In situ characterization of phase transformations in a magnetic field in Fe-Ni alloys

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Abstract. We have investigated the effect of magnetic field on the austenite(\(\gamma\))-ferrite(\(\alpha\)) equilibrium in Fe-xNi alloys with x = 0, 2, 4 wt%. The \(\alpha\rightarrow\gamma\) and \(\gamma\rightarrow\alpha\) transformations have been followed as a function of applied magnetic field by a laser dilatometer installed in a 16T superconducting magnet. In addition, magnetic measurements at high temperature have been used to follow the magnetic behavior of each alloy composition during a complete heat treatment.

We observe a shift of the phase diagram to higher temperature as the magnetic field is increased. We also find that the \(\alpha\)-phase is either in the paramagnetic or in the ferromagnetic state as the transformation proceeds, depending on the amount of Ni. This results in an increase of the transformation temperature which is respectively proportional to the magnetic field if ferrite is formed in the ferromagnetic state and proportional to the square of the magnetic field if ferrite is paramagnetic.

1. Introduction
Modification of thermal treatments by an applied magnetic field in the processing of iron based alloys has been growing substantially in the last decade [1-15]. The goal of such processing is to achieve major improvements in material properties that cannot be obtained through conventional thermo-mechanical treatments. The relevant advances in magnet materials and design technologies can now provide magnetic fields of sufficient magnitude for new effect to be highlighted. Among these effects, modifications of phase stability [1-4], phase diagram boundaries [5-7], and transformation kinetics [8-10] have been observed when the involved phases exhibit different magnetization behaviors. Several papers report on theoretical approaches to study the impact of external magnetic fields on \(\gamma/\alpha\) phase equilibria in Fe-C, Fe-X and Fe-C-X alloys [10-15]. These studies use the Weiss Molecular Field (WMF) assumptions [16] together with the Curie Weiss law to evaluate the change in the Gibbs Free energy of the individual phases and to calculate a field-modified phase diagram. All these studies result in a similar estimation of the magnetic field effect on the \(\gamma/\alpha\) equilibrium temperature. According to them, the equilibrium temperature is raised from 1 to 3°C per Tesla depending on the alloy composition and magnetic field intensity.

Among various Fe based alloys, Fe-Ni alloys with low Ni contents are of special interest because the Curie temperature of the \(\alpha\) phase (\(T_C\)) can be tuned by the alloy composition. Concomitantly, the \(\gamma\rightarrow\alpha\) and \(\alpha\rightarrow\gamma\) transformation temperatures are also changed. As an example, for Ni contents above 3%, the Curie point of the \(\alpha\) phase becomes higher than the structural transformation temperature [17]. Therefore we can expect that the magnetic field dependence of the transformation temperature (\(T_T\)) will differ depending on the value of \(T_C-T_T\). In the present work, we examine the \(\alpha\rightarrow\gamma\) and \(\gamma\rightarrow\alpha\)
transformation temperatures in Fe-xNi alloys with x = 0, 2, 4 wt% by dilatometry measurements up to 16 T. In addition, magnetic measurements at high temperature are used to follow the magnetic behavior of each alloy composition during a complete heat treatment and to evaluate the magnetic state of ferrite during transformation.

2. Experimental procedure
Ingots of Fe-Ni alloys with content of 0, 2 and 4 wt% are prepared by induction melting in a cold crucible under argon atmosphere and cast into a 10 mm diameter copper mold. The alloys have been treated for 48h at 700°C under vacuum for homogenization. Finally the ingots have been cold worked down to a diameter of 4 mm and cut into cylindrical specimens of 10mm in length.

Transformation temperatures in magnetic fields have been measured by dilatation measurements. This home-made in-situ measurement setup, together with the furnace, is installed in the room temperature bore of a 16T superconducting magnet [19]. In this system, homogeneous temperature and field are applied to the 1 cm long sample placed in the magnet center region. The dilatation of the specimens is measured by a Michelson interferometer across an airtight pyrex window. The beam is reflected by a spherical mirror fixed on the top of an alumina sample holder.

Specimens have been heated up to the maximum temperature with a heating rate of 3°C/min and this temperature has been kept constant for 30 min. The same rate has been applied upon cooling. The maximum temperature has been set at 30°C above the upper boundary of the two phase $\alpha + \gamma$ region [17]. This correspond to 940°C for pure Fe, 900°C for Fe-2Ni wt% and 850°C for the Fe-4Ni wt% alloys.

The magnetic behavior of the involved phases has been determined by the Faraday’s method using a magnetic balance. The measuring device together with the furnace are inserted in the vertical room temperature bore of a 7 Tesla superconducting magnet [19]. The sample temperature is measured with Pt/Pt-10%Rh thermocouples in direct contact with the sample. The maximum magnetic field obtained at the specimen location is about 4T with a magnetic field gradient of about 1.5T/cm.

The sample magnetization has been measured up to 1000°C with heating and cooling rates of 2°C/min. The magnetic field at the specimen location has been set to 2T with a magnetic field gradient of about 0.7T/cm.

3. Results
The dilatation signal of the Fe-2Ni wt% sample in a magnetic field of 16T is shown in Figure 1. The $\alpha \rightarrow \gamma$ phase transformation appears as a sharp contraction of the sample. The $\gamma \rightarrow \alpha$ phase transformation

![Figure 1. Dilatation measurement of Fe-2Ni wt% as a function of temperature in 16T.](image-url)
exhibits a sharp dilatation. For each $\alpha \rightarrow \gamma$ and $\gamma \rightarrow \alpha$ transformation, the onset of the transformation temperature ($T_S$) and the end of the transformation temperature ($T_E$) are evaluated by the tangents method. The transformation temperatures ($T_T$) are defined as $T_{\alpha \rightarrow \gamma}$ on heating and $T_{\gamma \rightarrow \alpha}$ on cooling. They are calculated as the mean value of each $T_E$ and $T_S$. The relative change of the transformation temperature due to the magnetic field ($B$), $\Delta T_{\alpha \rightarrow \gamma}$ and $\Delta T_{\gamma \rightarrow \alpha}$ are defined as:

$$\Delta T_{\alpha \rightarrow \gamma} = T_{\alpha \rightarrow \gamma}(B) - T_{\alpha \rightarrow \gamma}(0) \quad (1)$$

$$\Delta T_{\gamma \rightarrow \alpha} = T_{\gamma \rightarrow \alpha}(B) - T_{\gamma \rightarrow \alpha}(0) \quad (2)$$

As a reference, the transformation temperatures measured without any magnetic field $T_{\alpha \rightarrow \gamma}(0)$ and $T_{\gamma \rightarrow \alpha}(0)$ are given in Table 1.

Table 1. Measuring transformation temperatures on heating and cooling for Fe-Ni (0, 2 and 4 wt%) by dilatometry in zero field.

| wt% Ni | 0   | 2   | 4   |
|--------|-----|-----|-----|
| $T_{\alpha \rightarrow \gamma}(0) ^{\circ}C$ | 916 | 840 | 788 |
| $T_{\gamma \rightarrow \alpha}(0) ^{\circ}C$ | 903 | 775 | 695 |

The field dependences of $\Delta T_{\alpha \rightarrow \gamma}$ and $\Delta T_{\gamma \rightarrow \alpha}$ for the three alloys are plotted with solid marks in Figure 2(a) and 2(b) respectively.

Figure 2. Change in the (a) $\alpha \rightarrow \gamma$ and (b) $\gamma \rightarrow \alpha$ transformation temperature by the application of magnetic field.

Obviously, $T_{\alpha \rightarrow \gamma}$ and $T_{\gamma \rightarrow \alpha}$ increase with increasing the magnetic field intensity for the three alloys. The increase of $T_{\alpha \rightarrow \gamma}$ amounts to 17$^\circ$C in pure Fe, 24$^\circ$C in the Fe-2Ni wt% alloy and 29$^\circ$C in the Fe-4Ni wt% alloy in a magnetic field of 16T. This increase seems to be proportional to $B^2$ for the three alloys compositions. A similar increase is observed for the $T_{\gamma \rightarrow \alpha}$ temperature. This increase seems to
be also proportional to $B^2$ for the pure iron and the 2wt% Ni alloys composition, whereas it tends towards a linear behavior for the 4wt% Ni alloy.

Figure 3 shows the magnetization curves as a function of the temperature obtained for the three alloys upon heating in Figure 3(a) and cooling in Figure 3(b). Upon heating, a sharp decrease in the magnetic signals is associated to the respective Curie points of pure iron (770°C), Fe-2Ni wt% (764°C) and Fe-4Ni wt% (759°C) given by [13]. At higher temperature, the $\alpha \rightarrow \gamma$ transformation is observed as a small decrease in the magnetic signal. This effect can hardly be seen in the Fe-4Ni wt% alloy because the magnetic transition occurs in the same temperature range as the $\alpha \rightarrow \gamma$ transformation. Upon cooling, the $\gamma \rightarrow \alpha$ transformations and the magnetic transitions are well dissociated in pure Fe. In the Fe-2Ni wt% and Fe-4Ni wt% alloy composition, ferrite appears in its ferromagnetic state thus leading to a sharp increase in the magnetic signal.

4. Discussion

The effect of a high magnetic field on the respective stability of the ferrite ($\alpha$) and austenite ($\gamma$) phases can be qualitatively explained in term of the magnetic contribution to the total Gibbs energy of each phase. When a magnetic induction, $B$, is applied, the respective Gibbs free energy of ferrite and austenite are lowered due to the contribution of a magnetic energy term, $\Delta E_m$, to the total energy of each phase:

$$\Delta E_m = - \int_{0}^{B_{app}} M \ dB$$  \hspace{1cm} (3)

where $M$ is the field dependent magnetization projected along B. These respective contributions are proportional to the field dependence of the magnetization of each phase. As ferrite is more magnetic than austenite, this phase is stabilized, so that a shift in the equilibrium temperature is observed towards higher temperatures. Moreover, the magnetic contribution of the austenite phase to the Gibbs free energy can be neglected due to the very low magnetization of this paramagnetic phase.

The shift of the $\gamma \rightarrow \alpha$ transformation temperature was evidenced for all the alloy compositions studied in this work. However and depending on the Ni content, the variation of the equilibrium temperature shows two different behaviors with the magnetic field intensity: the transformation
temperature increases either linearly or follows a quadratic behavior with the applied magnetic field. These different variations are linked with the Curie temperature of the considered compound. When the $\gamma \rightarrow \alpha$ transition occurs below the Curie temperature of ferrite, the magnetization of the ferrite phase, $M$, is rapidly saturated so that the magnetic energy is roughly linear with the applied magnetic field value. On the contrary, when the $\gamma \rightarrow \alpha$ transition occurs above the Curie temperature, the ferrite is formed in the paramagnetic state and hence $M$ is almost linearly dependent on $B$. Since the magnetic energy is proportional both to the applied field and to the magnetization, the temperature shift is roughly proportional to the square of the field value. In this particular case, a more accurate analysis should use the Weiss Molecular Field model as the transformation occurs close to the Curie temperature of the ferrite phase. These two different behaviors have been recently reported in steels and cast irons [19] as well as in Fe-Co alloys [21].

5. Conclusion
In this work, a new experimental method based on in-situ measurements has been used to monitor the $\alpha \rightarrow \gamma$ and $\gamma \rightarrow \alpha$ transformations in Fe-Ni alloys as a function of applied magnetic field intensity. The transformation temperature is shifted towards higher temperatures in a magnetic field. This shift follows either a linear or quadratic behavior with the magnetic field intensity. Experimental determination of the magnetization of the ferrite phase at high temperature in various field values is in progress and will allow direct comparisons between WMF predictions and experiments.

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