The 8th International Conference of the Slovenian Society for Non-Destructive Testing
»Application of Contemporary Non-Destructive Testing in Engineering«
***September 1-3, 2005, Portorož, Slovenia*, pp. 251-260**

MAGNETIC MICRO-PROBE DETERMINATION OF THE CONDITION OF MARAGING STEEL

M. Zupančič, J. Grum

University of Ljubljana, Faculty of Mechanical Engineering,
Aškerčeva 6, 1000 Ljubljana, Slovenia,
E-mails: martin.zupancic@fs.uni-lj.si, janez.grum@fs.uni-lj.si

ABSTRACT

Microstructural and mechanical properties of precipitation-hardened maraging steel could be stated on a basis of magnetic properties of such a material. A correlation between mechanical and magnetic properties of the maraging steel is treated from the view of non-destructive testing during and after heat treatment as well as during use under unfavourable conditions at elevated operating temperatures.

A group of maraging steels shows numerous technical and technological properties, which substantially differ from those of classical steels, i.e. excellent weldability, high ductility in soft state, high hardening ability and favourable suitability for EDM machining. All of those properties are extremely important in machining of technologically exacting products such as die casting dies. The selected maraging steel contains 12 % of nickel and is primarily intended for the production of hot-working dies and various machine elements exposed to heavy mechanical and thermo-mechanical loads.

Non-destructive testing of maraging steels enables cheaper and more reliable production. Selected characteristic properties for testing of maraging steel after precipitation hardening are impact toughness, hardness, and residual magnetism. Supervision should be performed not only during precipitation hardening but also during the use of such a material at elevated temperatures at which additional precipitation may occur, which could lead to overaging of the micro-structure. A fast and reliable method of non-destructive testing of mechanical properties could significantly affect the time, price and quality of production as well as maintenance of products made of maraging steel.

***Keywords**:* Precipitation hardening, Maraging steel, Impact toughness, Hardness, Magnetization curve, Residual magnetism

1. Introduction

Maraging steels with lowered content of nickel show also a higher temperature of martensitic transformation and are therefore more suitable for demanding applications, such as different tools and die casting dies. Final properties of those steels are attained with the diffusion process that forms precipitates during precipitation hardening. Different precipitates have a size ranging from some atoms up to several hundred nanometres, which depends on a local content of individual elements and temperature/time conditions of precipitation hardening. Differences in the structure of crystal lattices and matrix induce strains, leading to the formation of local residual stresses of the third and the second order. The residual stresses and strains occurring in the vicinity of the precipitates result in distinctive mechanical and technological properties of maraging steels. Some of these properties include isotropy after heat treatment, high tensile strength, high hardness, and a favourable ratio of tensile strength to fracture toughness. Precipitates formed during precipitation hardening substantially change also the size and behaviour of micro-magnetic domains. Consequently, the movement of micro-magnetic boundaries or Bloch walls during magnetization will change too. Influence of the precipitations on the micro magnetic domains enables the use of non-destructive magnetic testing methods for searching optimal combinations of mechanical properties after precipitation hardening. Such combinations could be achieved with appropriate temperature and time conditions, temperature variations during heating and cooling and, in special cases, also with several consecutive heating/cooling procedures.

An increasing number of maraging steel applications in manufacture of die casting tools increases the importance of simple and fast non-destructive testing of materials concerned using the magnetic method. Measurement of magnetic properties as a function of the material state obtained in the precipitation process provides an option for non-destructive testing of ferromagnetic maraging steel. The measurement results obtained confirm that a magnetization curve can serve as a basis of statistically reliable prediction of relations between magnetic and mechanical properties of a certain material. The results of the mechanical and magnetic properties of the applied maraging steel were used to develop a new method of non-destructive testing.

The past studies have shown that mechanical properties of a certain ferromagnetic material can efficiently be related to the magnetic properties via the basic parameters of the magnetization curve. The basic magnetic properties include, for example, coercivity H_c, residual magnetic-flux density B_r, permeability μ_r and μ_m, as well as an area and flattening of a hysteresis loop defined by A and η. In technical literature relations between mechanical properties and coercivity H_c, residual magnetic-flux density B_r and saturation flux density B_s are described.

Hardness of quenched and tempered steel in relation to magnetic properties of steel was analysed by Bussiere [1], who found a favourable relationship between hardness and coercivity H_c, and a less favourable one between hardness and residual magnetic-flux density B_r.

Kröning [2] treated a testing method in a batch production of induction-hardened steel shafts. Measurements of amplitude values U_A of a Barkhausen-noise voltage signal U_{BN} as well as an envelope of a voltage signal M_{max} were applied to control material hardness. In the shaft production, shafts to be discarded could be set apart during the production cycle, which lowered a cost price of the shafts.

Mitra [3] treated the martensite content in austenitic stainless steel in relation to variations of the magnetization curve. It turned out that the magnetic method was not the most reliable method in this case since the martensitic phase could be perceived only when its content exceeded 18 %. Moriya [4] measured characteristics at the magnetization curve to determine abrasion and corrosion damages to supporting steel wires. A difference between indirectly magnetically determined and actually measured results of a wire cross-section was in the order of magnitude of up to 1.4%.

The micro-magnetic method of measuring the induction as a consequence of directing individual micro-magnetic walls is a bit more demanding. The micro magnetic domains in ferromagnetic materials, known as Weiss domains comprise volumes in a range of 10^{-5} mm^3. For the first time the domains were proved by a German professor Barkhausen and since then numerous publications in that field are available, e.g. [5].

On the basis of the measurements performed at the selected maraging steel, a statistical relation was determined between the impact toughness, hardness and magnetic flux Φ_r at the point of the residual magnetic-flux density B_r.

2. Experimental procedure

2.1 *Preparation of material and precipitation hardening*

The maraging steel with the composition given in Table 1 was selected for measurements. The relatively low nickel content in comparison to many other maraging steels provides high resistance to reverted austenite. This provides higher resistance to elevated temperatures of employment, which is especially important with highly thermo-mechanical loaded die-casting dies.

Table 1: Chemical composition of the maraging steel concerned.

Elements [wt. %]					
C	Mo	Ni	Co	Ti	Al
< 0.02	8.0	12.0	8.0	0.5	~0.1

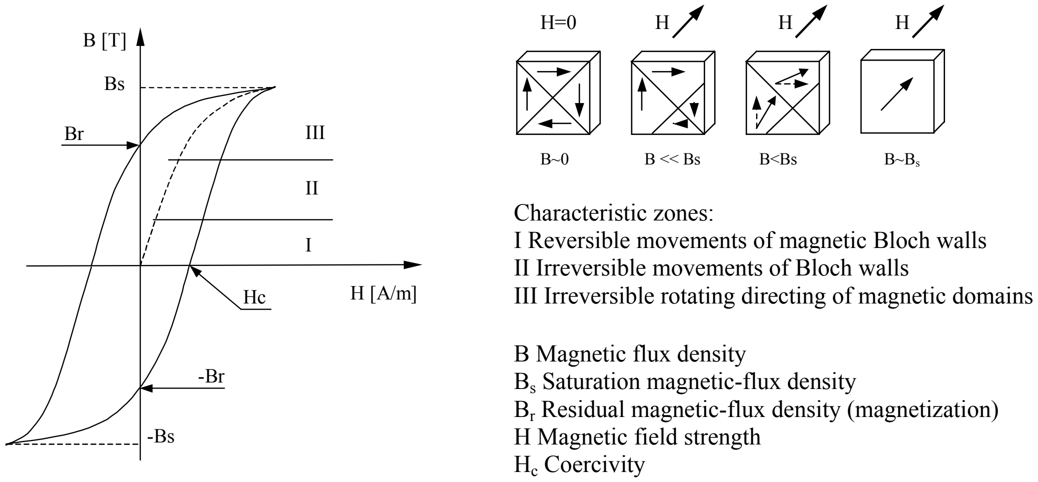
All samples were subjected to the same method of precipitation annealing; only temperature and time were varied. Precipitation hardening was accomplished with eight different temperature conditions and eight different time conditions. The samples were thus subjected to 64 different temperature/time conditions in total, i.e.:

- Precipitation annealing in accordance with conditions recommended by the manufacturer (450, 490, and 550 °C; duration: 2 h);
- Precipitation annealing at low temperatures (410 and 425 °C; duration: 10 min, 15 min, 1, 2, 6, 10, 20, and 50 hours);
- Precipitation annealing at elevated temperatures (600 and 650 °C; duration: 10 min, 15 min, 1, 2, 6, 10, 20, and 50 hours).

The precipitation-hardening conditions chosen for the magnetization curve and the attained hardness and impact toughness are evident from Fig. 4.

2.2 *Demagnetization and magnetization of the samples*

Fig. 1 shows the magnetization curve with the characteristic features that are a basis for a comparison of magnetic properties of different ferromagnetic materials.



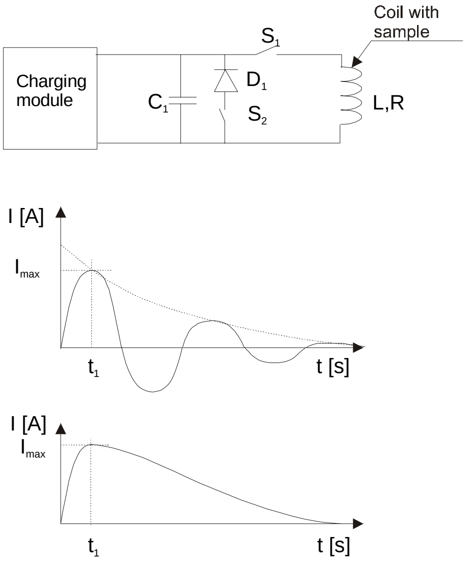
Characteristic zones:
I Reversible movements of magnetic Bloch walls
II Irreversible movements of Bloch walls
III Irreversible rotating directing of magnetic domains

B Magnetic flux density
B_s Saturation magnetic-flux density
B_r Residual magnetic-flux density (magnetization)
H Magnetic field strength
H_c Coercivity

Fig. 1: Magnetization curve of ferromagnetic material with characteristic zones and values [6].

Residual magnetic flux Φ_r in relation to conditions of the precipitation hardening was taken into account in measurement of characteristic magnetic properties of the selected steel. There are many factors influencing results of magnetic measurements, which include a local material composition and microstructure, a sample size, sample geometry and other influences. The influence of geometry could be diminished with a relatively small size of magnetic measurement coil with relation to the sample size; alternatively one could also produce samples with the same geometry. With our experiment the second alternative was considered.

The measurements showed a strong dependence of the magnetic flux on geometrical properties of the samples. To eliminate this influence, additional machining, i.e., grinding, was accomplished. Prior to measurement of the magnetic properties, the samples were demagnetized so that the same initial values were obtained for all the samples. Fig. 2 shows demagnetization /magnetization circuit. The magnetic measurements were accomplished using a magnetic flux gauge Magnetic Instrumentation ML40D.



R = 96.7 mΩ
L = 259.8 μH
n = 121 coils

Demagnetization: Initial voltage U_C (t=0) = 500 V

$$U_C(t) = R \cdot I_L(t) + L\frac{dI_L(t)}{dt}$$

$$I_L(t) = I_C(t) = -C\frac{dU_C}{dt}$$

Magnetization: I_{max} = 150, 160 and 360 A

$$I_{\max} = \frac{U_{C0}}{\omega L}e^{-\frac{\pi}{2\omega\tau}}; \quad I(t > t_1) = I_{\max}e^{-\delta t}; \quad \delta = \frac{R}{2L}$$

Fig. 2: A system for demagnetization and magnetization of the specimen.

2.3 Execution of the measurements

Mechanical and magnetic properties were measured on the samples after precipitation hardening. The characteristic properties chosen were Rockwell hardness at three selected points on each sample as well as impact toughness and magnetic flux Φ_r at the point of the residual magnetic-flux density B_r.

Each sample was demagnetized after precipitation annealing under different temperature/time conditions. Afterwards it was inserted in the magnetizing coil and a magnetic field of a certain strength H [A/m] was established. The magnetized sample was then inserted in the measuring coil and magnetic flux Φ_r [Wb] was measured. An experimental setup is shown in Fig. 3.

3. Experimental results

3.1 Hardness and impact toughness after precipitation hardening

The selected steel X2NiCoMoTi 12 8 8 belongs to a group of maraging steels characterised by a formation of precipitated phases in the course of precipitation annealing. Its microstructure changes significantly during the precipitation process, which affects also mechanical properties. Thus the chosen steel could be substantially deformed when in a “soft state” using high degrees of plastic deformations. Initial hardness amounts to a value of 31 to 32 HRC, tensile strength to 1020 MPa, and impact toughness to around 1.13 J/mm^2.

The steel shall be precipitation-hardened after machining and, consequently, hardness values exceeding 53 HRC and tensile strength exceeding 1950 MPa will be obtained. On the contrary, in the course of precipitation annealing impact toughness will reduce even down to 0.21 J/mm^2, as shown in Fig. 4. Stated data apply only to test specimens Charpy U having a size of 6x6x50 mm.

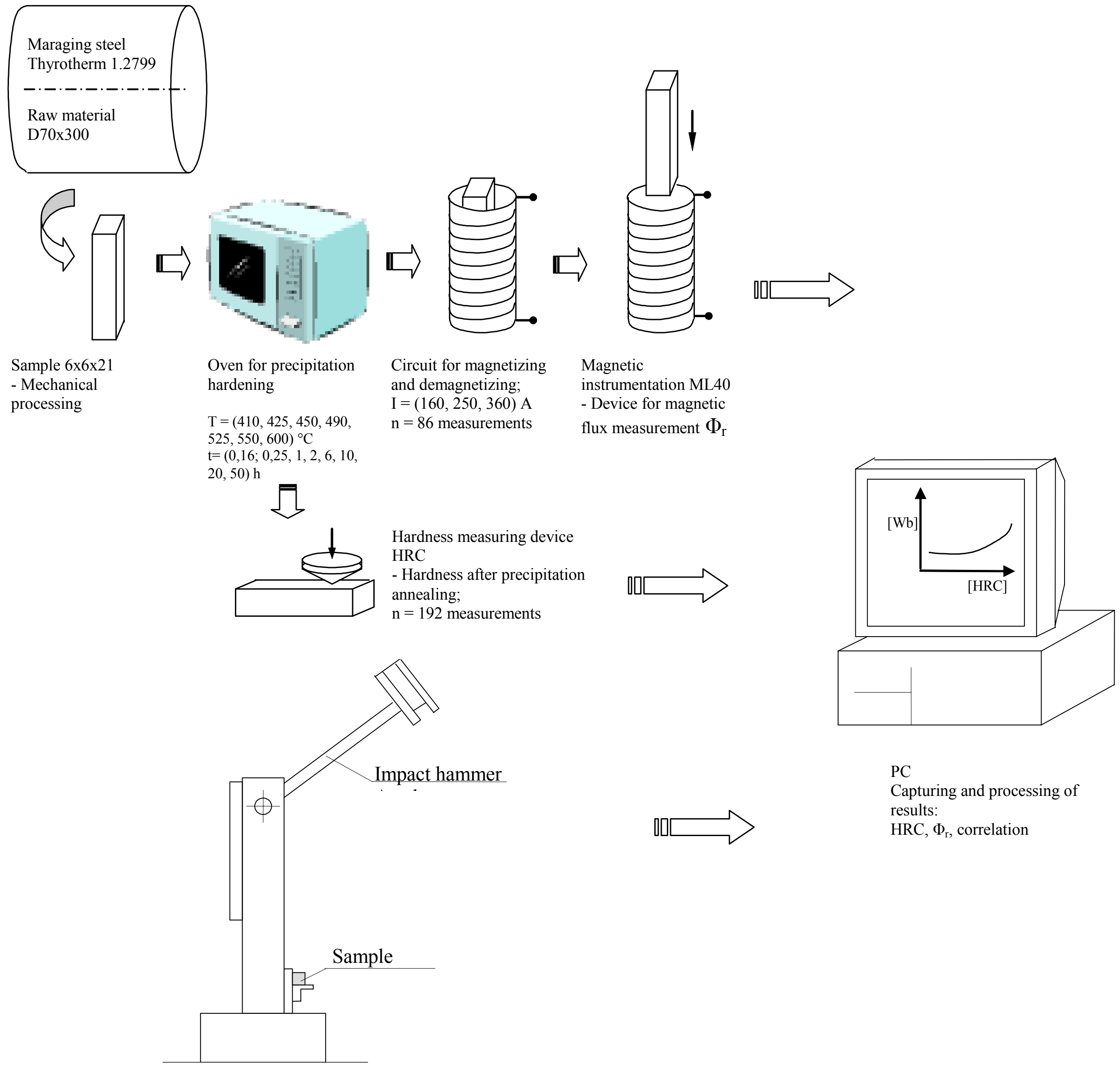


Fig. 3: An experimental system for measurement of mechanical and magnetic properties.

The temperature of precipitation hardening has significant effect on the diffusion process and, consequently also on hardening as represented in Fig. 4. According to data in the diagram it can be confirmed that there is a strong relation between the temperature of precipitation hardening and promptness of a change of hardness or impact toughness. A similar relation is valid also for other mechanical properties, for example tensile strength and elongation, which was also measured during testing [7].

High temperatures of precipitation hardening could produce a fast change of mechanical properties. Thus it is possible to reach maximum achievable hardness at 550 °C after 1 hour precipitation annealing whereas this time is shortened to only 15 minutes at 600 °C as shown in Fig. 5. Such results agree with our knowledge of the diffusion process and expectation that the temperature is a leading influencing factor of occurrence and growth of the precipitates. At temperatures below 400 °C the mentioned processes are much slower but also present and they could sometimes significantly change properties of the maraging steel used for applications in hot environment. Strong dependence of the precipitation process on the temperature requires appropriate conditions of employment to prevent overaging of the microstructure. Generally, overaging could be defined as a microstructural state when there is a decrease of hardness or tensile strength perceived due to changing and coarsening of the precipitates.

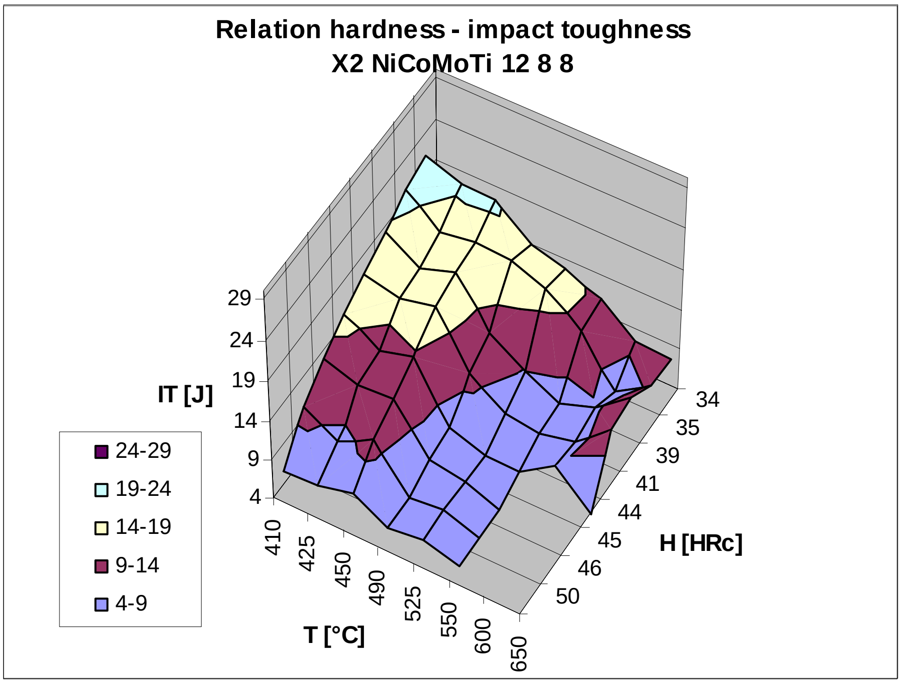


Fig. 4: Hardness and impact toughness variation of the selected maraging steel according to conditions of precipitation hardening.

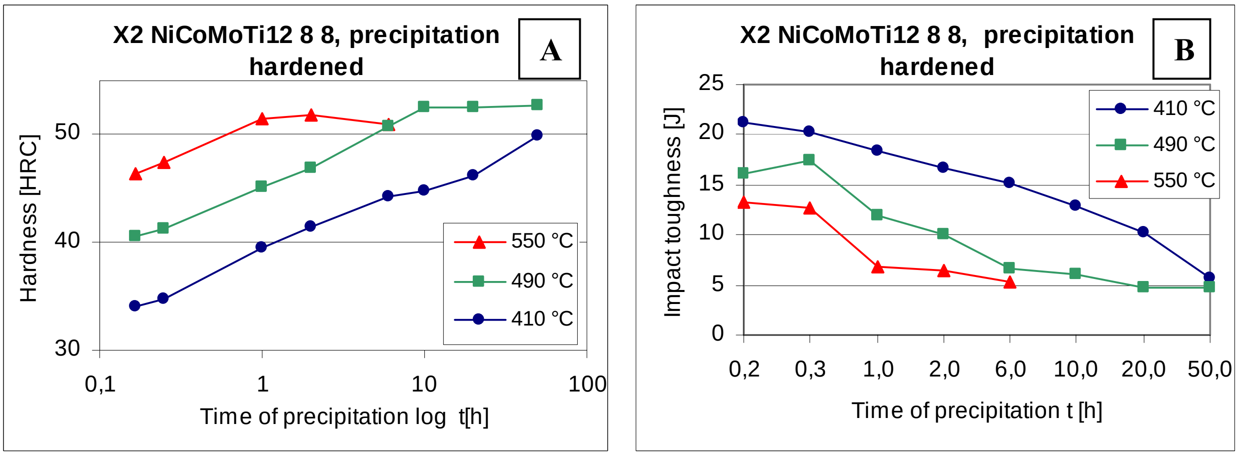


Fig. 5: A) Hardness and B) Impact toughness with regard to selected temperature and time conditions of precipitation hardening.

3.2 *Magnetic flux*

Measurements of residual magnetism, i.e., relevant magnetic flux Φ_r, were performed at three the most characteristic precipitation-annealing temperatures selected, i.e., annealing in a lower temperature range, that is at 410 °C, annealing in a medium range, that is at 490 °C, and annealing at an upper limit of a range recommended, that is at 550 °C. The precipitation-annealing temperatures below 410 °C require exceptionally long annealing times, which, however, are not fit for use in the industrial practice. At the comparatively elevated temperatures, i.e., above 550 °C, in a relatively short time, overaged microstructure is obtained, which is accompanied by coarse precipitated phases. In practice high precipitation-annealing temperatures shall be avoided to eliminate difficulties in the control of precipitation annealing.

Fig. 6 shows the measured magnetic flux Φ_r in relation to the precipitation-annealing conditions. With low temperatures of precipitation annealing, magnetic flux Φ_r changes only insignificantly with longer precipitation times. This can be expected since at the lower precipitation temperatures diffusion rates are very low. Under such conditions the precipitates are usually small.

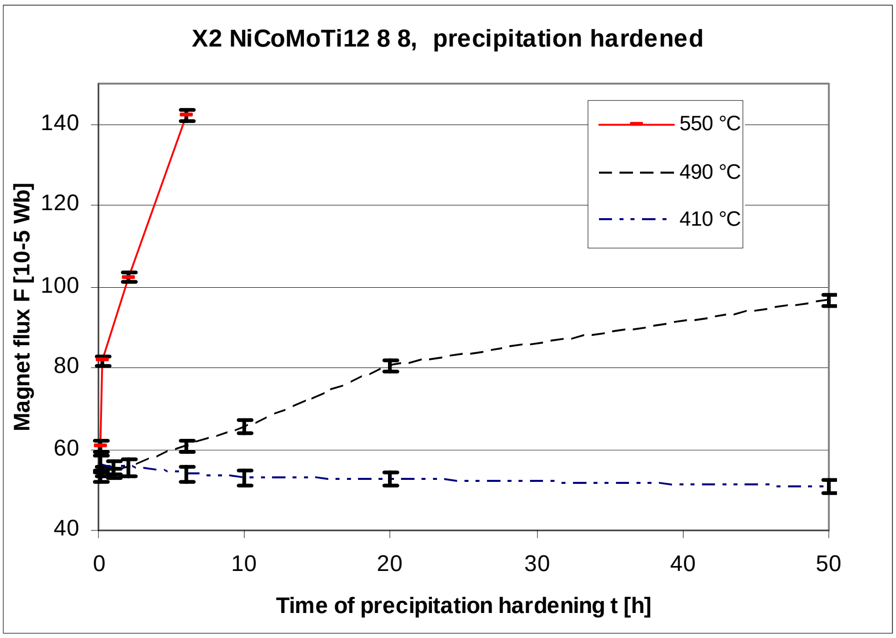


Fig. 6: Measured magnetic flux Φ_r as a function of precipitation-annealing conditions. Each point in the diagram also shows deviations related to sample orientation.

The elevated temperatures of the precipitation produce major changes in material hardness and considerably change magnetic flux Φ_r. Thus after 5 hours of precipitation at 490 °C, a change of magnetic flux Φ_r will amount to over 10% and at 550 °C to around 170% with regard to a thermally untreated state. Such strong differences are caused by the initiation and growth of precipitates and a local change of composition both in precipitates and matrix.

Results of the measurements indicate that the magnetic method may be applied as a fast and relatively cheap control to prevent overaging. In all cases of the measurements a higher precipitation rate essentially increased the magnetic flux Φ_r. In practice the only problem encountered would be overlapping of individual results for lower precipitation rates, i.e., low temperatures and short precipitation times, but in such cases there is no risk of overaging.

It turned out that the magnetic method proposed is so sensitive that very small changes of material properties can be detected. Measurements were therefore repeated at the same sample by inserting it into the measuring coil at an angle of 180° with regard to the initial measurement. In this way anisotropic material properties emanating from differences in microstructure showed. Fig. 6 shows also an influence of the sample orientation on measured magnetic flux Φ_r. The changes of magnetic flux Φ_r due to the sample orientation for individual spots are shown as deviations from the mean value.

The magnetic flux measurements confirmed that differences in magnetic flux, with regard to sample orientation in the coil, were small. The differences measured amounted to 2.8% with the precipitation-annealing temperature of 550 °C and to 5.7% with 410 °C. From the results it is evident that the influence on the anisotropy decreases with the precipitates formed at elevated precipitation-annealing temperatures.

3.3 Influence of magnetization current

The stated influence of the magnetization current on residual magnetism was very small. For example, with magnetization currents of 150 and 360 A at the precipitation-annealing temperature of 450 °C, values indicated in Table 2 were obtained. The table indicates that the differences between magnetic fluxes Φ_r with each magnetization current amount to less than 0.9%.

Table 2: The influence of magnetization current on magnetic flux Φ_r.

Magnetic flux Φ_r [10-5 Wb]						
Magnetization current I [A]	Temperature T = 450 °C **Precipitation-annealing time t [h]**					
	0,25	**2**	**6**	**10**	**20**	**50**
150	54,6	53,8	53,2	53,8	50,85	50,75
360	54,15	53,95	53,15	53,65	50,9	50,35
Difference [%]	**0,82**	**0,28**	**0,09**	**0,28**	**0,10**	**0,79**

3.4 Calibration curves for different precipitation-annealing conditions

Efficient introduction of the magnetic method of non-destructive testing of maraging steel requires determination of a relationship among the magnetic and mechanical properties after precipitation annealing with certain parameters. For this purpose hardness, impact toughness and residual magnetism were chosen as properties to be compared. A calibration curve could be used for non-destructive testing of maraging steel during precipitation annealing or during employment of a hot-working tool. For the precipitation-hardening process it is possible to control the annealing time at a chosen temperature or to control the influence of temperature on the precipitation processes as well as to assure a safe margin against overaging.

The statistical relationship obtained between HRC or impact toughness and magnetic flux Φ_r is defined by a linear approximation and dispersion of the measured magnetic flux around a straight line. A dispersion of the results around the approximated straight line is defined by a correlation coefficient R. The results of the measurements shown in Fig. 7A indicate that a relation between hardness and magnetic flux is favourable under selected temperature/time conditions of precipitation annealing since the correlation coefficients R are ranging from 0.74 to 0.94. An even better correlation is achieved between impact toughness and magnetic flux with R ranging from 075 and 0.975, Fig. 7B.

The results show that the magnetic method is suitable for non-destructive testing of the surface layer of die-casting tools. During operation of such tools each die casting cycle causes a temperature cycle, which is exacting for the surface layer of the tool. This is especially demanding for tools operating at high temperatures. Thermal cycles cause fatigue and may also provoke precipitation processes. Non-destructive magnetic testing is an elegant method to test a progress of the precipitation process in the surface layer, which could lead to an undesirable failure of such a tool. The proposed testing method could provide reliable information related to the timely maintenance or service operation at such a tool.

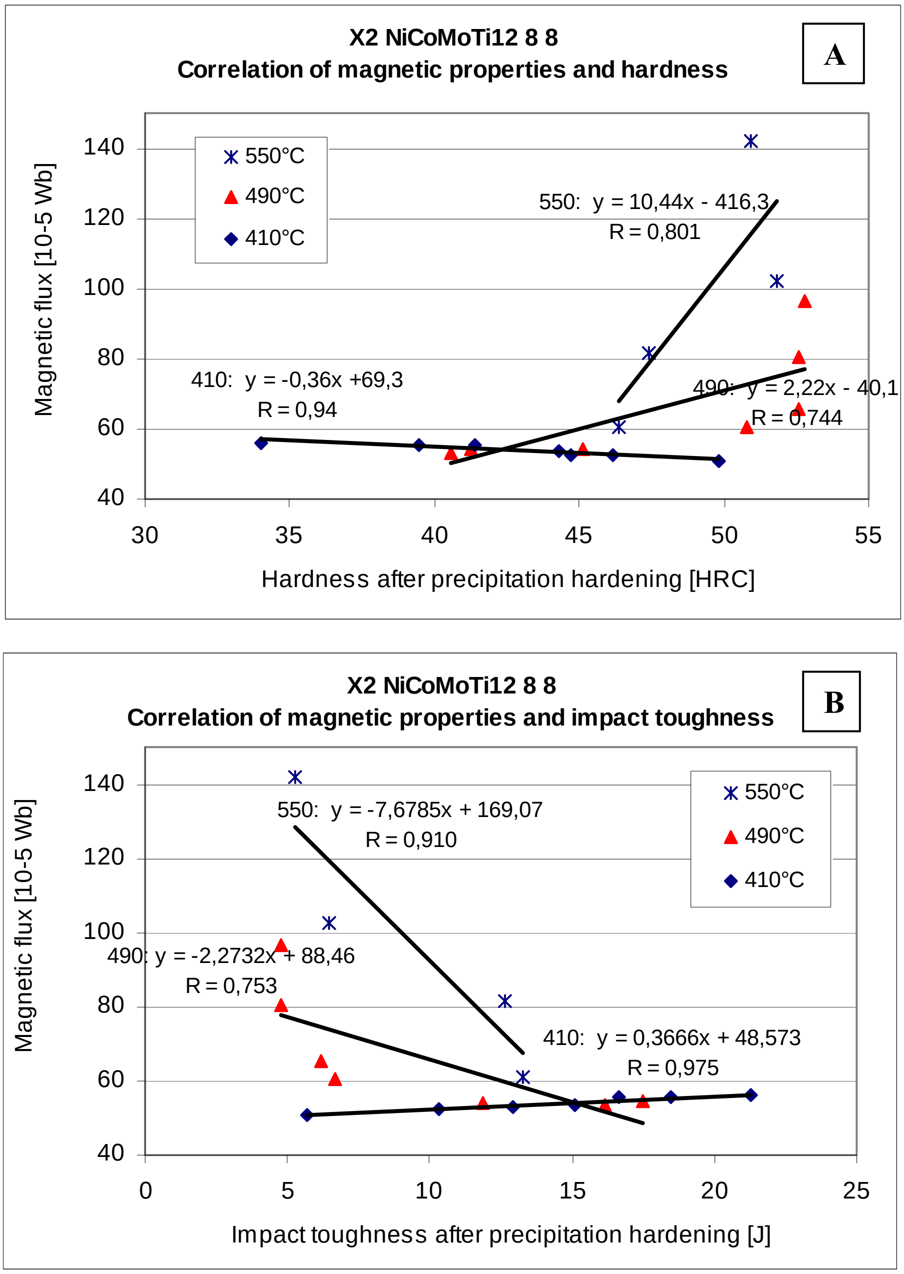


Fig. 7: A) Correlation between hardness and magnetic flux Φ_r. B) Correlation between impact toughness and magnetic flux Φ_r for selected temperature-time conditions.

Difficulties encountered in the application of the magnetic method with the given measurement method may result from small and non-uniform changes of residual magnetism at lower precipitation-annealing temperatures. Below 450 °C, magnetic flux Φ_r is, namely, a bit reduced with longer precipitation-annealing times. This phenomenon may be related to a decrease in Ni and Mo contents in the matrix due to the formation of Ni_3Mo and Ni_3Ti precipitates, which was confirmed by separate and additional microstructural and microchemical analyses of the samples concerned. The precipitates had an extremely small size, i.e., up to some 10 nm, due to a relatively low precipitation-annealing temperature. Consequently, their influence on residual magnetism is essentially different from that with higher temperatures of hardening. Due to the low precipitation-annealing temperature, the diffusion processes were slow, which resulted in a high density of the fine distributed precipitates. Thus at lower temperatures, the change of the magnetic properties was more strongly affected by deviations in the chemical composition and other influences than by the changes of microstructure due to precipitation annealing.

4. Conclusions

With increasingly wide use of maraging steels in manufacturing of complex die-casting tools in last years, a higher number of non-destructive testing methods can be expected to be required for determination of a material state and properties during precipitation hardening, as well as during employment at high temperatures and also after service operations.

Measurements of magnetic properties provide a fast, relatively cheap and reliable method, which could be used for such a purpose; as such it has its future in the field of testing die-casting dies.

Precipitation annealing of the selected maraging steel with 12% Ni was performed with the characteristic temperature/time parameters to obtain the mechanical properties that can be efficiently related to the magnetic properties of the material. The selected properties are defined with the calibration curves hardness - magnetic flux Φ_r and impact toughness - magnetic flux Φ_r at the point of residual magnetic flux density B_r .

Maximum hardness achieved is, expectedly, a function of the precipitation-annealing temperature and time and can be increased from initial 32 HRC to around 53 HRC. On the contrary, impact toughness decreases from 1.13 J/mm^2 to about 0.21 J/mm^2.

The study showed that residual magnetism strongly depended on the precipitation-annealing temperature and time. Thus after 5 hours of precipitation at 490 °C, the change of magnetic flux Φ_r amounted to over 10% whereas at 550 °C to around 170% with regard to the thermally untreated state. At the medium and elevated precipitation-annealing temperatures, the precipitates are considerably coarser and exert a stronger influence on the mechanical properties.

A correlation coefficient R between hardness and residual magnetism for the selected precipitation-annealing conditions ranged from 0.74 to 0.94 while a correlation between impact toughness and residual magnetism amounted 0.75 to 0.975. Such correlations between hardness and residual magnetism after precipitation annealing of different samples are satisfactory to control the precipitation-annealing process or the material state during operation.

5. References

[1] Bussiere J. F.: On-line Measurement of the Microstructure and Mechanical Properties of Steel, Materials Evaluation, Vol. 44, No. 1, 1986, 560-567.

[2] Kröning M.: Benefits and Challenges of the Integration of Nondestructive Testing Into Material and Process Engineering, 14th WCNDT, New Delhi, 1996, 87-92.

[3] Mitra A., Sagar S. P., Bhattacharya D.K.: Evaluation of Magnetic Phases in Cold Worked and Weldments of AISI 304 Stainless Steel, 15th WCNDT, Rome, 2000.

[4] Moriya T., Tsukada K., Hanasaki K.: A Magnetic Method for Evaluation of the Deterioration of Large Diameter Wire Ropes, 15th WCNDT, Rome, 2000.

[5] Langman R.: Some Comparisons between the Measurements of Stress in Mild Steel by Means of Barkhausen Noise and Rotation of Magnetization, NDT International, Vol. 20, No. 2, 1987, 93-95.

[6] Shackelford F. J.: Introduction to Materials Science for Engineers, 5th Edition, Prentice Hall, 2000, 670-703.

[7] Grum J., Zupančič M.: Suitability assessment of replacement of conventional hot-working steels with maraging steel. Part 1, Mechanical properties of maraging steel after precipitation hardening treatment, *Z. Met.kd.*, 2002, Vol. 93, No. 2, 164-170.