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Copper and phosphorus effect on residual embrittlement of irradiated model alloys and RPV steels after annealing

M. Valo[a], L. Debarberis[b,*], A. Kryukov[c], A. Chernobaeva[c]

[a] *VTT Industrial Systems, P.O. Box 17042, FIN-02044 VTT, Finland*
[b] *JRC-IE, CEC, P.O. Box 2, 1755 ZG Petten, The Netherlands*
[c] *RRC-KI, Kurchatov Institutes, Moscow, Russian Federation*

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Abstract

The dependence of the recovery of the transition temperature shift after annealing (475 °C, 100 h) on copper and phosphorus contents has been studied on irradiated reactor pressure vessel (RPV) materials. A set of model alloys with low nickel content, lower than 0.2 mass%, was used for the study. Copper and phosphorus contents were varied in a wide range: 0.005–0.99 and 0.002–0.039 mass%, respectively.

Recovery efficiency has been estimated by the value of residual embrittlement after annealing, measured in terms of a shift in transition temperature (ΔT_K^{res}).

A comparison of the results obtained on model alloys with data for VVER-440 RPV materials has also been carried out. Comparative analysis has confirmed the conclusion that ΔT_K^{res} is independent of phosphorus content while the effect of copper on ΔT_K^{res} is not significant for typical VVER-440 RPV materials with a typical range of Cu contents between 0.10 and 0.24 mass%. However, for model alloys with a wider range of copper content, copper mainly controls the value of ΔT_K^{res}.

Keywords: Annealing; Irradiation; Recovery; Transition temperature; Copper; Phosphorus

1. Introduction

Several reactor pressure vessels (RPVs) approaching the end of design life have been successfully annealed in recent years by means of thermal treatment at an optimized temperature–time regime (475 °C, 100 h) [1]. Thermal annealing is still considered as an option in several plant life management strategies, in particular for RPVs approaching end-of-life. It is generally assumed that annealing at a properly chosen temperature and time can recover completely matrix damage. However, the recovery of the damage due to precipitates and segregates is not fully understood [2]. It is therefore very important to quantify the annealing efficiency and its dependence on phosphorus and copper.

In order to understand the roles of phosphorus and copper in the changing RPV steel's structure and its behaviour under re-irradiation, it is necessary to analyse the evolution of the structure taking place during the processes of irradiation and annealing. This can be done qualitatively with atom probe studies and TEM analysis [3–5]. For typical annealing conditions of VVER-440 materials, it is observed that during annealing, copper precipitates do not massively re-dissolve back into the matrix but they grow larger, decreasing their density. In contrast, phosphorus almost completely re-dissolves back into the matrix. As it is mainly a high density of small defects that are very effective barriers for dislocation movements, it is expected that damage due to phosphorus can be completely recovered by thermal annealing. The damage contribution due to copper is expected to be recovered effectively well enough, for practical cases. An experimental study of annealing efficiency is presented in this paper.

*Corresponding author. Tel.: +31 22456 5413; fax: +31 22456 1432.
E-mail address: debarber@jrc.nl (L. Debarberis).

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2. Materials, irradiation and annealing parameters

A set of very well-characterized model alloys was chosen as the subject of the study [6]. A group of materials with low nickel content has been considered to evaluate phosphorus and copper effects on residual embrittlement after annealing of irradiated materials. The variation ranges for nickel, phosphorus and copper in these alloys are given in Table 1.

All the model alloys studied in this work were irradiated in the high flux reactor (Petten, The Netherlands) at a fluence of $3\times10^{18}\,\mathrm{cm}^{-2}$ ($E>1$ MeV) at 270 °C. After irradiation they were annealed at a temperature of 475 °C for 100 h.

Residual embrittlement was evaluated from the results of impact tests using mainly KLST small CV specimens ($4\times3\times27$ mm). The value of residual embrittlement, $\Delta T_K^{\mathrm{res}}$, was determined as the difference between absolute values of transition temperature after annealing, T_K^{ann}, and initial transition temperature, T_{K0}:

$$\Delta T_K^{\mathrm{res}} = T_K^{\mathrm{ann}} - T_{K0}. \tag{1}$$

Values of $\Delta T_K^{\mathrm{res}}$ as functions of phosphorus and copper contents are given in Figs. 1 and 2. Different symbols correspond to the experimental data for different copper contents; Fig. 1. It is apparent that an increase of phosphorus content from 0.002 to 0.039 mass% does not result in a noticeable increase of residual embrittlement if copper content is lower than 0.1 mass% ($\Delta T_K^{\mathrm{res}} \leqslant 5$°C).

Table 1
Variation ranges of Ni, P and Cu in the model alloys

	Ni, mass%	P, mass%	Cu, mass%
Min.	0.01	0.002	0.005
Max.	0.20	0.039	0.99

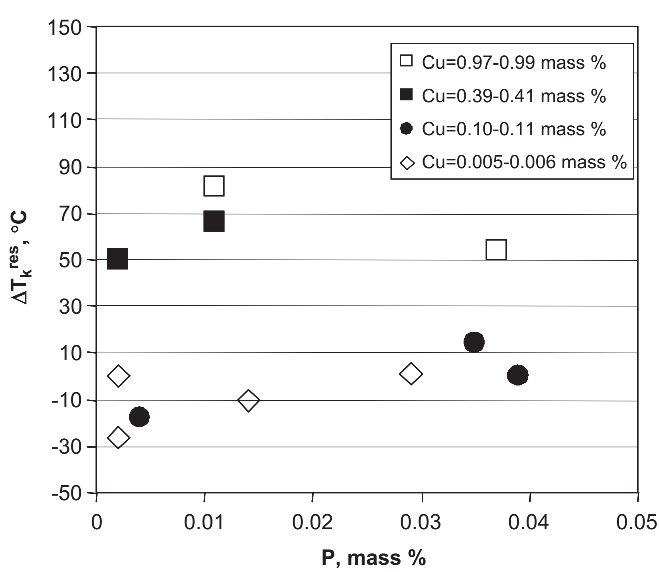

Fig. 1. $\Delta T_K^{\mathrm{res}}$ for model alloys versus phosphorus content.

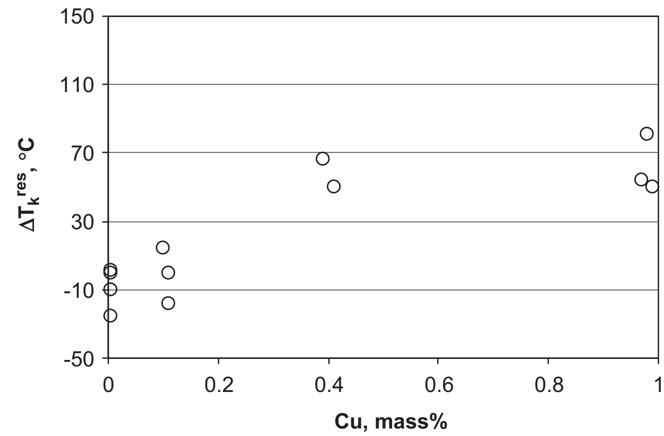

Fig. 2. $\Delta T_K^{\mathrm{res}}$ for model alloys versus copper content.

Table 2
Estimates of the parameters in Eq. (2) and their statistical characteristics for model alloys

Parameter estimated	Coefficient	Std. error
C_1	462.52	406.58
C_2	74.47	14.79
C_3	−11.89	10.66

However, higher values of $\Delta T_K^{\mathrm{res}}$ (50–80 °C) are typical for alloys with high copper content (0.40–0.99 mass%). An increase of copper content from 0.005 up to 0.99 mass% results in a $\Delta T_K^{\mathrm{res}}$ increase from 0 to 80 °C as shown in Fig. 2.

To estimate quantitatively the effects of phosphorus and copper on $\Delta T_K^{\mathrm{res}}$, a parametric statistical evaluation based on Eq. (2) using three parameters, C_1, C_2 and C_3, was performed.

$$\Delta T_K^{\mathrm{res}} = C_1 \times \mathrm{P} + C_2 \times \mathrm{Cu} + C_3, \tag{2}$$

where P and Cu are the phosphorus and copper bulk contents in mass%.

The values of the parameters and their statistical characteristics are presented in Table 2.

The analysis of the data given in Table 2 shows that only the parameter C_2 is significant (95% confidence). This means that for the ranges of phosphorus and copper contents considered there is a linear dependence between $\Delta T_K^{\mathrm{res}}$ and copper and no dependence between $\Delta T_K^{\mathrm{res}}$ and phosphorus content. Similar results were obtained in [6,7].

The experimental results obtained in this work were compared with available data for VVER-440 RPV steels [8–11]. The data were obtained at a nominal operating temperature of approximately 270 °C and at different fluences covering the same range of the studied model alloys and annealed at the same already mentioned optimised annealing regime.

The ranges of phosphorus and copper, typical for VVER-440 RPV steels, are significantly smaller for copper