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Hydrogen charging in nickel and iron and its effect on their magnetic properties

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Abstract

The current study was undertaken to explore the possibility of detecting hydrogen cavitation in magnetic materials through magnetic propertymeasurements. It is known that dissolved hydrogen in a material causes microvoids. These voids may affect the structure-sensitive magnetic properties such as coercivity and remanence. In this study, hydrogen was introduced into nickel and iron by two processes, namely thermal charging and cathodic charging. The effect on the magnetic properties was measured. In addition, the variation of the magnetic properties with porosity was studied.

Keywords

Electrical properties, Iron, Magnetic effects, Materials properties, Nickel

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The current study was undertaken to explore the possibility of detecting hydrogen cavitation in magnetic materials through magnetic property measurements. It is known that dissolved hydrogen in a material causes microvoids. These voids may affect the structure-sensitive magnetic properties such as coercivity and remanence. In this study, hydrogen was introduced into nickel and iron by two processes, namely thermal charging and cathodic charging. The effect on the magnetic properties was measured. In addition, the variation of the magnetic properties with porosity was studied. [S0021-8979(96)30608-3]

I. INTRODUCTION

The presence of hydrogen in metals is known to have a significant impact on the mechanical properties.[1] Generally, hydrogen is taken up in interstitial sites, but at higher concentrations it can lead to the formation of voids. Oriani[2] has confirmed that the exact nature of hydrogen dissolved in a metal is that of a proton which has given up its electron to the electron gas of the metal. The solubility of hydrogen in nickel and iron is proportional to the square root of the external hydrogen pressure, at least for small concentrations according to Sieverts.[3] Also the diffusion of hydrogen through a metal follows the Arhennius equation and is therefore much faster at higher temperatures. Lord[4] has suggested that hydrogen is more likely to reside in octahedral rather than the tetrahedral sites in iron.

When hydrogen is introduced into a lattice at high temperature and pressure and the material is then quenched back to ambient conditions, the lattice is no longer capable of retaining all of the hydrogen. The excess hydrogen is taken up by internal traps such as dislocations, grain boundaries, and vacancies. Thus the physical picture is that of hydrogen dissolved in the lattice with the excess amount residing in internal voids. The presence of dissolved hydrogen in a metal causes an expansion of its lattice. Smialowski[5] concluded that the irreversible external dimensional changes of iron samples caused when they are charged with hydrogen, are due to molecular hydrogen gas at very large pressures. However, detection of hydrogen in a material is very difficult due to its extremely small equilibrium lattice solubility of only 5×10^{-10} at normal temperature and pressure.

In a ferromagnetic material, the process of magnetization occurs by the movement of the domain walls. This motion can be hindered by the presence of pinning sites such as second phase particles and inclusions resulting in hysteresis. According to the model of Kersten and Neel,[6,7] the presence of nonmagnetic inclusions or voids causes pinning of magnetic domain walls and leads to an increase in the coercivity and a decrease in permeability. Baum[8] noted that the structure-sensitive properties such as coercivity and permeability appear to be dependent on the porosity of iron. More recently, Jiles *et al.*[9] have reported that the coercivity of iron is at maximum when the pores have a diameter of about 8–11 μm.

Oriani and Josephic[10] studied the effect of hydrogen on the plastic deformation of steel and reported that the basic effect is an increase in the population of the microvoids at a given strain. Hydrogen of very high fugacity nearly doubles the microvoid population. This causes an increase in the number of voids present in the material which acts as pinning sites, thus leading to changes in its magnetic properties. In addition, the presence of hydrogen causes local strain in the material, which also impedes the motion of the domain walls as suggested by Becker.[11] In view of these effects, an attempt has been made in the present work to study the effect of hydrogen on the magnetic properties of materials based on iron and nickel.

II. EXPERIMENTAL PROCEDURE

Hydrogen can be introduced into the metal in several ways. These include thermal charging, cathodic charging, and acid corrosion.[12] Of these methods the first two were used in the present work and the change in the magnetic properties was studied.

A. Thermal charging

Pure nickel rods of 10 mm diameter and 100 mm length were put in a chamber which was evacuated and then subjected to a pressure of 100 atm of hydrogen at 500 °C. The maximum solubility of hydrogen under these conditions was calculated as 0.0015 atoms of H_2/atom of nickel. The time taken for saturation of hydrogen was 2.5 h. During charging, the samples were subjected to hydrogen charging for different periods of time, causing varying uptakes of hydrogen. Baseline magnetic measurements were taken after annealing the sample at 500 °C for 12 h. Magnetization measurements were made using a magnetic hysteresisgraph. Results were analyzed using a software package written exclusively for the experimental system.

B. Cathodic charging

Cathodic charging has often been used to introduce hydrogen into metals because of its convenience and its ability to produce high fugacity of hydrogen. This is an electrolytic process in which hydrogen is charged into the metal cathodically using platinum wire as the anode. Boniszweski and Smith[13] have used this method to study the effect of hydro-

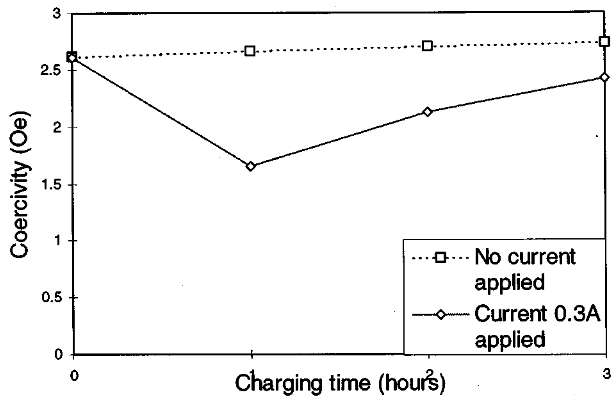

FIG. 1. Variation of coercivity of iron as a result of cathodic charging.

gen in the plastic deformation of nickel. In the current study charging was done in a 1 N sulfuric acid bath at room temperature with a current density of about 500 A/m^2 for different times. Thiourea was used as the catalyst, causing a hydrogen input fugacity of 10^7 MN/m^2. The magnetic properties were measured as a function of the amount of hydrogen in the sample. These are shown in Fig. 1.

III. RESULTS AND DISCUSSION

A. Thermal charging

The results of thermal charging are shown in Fig. 2. Initially, the coercivity and remanence decreased as expected,since a large number of dislocations and stress fields which act as pinning sites for the domain walls are removed as a result of annealing. Upon charging to 100 atm of hydrogen at 500 °C, the magnetic properties varied due to both the addition of hydrogen and exposure to high temperature which further relieved the residual stress. During the initial stages of charging, the main effect was simply the annealing of residual stress, thus the coercivity decreased. However, as the hydrogen content increased, it caused an increase in the density of voids and resulting internal stress and led to an increase in the coercivity. Quantitatively, the changes due to hydrogen charging were small, amounting to a maximum of 0.5 Oe. The small changes in the magnetic properties resulted from a low hydrogen intake which resulted in a small number of additional voids formed. Also, the excess hydrogen was preferentially chemically adsorbed on to inclusions and grain boundaries which acted as pinning sites even in the absence of hydrogen.[1] In these cases, the number of new pinning sites introduced by the hydrogen absorption would be small, leading to only a small increase in coercivity.

B. Cathodic charging

Hydrogen introduced by charging may affect the magnetic properties of iron-based alloys either by the formation of hydrides or by the formation of voids. Kimura and Birnbaum[14] have reported observations using transmission electron microscopy of small gas bubbles in iron cathodically charged at 20 A/m^2 which caused blisters to form due to the high pressure of hydrogen. According to Boniszweski and Smith,[13] hydrides could be formed on the surface layers of the material under certain charging conditions. It has also been reported in the literature that the thickness of the hydride layer formed on the surface by cathodic charging at room temperature for 48 h was as small as 10 μm. The high fugacity of hydrogen, and the corresponding high hydrogen concentration gradients, may therefore affect only the surface layers of the specimen. This has also been confirmed by Kimura and Birnbaum.[14] While cathodic charging of relatively thick specimens had relatively little effect on their measured flow stress, large effects were observed for smaller specimens with diameters of 0.1 mm or less. Thus it is established that the effect of hydrogen on the magnetic properties is entirely a surface phenomena. The measured hysteresis properties were however averaged values over the bulk sample and therefore these magnetic properties do not exhibit significant changes due to the addition of hydrogen.

IV. EFFECT OF POROSITY

In order to confirm these conclusions concerning the effect of hydrogen on the magnetic properties, it was decided to study the effect of uniformly distributed pores on the mag-

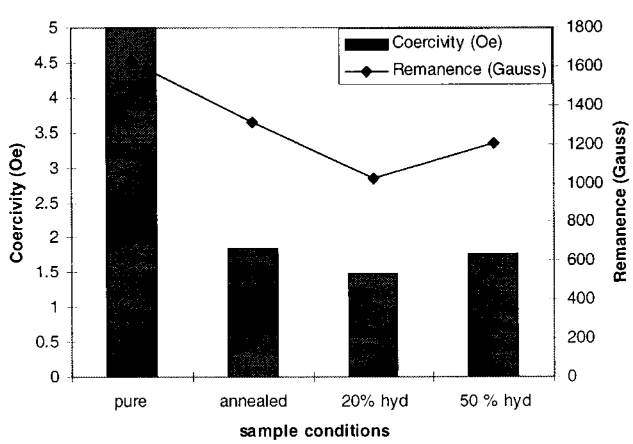

FIG. 2. Variation of coercivity and remanence of nickel as a result of thermal charging.

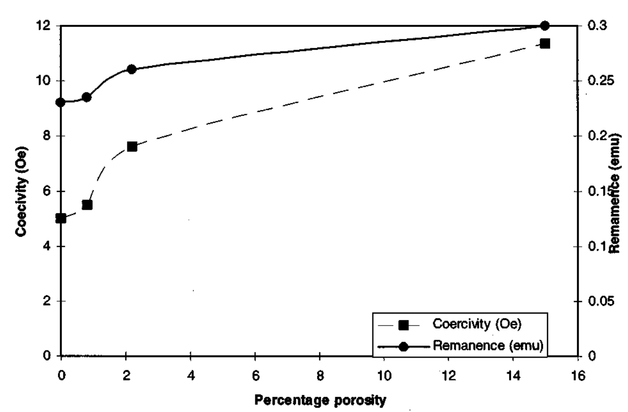

FIG. 3. Variation of coercivity and remanence with the density of nickel specimens with controlled amounts of porosity.

netic properties of pure nickel. For this purpose, pure nickel rods with different levels of porosity were prepared and their magnetic properties were studied. Compacts were made using nickel powder mainly composed of individual particles that were typically a few microns in diameter. The compacts were cold isostatically pressed by applying a pressure of 4400 atm and sintered under different conditions to obtain different densities. The four compacts thus obtained were tested for their magnetic properties at room temperature. From the data plotted in Fig. 3, it can be seen that there was a regular decrease in some of the magnetic properties such as the coercivity and remanence, as the density increased. This is because the number of pores decreases as the density increases and these act as the pinning sites for the motion of the domain walls. As a result, the irreversible nature of the magnetization was enhanced and therefore the coercivity, remanence, and hysteresis loss increased at low densities.

V. CONCLUSIONS

Hydrogen absorption in nickel under extreme conditions of high temperature and pressure had a small effect on the bulk magnetic properties, since the hydrogen does not form new pinning sites, but is adsorbed on those that already exist. The largest measured change in coercivity was from 2.0 to 1.5 Oe. Cathodic charging of nickel or iron resulted only in changes to the surface from the formation of hydrides or voids and this did not greatly affect the bulk magnetic properties. In this case the largest change in coercivity was from 2.6 to 1.6 Oe. The presence of voids in the bulk of the material affected the magnetic properties and can be related directly to the density of the material. These results led to the conclusion that the detection of hydrogen by bulk magnetic measurements is problematic unless the amount of hydrogen is very high, the hydrides can be formed in the bulk material, or the resolution of the magnetic measurements is sufficiently high.

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