Compulsory Checking of Nuclear Power Engineering Materials by Direct and Eddy Current

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Abstract. The testing technology of copper parts designed for dry storage of spent nuclear fuel with application of direct and eddy current has been developed. Measurements results of flaw quantity caused hydrogenation and oxidation processes are presented. Evolution of copper M 001 flaw structure during hydrogenation from gaseous medium is analyzed. It has been demonstrated that the dependence of copper ρ electrical resistance on number of flaws in its structure has dome shaped character and changes with eddy current frequency change. Number of flaws formed by hydrogen depends on direction (100) or (200) of the crystal structure of copper lattice.

1. Introduction

Copper containers are widely used for dry storage of spent nuclear fuel [1]. Hydrogen plays a significant role during their production and operation as it dissolves in melting during welding metal. Hydrogen ingresses into metal from air which contains water vapour, from moisture covering electrodes, from oxides located on the surface of metal product and electrodes. Under high temperature moisture fumes and dissociates with heat absorption. Hydrogen is contained in electrode coatings and in metal. As a result of components production, gasing under operating conditions, especially during welding, when hydrogen inevitably precipitates, «hydrogen disease» occurs under high temperature in copper. For express diagnostics of welded joints in particular, various methods of nondestructive testing and their combinations are used [2].

During operation by hydrogenation complex structural changes are frequently observed in the surface layer and generally in metal. Saturated layers, characterized by the presence of hydrogen implementation phases and changes of the parameters of the crystal lattice at different depths of the metal, are formed. As a result, there are structural defects. Despite the diffusion effects, which are characteristic of titanium, hydride formation, reducing the mobility of hydrogen, there is hydrogen distribution in the depth of the titanium sheet. The analysis of copper is of great importance. In particular, in the manufacture of electro-vacuum devices, OFC with oxygen of not more than 0.0005% is used. When oxygen in copper is excessive the "hydrogen disease", caused by the presence of hydrogen in copper, occurs. The quality of copper is determined by measuring the number of bends of copper strips, pre -annealed in a hydrogen atmosphere. This process is not technological. Thus, the analysis proves the relevance of finding alternative non-destructive methods for determining the concentration of hydrogen in metals. Eddy current transducers are used for a variety of measurements of properties and for detail and material control. However, in this case there is a following problem. If

the controlled samples are sufficiently thin, the signal measured with an eddy-current transducer is dependent not only on the material properties of the sample, but also on its thickness. Due to the fact that the conductivity is a function of hydrogen concentration in metals, the eddy-current method can be used to measure the hydrogen content in metals. However, in all cases, the topical problem is the accuracy and sensitivity of the measurements.

In the present paper the evolution of copper M 001 flaw structure during oxygen and hydrogen saturation is analyzed. Principal physics for application of current methods for these purposes is change in ρ copper electrical resistance dependant on the presence of gases and flaws in copper. Defects of crystal lattice influence specific resistance of metals. Change in electrical resistance in different frequencies of eddy currents leads to deformation of obtained dependence.

The goal of the work is to develop the technology for diagnostics of copper containers with spent nuclear fuel using the method of current analysis.

2. Materials and Methods. Experimental Procedure

Copper M 001 having the following composition [%wt.]: 0.01 Fe; 0.1 Si; 0.07 C; 0.024 O; 0.04 N was used for studies. Hydrogenation was performed according to Sievert's method [3] under the temperature up to 1000 ^{0}C and oxidation equal to 600 ^{0}C. Measurements of hydrogen concentration were performed using hydrogen analyser RHEN602 by LECO. Density of dislocation in titanium was determined by broadening of X-ray lines using diffractometer Shimadzu XRD-6000 in radiation Cu-Kα. Analysis of phase composition was performed using PCPDFWIN and PDF-4+ data base and software for full profile analysis POWDER CELL 2.5. Hydrogen depth distribution was performed by means of magnetic spectral analyser (3MA, Germany). Resistance measurement of titanium samples in direct current was performed by 4 probe method [4] using hard- and software by «KEIHLEY INSTRUMENTS».

3. Results

Hydrogen interacts with the present structural defects as well as it induces new defects formation and dislocations appearance [5, 6]. This leads to changes in electrical resistance. Change in specific resistance of dislocations, related to the unit of their density N_d in the unit of volume in dependence to hydrogen concentration in metal was estimated using the formula [7]:

$$D = \frac{\rho_d}{N_d} = \frac{\hbar k_F \Omega_a Q}{n_s e^2}, \quad (1)$$

where k_F – volume of wave vector on Fermi level, Ω_a – atomic volume, Q – transport cross-section of electrons scattering, n_s — number of current carriers per atom, e – volume of electron charge, n_s – number of charge carriers.

Dislocations density N_d was determined using the formula [8] based on X-ray diffraction analysis data

$$N_d = \pi\beta^2 ctg^2\Theta / 16b^2, \quad (2)$$

where β – broadening of X-ray lines conditioned by lattice microdeformation, θ – the angle which corresponds to the maximum of X-ray line, b – Burgers vector. For each weight concentration of hydrogen in copper, specific resistance of samples was measured using probe method [4].

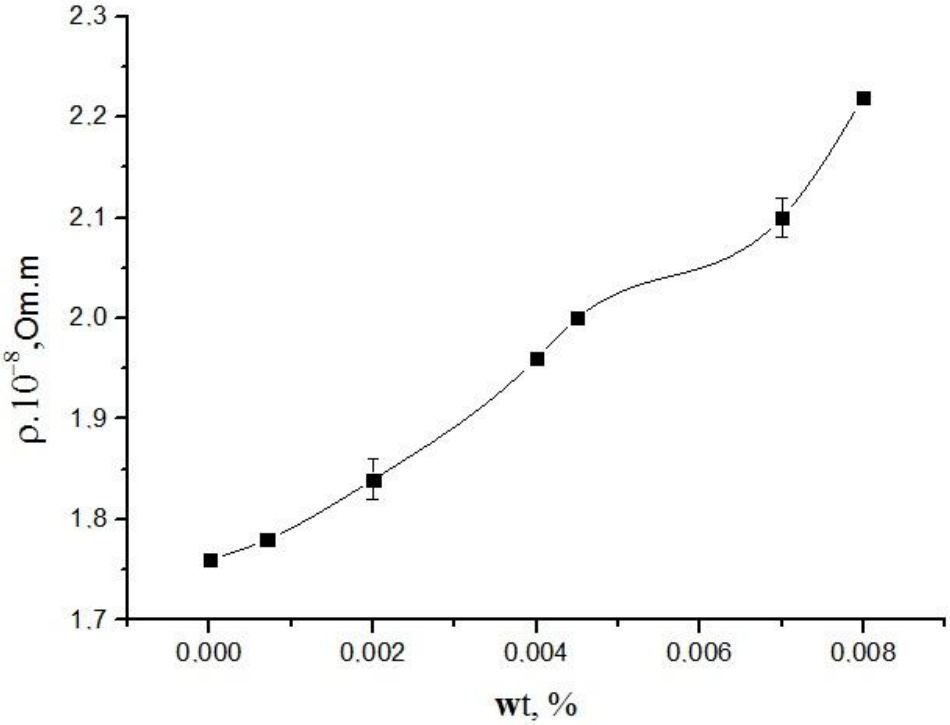

Figure 1. Dependence of Specific Electrical Resistance on Hydrogen Concentration in Copper

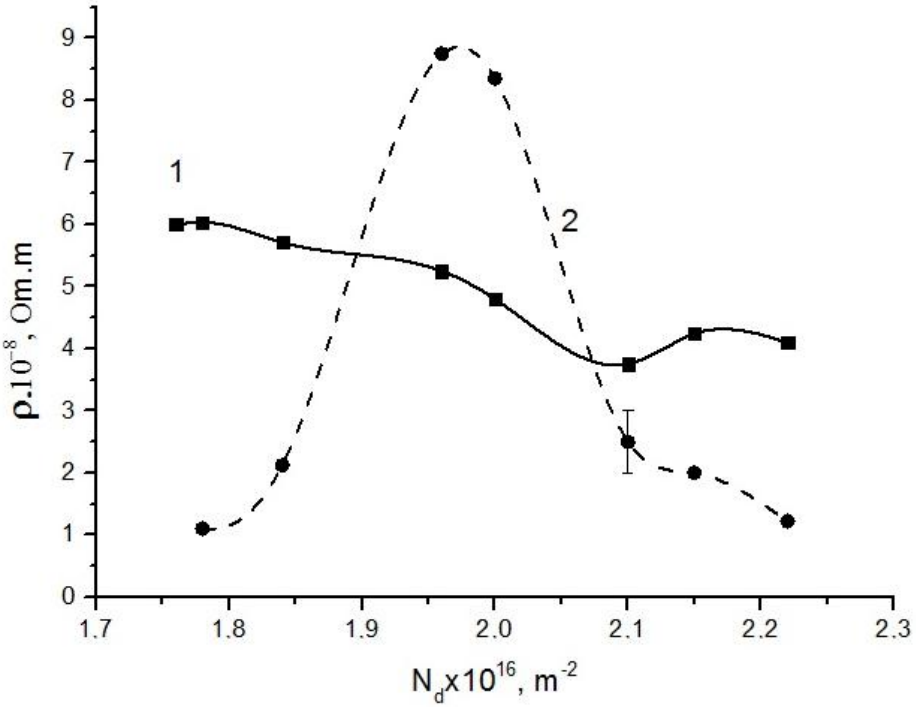

Figure 2. Dependence of Specific Electrical Resistance on Defects Concentration in Hydrogenated Copper (1- direction (100), 2 – (200))

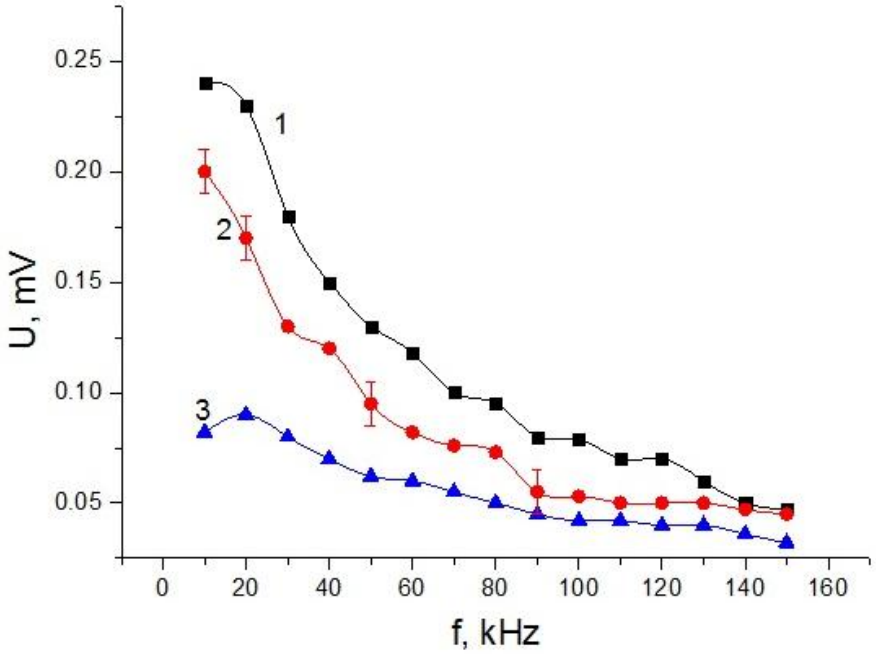

Figure 3. Dependence of Magnetic Spectrometer Indications on Eddy Current Frequency, 1 – Hydrogenated and then Oxygenated Copper, 2 – Sample Hydrogenated up to Oxidation, 3 – Initial Sample

The change of parameter P can be caused by different area of a sample covered by eddy current at different depth of its penetration. The hydrogenation can change both electric conductivity and the area of a layer located at depth δ. Since the experimental separate determination of the area value is impossible, the relative change of parameter P during hydrogenation allows evaluation of metal structure change extent, which enables the analysis of hydrogen content at different depth of a material. This is demonstrated by a plot in fig 4. The change of parameter P at frequencies varying from 50 to 100 kHz testifies not the change of thickness, but a prevailing change of metal structure caused by the hydrogenation.

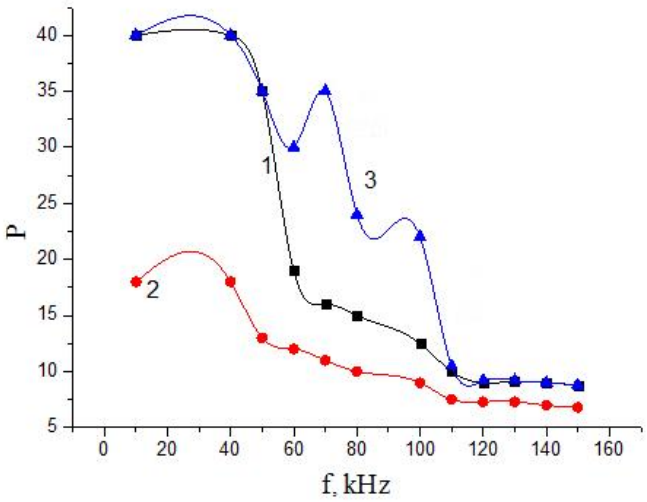

Figure 4. Dependence of parameter P on frequency of 3MA magnetic spectrometer eddy current (1 – Δd/d=0.05/1.7); 2 – Δd/d=0.05/1.75; 3 is hydrogenated metal)

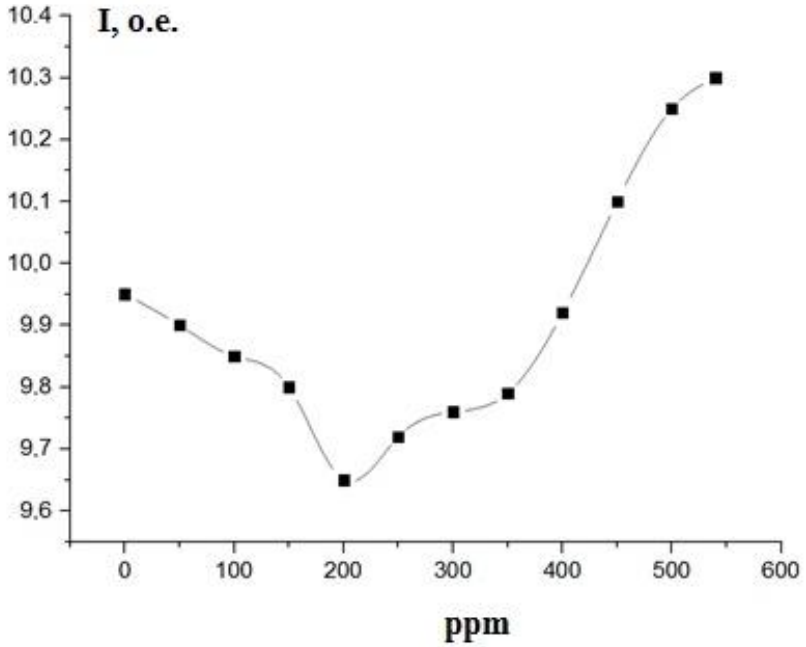

Figure 5. Dependence of active component of eddy current (frequency of 200 kHz) on hydrogen content in VT1-0 titanium

The dependence of eddy current transducer signal on hydrogen concentration in VT1-0 titanium for different frequencies is depicted in fig. 5. The dependence can be specifically used as calibration curves for the analysis of hydrogen content in every particular case.

Thus, the study results allow creation of nomogram atlas for determination of the condition of exposed products [9]. Alternatively, application of nomograms provides an intelligent control in the conditions of personnel limited access to continuously exposed objects.

4. Discussion

Figures 1-3 represent the results of experimental studies of copper samples. In all cases the electrical resistivity and half-width of X-ray spectrum were measured. Within the studied hydrogen concentrations in copper the virtually linear dependence of electrical resistance increase on hydrogen concentration in copper is observed (Fig.1). Virtually decreasing dependence of electrical resistivity on number of defects calculated using the formula (1), is characteristic for the direction (100) of crystal lattice of copper (Fig.2). Equally the dependence of specific resistance of hydrogenated copper samples for direction (200) of crystal lattice possesses dome-shaped character. The difference between the curves 1 and 2 in Fig. 2 on directions of crystal lattice is probably related to change in the number of hydrogen atoms introduced to copper lattice and changes of dislocations character. Dependence of the parameter $D = \rho d/Nd$ (formula 1) on hydrogen concentration in copper in the first case carries decreasing character, in the second case this value virtually does not change. This indicates the decrease of total transport cross-section of electrons scattering on defects caused by hydrogen (formula 2).

Fig. 3 shows the results of copper samples measurements performed by eddy currents. The difference between the curves is conditioned by the following defects. Under regulated by standards the oxygen concentration in copper is up to 0.065 % on the mass, some of oxygen within the limits of solubility is located in copper crystal lattice. The other part in the form of oxide particles is stochastically located in copper matrix. Voids occurrence is possible in the proximity of copper oxides particles in oxygen-containing copper samples [10], due to hydrogen and oxygen interaction.

Under low frequencies the maximum difference for initial hydrogenated and oxidized samples is observed. In this case the penetration depth of eddy current in sample is maximum (Fig. 3). The difference is decreasing with increase of eddy current frequency. This is conditioned by the fact that electrical conductivity on the copper samples surface is for the main part performed by impurities rather than hydrogen and oxygen. It should be underlined that the behavior of the curves within the limits of ± 0.02 mV depends on the degree of initial samples preparation for studies.

5. Conclusion

The potential of using eddy current method for testing of nuclear power engineering materials is mainly provided by maintenance personnel safety. In research, certain interest is represented by the comparison of electrical resistance measurements performed under direct and eddy current along with X-ray diffraction analysis in order to identify the conditions of current propagation on defects of hydrogenated metals. Imperfection of hydrogenous copper structure is evident with the presence of electrical resistance dependences on directions 100 and 200 of copper crystal lattice and is identified by measurements of eddy current of different frequencies.

Acknowledgements

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