating element with welded thermocouple. At the end of the holder, just beyond a vacuum, there are electrical connections to the unit of measurement and control (fig. 2b). It contains the power supply to the heater and allows the measurement of the voltage generated by the thermocouple.

a) 

b) 

Fig.2. TEM heating holder with a) 1)furnace and 2) thermocouple and b) control unit.

The heating process is controlled by changing the voltage and current supply. The power supply unit after switching on must be calibrated. Heating process begins with the gradual increase of voltage (from 0 to 10 mV). The maximum allowable temperature rise is 400 °C per minute. The maximum temperature of heating should not exceed 1000 °C , which corresponds to a voltage of about 10 ± 0.3 mV [Phillips, 1973]. Recorded temperatures are repeatable for different sample. What is observed, is about 100 °C difference between the temperatures obtained using holder and temperatures of the processes in the same material measured using a calorimeter [Sinclair, 1993] .

The main goal of the in situ observation is not a measurement of the temperature of the processes, but the observation of changes in the material with increasing temperature. The first direct observation using heating holder was carried out by Bailey [Bayley, 1960], and Hu [Hu, 1963] . In these works authors noted the difference in recrystallization processes in bulk materials then in thin foils. Further works of Roberts and Lethinen'a [Roberts, 1972] , Hutchinson and Ray [Hutchinson, 1973] , Sztwiertnia and Haessnera [Sztwiertnia, 1994] proved that ensuring proper conditions of in situ experiment, assures to observe changes in the annealed material which are at least close to that occurring in a bulk sample.

In the case of materials subjected to the rolling specific microstructure is obtained in which the grain boundaries are parallel to the plane of the rolled sheet.

It turns out that in the samples cut from planes parallel to the plane of the sheet is a small amount of well-defined grain boundaries, which makes both the nucleation and the migration of grains limited. Properly prepared samples should be cut from the plane perpendicular to the plane of the sheet, which has been confirmed in studies presented in the literature [Hutchinson, 1973; Sztwiertnia, 1994] .

An important feature of the in situ investigations is that front of recrystallization process is stopped in the areas of foil thinner than a certain critical value (Fig. 3). Thin foil areas are not recrystallized due to the formation of the thermal grooving at the surface, which can inhibit the migration of boundary occurring under the influence of large recrystallization motion [Roberts, 1972; Lethinen, 1973]. Effect of sample thickness on the movement of grain boundaries is explained by Mullins [Mullins, 1958]. Mullins showed that the movement of boundaries is stopped by the associated thermal grooving (fig. 4) when a certain grain size is reached. The appearance of the grooves is a result of extensive surface tension at positions where grain boundaries intersect upper or lower surface of the film. Inhibition of grain boundaries in thin areas of the foil is related to the intensive growth of thermal grooves in these areas during the annealing in the microscope. It is also possible to observe thermal grooving in SEM in situ experiments.

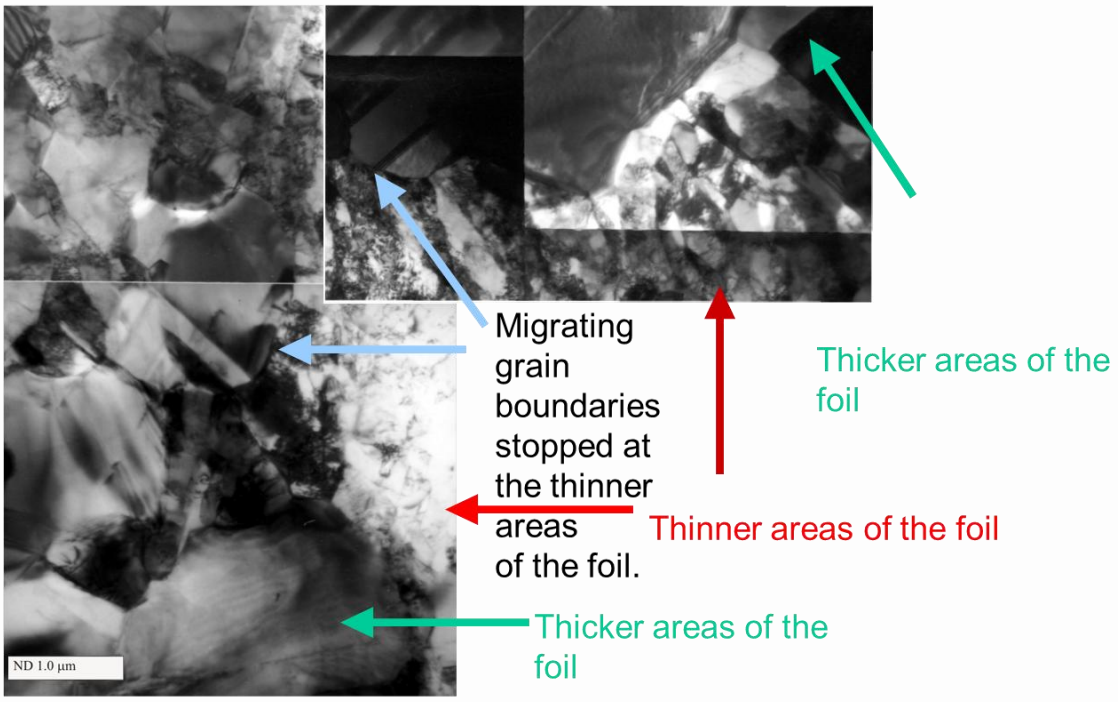


Fig.3. Bright field image, TEM from CuZn28 presenting recrystallization front stopped in the thinner area of thin foil.

Phenomenon of recrystallization front frozen in thin areas of the film may be used to obtain the characteristics of misorientation of boundaries migrating during recrystallization annealing. The results are presented in [Sztwiertnia, 2006].

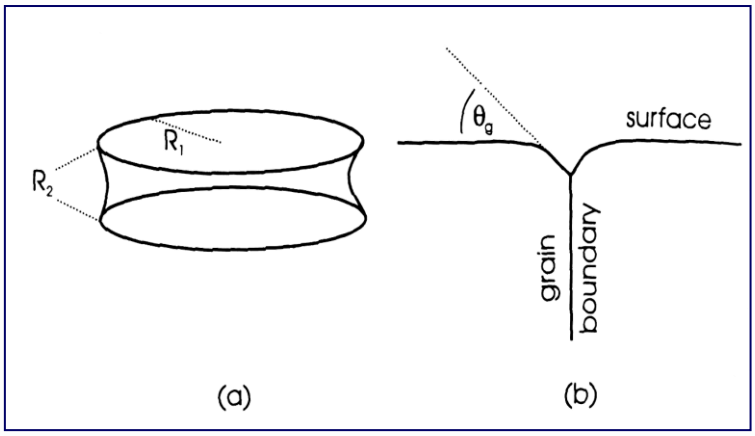


Fig 4. Thermal grooving of a thin specimen. (a) The shape of an isolated grain in a thin sheet, (b) Thermal grooving at the surface. [Humphreys, 2002]

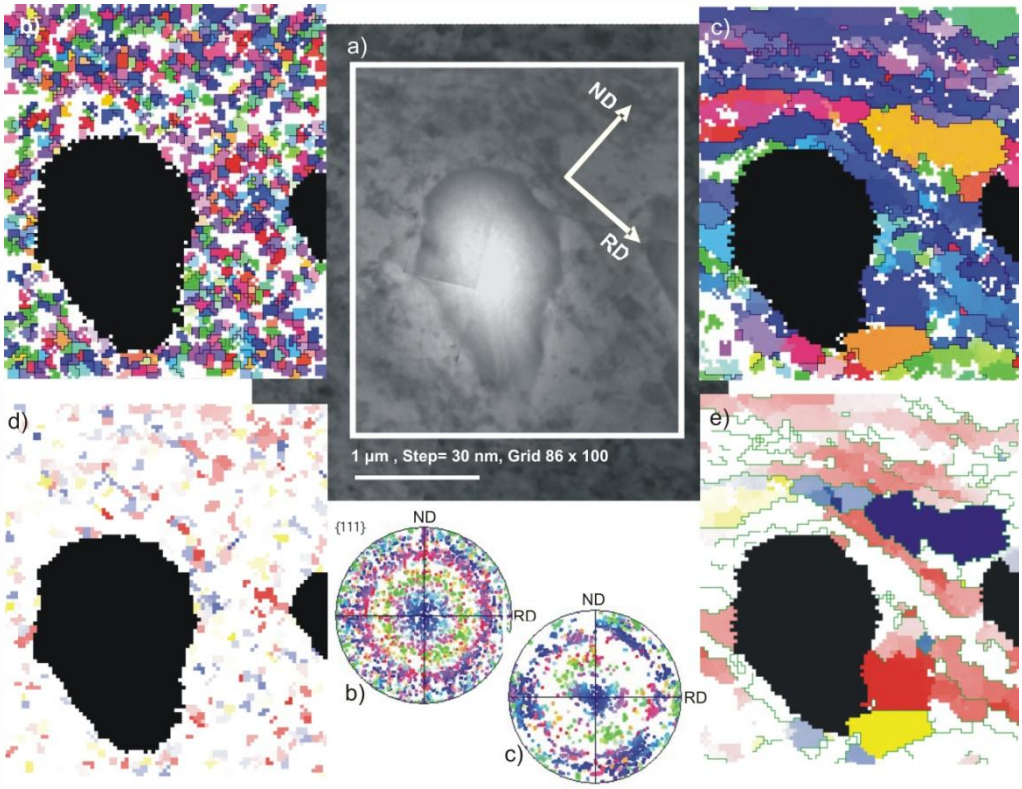


Fig.5 Microstructure of 6013 aluminum alloy cold rolled up to 90%, the deformation zone surrounding large particle (a), longitudinal section, TEM. Orientation topographies and pole figures in areas of the deformation zone before (b) and after (c) heating in-situ in TEM; particles of the second phase - black, white regions – not indexed; thick lines – high angle grain boundaries, thin lines – low angle grain boundaries. Areas with similar orientation (blue, red, yellow) were imaged before and after annealing (d, e). [Bieda, 2010.

Presented results on fig. 5 are from the sample of aluminum alloy 6013 reversibly cold rolled to 90%. The thin foil was prepared from the planes perpendicular to the transverse direction (TD).
After placing the sample in the holder first investigation of microstructure before annealing were performed. Bright field images and orientation maps of selected areas were recorded, particularly in areas of localized deformation around the particles of large precipitates. Then the foils were slowly heated. Changing in the bright field image was observed during heating. The heating was stopped immediately after the first observation of change. After cooling the sample orientation maps measurements were performed in the same places, as in deformed state. This makes it possible to measure the orientation of the new nuclei, appearing in the localized deformation zones. This make possible to compare the microstructure before and after annealing. Result of this investigations are presented in [Bieda, 2010, Sztwiertnia, 2013]

3. In situ investigations in SEM

There are several advantages of scanning electron microscope in situ investigation. The sample possess only one free surface and is resistance to buckling [Humphreys, 1996]. It means that in spite of worse spatial resolution obtained data can be more reliable and more resemble to the analysis of bulk materials. Fast data acquisition systems which are used for EBSD analysis can provide larger scale of analysis then in SEM.

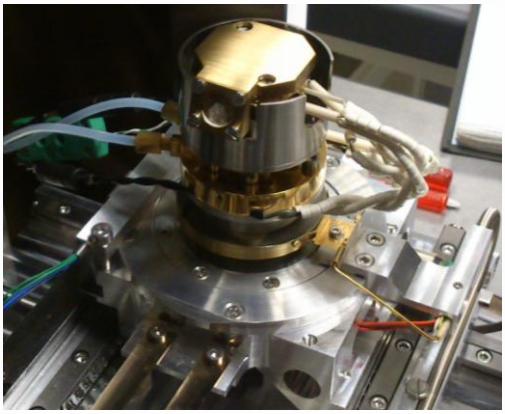


Fig.6. Pretilted SEM heating stage for EBSD application.

For simultaneously EBSD and heating experiments performance special holder which can be tilted up to 70 ° is needed. In market are also available pretilted heating holders as presented on Fig.6.
Main application of this kind of stage is observation of nucleation and grain growth or phase transformation [e.g. Brisset, 20013, Humphreys, 1996, Hurley, 2004, Lens 2005, Wright, 2009]. The unique opportunity of compering the same area of the sample before and after heating for several consecutive temperatures is given. Example of results are presented on fig. 7

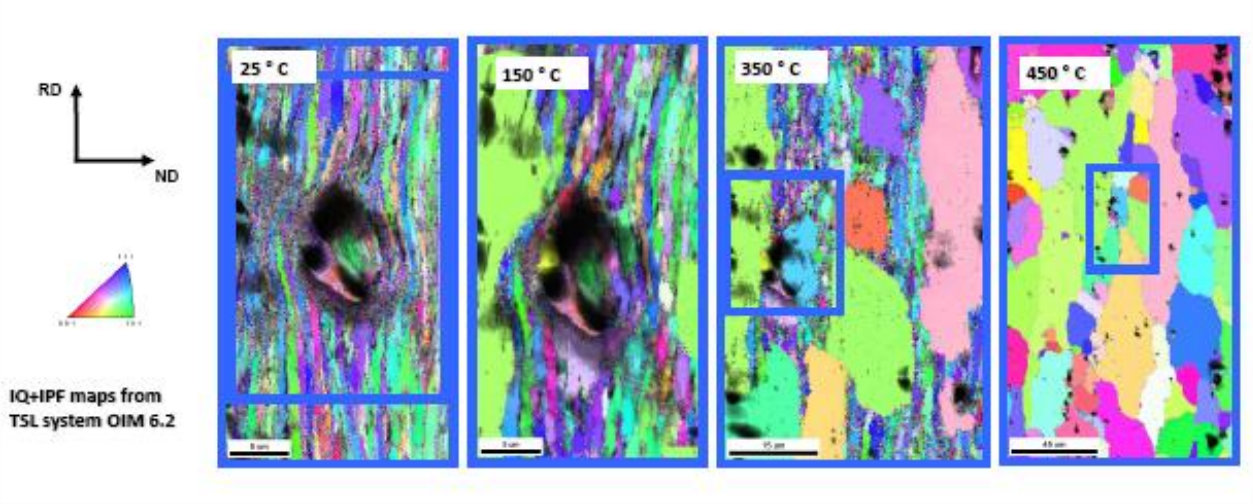


Fig.7. Series of EBSD maps from aluminium alloy 6013 after KOBO and cold rolling annealing by means of SEM heating stage.

4. Summary

Complex investigations of microstructural changes during annealing of the material after derformation can be carried out by means of in-situ heating experiments combined with orientation maps in TEM and SEM. Essential for proper analysis of obtained results are parallel calorimetric study. Applied methods are powerful tools to provide complex information about material behaviour during annealing.

5. References

[Bayley, 1960] J.E. Bailey *„Electron microscope observations on the annealing process occuring in cold-worked silver”* Phil. Mag. 5 s. 833 (1960)
[Bieda, 2010] M. Bieda, K. Sztwiertnia, A. Korneva, T. Czeppe, R. Orlicki – „Orientation Mapping Study On The Inhomogeneous Microstructure Evolution During Annealing of 6013 Aluminum Alloy” Solid State Phenomena Vol. 163 (2010) 13-18
[Brisset, 2013] F. Brisset, A-L. Helbert, T. Baudin Microscopy and Microanalysis 969-977, 19 (2013)
[Humphreys, 1996] F.J. Humphreys, M. Ferry Mater Sci Forum 217–222, 529(1996)
[Humphreys, 2002] F. J. Humphreys, M. Hatherly *„Recrystallization and Related Annealing Phenomena”* Pergamon (2002)
[Hurley, 2004] P.J. Hurley, F.J. Humphreys J Microsc 213,225 (2004)
[Lens, 2005] A. Lens, C. Maurice, J.H. Driver Mater Sci Eng A 403, 144(2005)
[Lehtinen, 1973] B. Lehtinen, W. Roberts „*In Situ Observations of Precipitation and Recrystallization With a 1-MeV Electron Microscope”* J. Microsc. 97 s. 197 (1973)
[Mullins, 1958] W.W. Mullins *„The Effect of Thermal Grooving on Grain Boundary Motion”* Acta metall. 6 s. 414 (1958)
[Philips, 1973] instrukcja do uchwytu grzejnego firmy Philips (1973)
[Roberts, 1972] W. Roberts, B. Lehtinen *„On the feasibility of in situ observations of recrystallization in the high voltage electron microscope”* Phil. Mag. 26 s.1153 (1972)

[Sinclair, 1993] R. Sinclair, J. Morgiel, A.S. Kirtikar, I.-W. Wu, A. Chiang *„Direct observation of crystallization in sillicon by in situ high-resolution electron microscopy”* Ultramicroscopy 51, s. 41 (1993)
[Sztwiertnia, 1994] K. Sztwiertnia, F. Haessner *„In situ observations of the initial stage of recrystallization of highly rolled phosphous copper”* Mater. Sci. Forum, 157-162, s.1069 (1994)
[Sztwiertnia, 2006] K. Sztwiertnia, M. Bieda, G. Sawina *„Charakterystyki dezorientacji migrujących granic w wyżarzonym „in situ” mosiądzu α”* s.747 Materiały konferencyjne „Metalurgia 2006” Krynica (2006)
[Sztwiertnia, 2013] K. Sztwiertnia, M.Bieda, A.Korneva Continuous and discontinuous recrystallization of 6013 aluminum alloy Materials Science Forum Vol. 753 (2013) pp 221-224
[Wright, 2009] S. Wright and M. Nowell *Review of In Situ EBSD Studies* A.J. Schwartz et al. (eds.), Electron Backscatter Diffraction in Materials Science, Springer 2009A.