L. ŘEHÁČKOVÁ, J. DOBROVSKÁ, J. KALOUSEK

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CHARACTERIZATION OF CHEMICAL HETEROGENEITY OF THE WELDED JOINT OF TWO STEELS BY MODELLING OF DIFFUSION PROCESSES

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Preliminary Note –Prethodno priopćenje

The paper deals with original methodology for characterization of chemical heterogeneity of welded joint by measurement of concentration profiles of substitution elements in three straight lines in a plane perpendicular to the weld interface. Three concentration profiles were evaluated by modelling of diffusion based on solution of the 2nd Fick's law. Primary goal of evaluation consists in characterization of chemical heterogeneity in the narrow area on both sides of the welded joint on the basis of their mutual position. Experimental data are optimised by original adaptation of the Levenberg-Marquardt's algorithm of non-linear regression. The paper deals with diffusion of nickel in the welded joint of two different steels as an example of investigated elements.

Key words: diffusion, distribution of substitution elements, chemical heterogeneity, weld, steel

INTRODUCTION

Welded joints of steel with increased contents of alloying elements are almost always accompanied by higher chemical and structural heterogeneity that is particularly characteristic for the transition zone of the welded joint. The accompanying redistribution of interstitial carbon strengthens the tendency of heterogeneity. Chemical and structural alterations in the transition zone caused by a redistribution of elements can also considerably determine strength and other mechanical properties of contacting metals. In technical practice adverse effects of chemical and structural heterogeneity manifest themselves in welded joints of heat-resistant steels that are subjected to a load which is both of high-temperature and long-duration. The margin of safety – or vice versa danger – certainly depends on time and temperature of annealing.

Monographs [1] and [2] have introduced various models of diffusion solution in welded joints with application on interstitial elements, particularly carbon. Works [3 - 5] have applied these models on ternary and polycomponent systems of austenitic steels.

The pilot study of the diffusion processes of substitution elements at the interface of two different steels from the point of chemical heterogeneity has been given in [6 - 8].

The term "chemical heterogeneity" means the difference in chemical composition in different places of the microstructure of an analysed system. In metallic materials, e.g. in steels, the chemical heterogeneity is mainly caused by the segregation behaviour of elements in the course of material solidification. As both constitutive elements and impurities segregate into interdendritic areas during steel solidification, the difference in chemical composition in dendrites and in interdendritic areas can be relatively high in cast steels.

The aim of the present work is primarily to show a possibility of an assessment of chemical heterogeneity of the weld joint on the basis of a difference in concentration profiles calculated from data sets measured along three lines perpendicular to the welded interface. This problem is illustrated on Ni redistribution in the welded joint of two different steels.

EXPERIMENT DESCRIPTION

A heat-resistant low-alloy CrNiMoV steel (P2) and low-alloy silicon steel (V) were used for the welded joint. Both steels are of ferrite structure. Table 1 gives the chemical composition of the steels.

Table 1 **Chemical composition of steels of welded joint / wt.%**

Steel	C	Mn	Si	P	Cr	Ni	Cu
P2	0,16	0,48	0,28	0,009	2,20	1,29	0,06
V	0,80	0,66	2,34	0,026	0,09	0,05	0,09

Steel	S	Mo	V	Al	Fe
P2	0,010	0,59	0,100	0,01	rest
V	0,033	0,004	0,007	0,15	rest

Samples in the form of small flat cylinders with a diameter of 12 mm and a height of 4 mm were prepared from both steels. A polished metallographic section was prepared on the frontal surface of each cylinder. Samples were welded on polished surfaces in diffusion pairs by electric shock under a protective argon stream.

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Welded diffusion pairs were afterwards sealed into quartz tubes with titanium splinters and isothermally annealed in an electric resistance furnace in the temperature range from 500 to 1 000 °C with a grading of 50 °C with a different time of annealing [6]. Twelve samples were prepared altogether. After annealing, the welded diffusion pairs were removed from the quartz tubes, cut in half perpendicularly to the welded interface and polished; metallographic sections were prepared on the cut surfaces.

Distribution of the selected elements was investigated by energy dispersion X-ray micro-analysis with use of the instrument JEOL – JXA 8600/KEVEX. This analysis was made in selected points, along three straight lines, perpendicularly to the weld interface, approx. 180 μm long with step of 3 μm. Concentration sets containing 61 values of concentrations of the measured elements (Al, Si, Ti, Cr, Mn, Fe, Ni, Mo) were obtained from one measured segment. Three such sets were obtained from each sample.

THEORETICAL BASIS

Solution of the 2nd Fick's law, used for this special case, results in the following relation [3 - 5]:

$$N(x,t) = N_1 + 0{,}5(N_2 - N_1)\,erfc\left(\frac{x - x_0}{2\sqrt{Dt}}\right) \qquad (1)$$

Here $N\,(x,t)$ corresponds to the concentrations at the distance x from the welded joint in the time t. Symbols N_1, N_2 correspond to initial concentrations in both areas of the welded joint, diffusivity D is equal for both sides of the weld for the element in question, x_0 is a correction of the coordinate eliminating a possible inaccuracy of measurement of the interface between metals of the welded joint. Eq. (1) was used for constant temperature. Eq. (1) was derived from the fundamental solution, given e.g. [9].

EVALUATION OF EXPERIMENTAL DATA

The objective of the data evaluation consists of the calculation of concentration profiles. The tool for achieving this objective is the original calculation optimization program, which determines free optimization parameters of the diffusion Eq. (1) i.e. D, x_0, N_1, N_2. Calculation of concentration profiles and free parameters of Eq. (1) is based on non-linear regression. A proprietary original adaptation of Levenberg-Marquardt' algorithm [10] was developed. Short description of the mentioned program was published [11].

Three sets of concentrations measured along three lines enable variant access of optimization processing:

A) The complex of all concentration data measured along all three lines was used for input into computation program (i.e. 183 concentration values).

B) Only the mean concentrations were used for input into computation program (i.e. 61 concentration values). Each such mean concentration was calculated from three concentrations measured along three lines at the same distance from the weld interface.

C) Each set of measured concentration data was used separately for input into computation program (i.e. 3 × 61 concentration values), (see Figure 1).

D) From the output of method C) an arithmetic mean of parameters N_1, N_2, D and of their errors were determined.

E) Only the medians of measured concentrations were used for input into a computation program (i.e. 61 concentration values). Each such median was calculated from three concentrations measured along three lines at the same distance from the weld interface. This procedure is a certain form of robust regression; that is, it does not exclude boundary points, but gives lesser weight to them.

Table 2 presents the results of evaluation of diffusion coefficients according to the above mentioned methods. With regard to great errors of these coefficients the calculated values were rounded off. Due to the limited scope of the paper only results of one selected sample are presented. This selected sample most markedly encompasses solved problems, i.e. the influence of chemical heterogeneity on diffusion characteristics (particularly diffusivity) of an analysed element in a welded joint.

DISCUSSION

A great many calculations of parameters for the elements Cr [6, 14], Si [7], Mo and Ni were made by the

Table 2 **Comparison of diffusivities of sample (700 °C, 40 h) according to different methods**

method	$10^{15}D_{Ni}$ /cm^2 s^{-1}	Coefficient of determination
A	900 ± 300	0,905
B	900 ± 400	0,957
C - line 1	200 ± 200	0,913
C - line 2	500 ± 400	0,923
C - line 3	3500 ± 1700	0,907
D	1400 ± 300	0,914
E	1000 ± 400	0,961

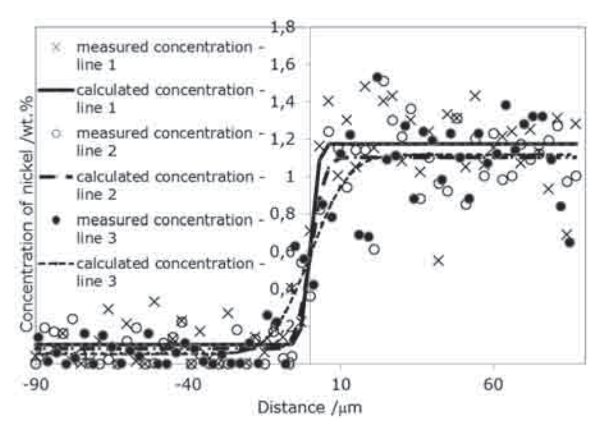

Figure 1 Redistribution of nickel in 1st, 2nd and 3rd measured line, sample (700 °C, 40h)

authors by the methodological approach described above (Figure 1). They were successful even in case of large scatter of experimental data. Less favourable side of large scatters are the magnitude of parameters' errors.

The model used generally assumes diffusion in a homogenous environment. This condition need not always be sufficiently fulfilled. Various rates of diffusion in the mass of crystal and along grain boundaries can occur [12], forming of intermetallic compounds [13, 14], segregation in poly-component system, influencing of diffusion by phase transformations, by changing activities of diffusing components and by other situations of thermodynamic non-equilibrium. These phenomena may not be of random character and also may not have Gaussian distribution. Until they are not sufficiently quantified and implemented by the system of physical-chemical laws into a calculation program, the statistic assessment of scatters remains an indirect quantitative indicator possible secondary processes in the area of welded joint.

The submitted model is based on use of the 2nd Fick's law individually for each element, it neglects the influence of interdiffusion coefficients, considered particularly in ternary systems, see e.g. [15]. Systematic error of simplified method is also reflected in errors of parameters. The coefficient of determination belongs to the statistical quantities appraising the fitting of the optimised function to the measured points. It is advisable to pay enhanced attention to phenomena with irregular standard deviations of calculated parameters.

In spite of unfavourable scatter of data in the case of nickel the developed methodology brings results. The obtained results show that diffusion characteristics of a system can significantly differ depending on the selected locations for concentration data measuring. Thus, they can differ depending on the chemical heterogeneity of the system. The chemical heterogeneity is the reason for different concentration profiles calculated in three investigated areas. The scatter of the measured concentrations (i.e. chemical heterogeneity) results in the difference in optimization parameters of the diffusion equation.

Although the estimation of diffusivities is not the main aim of this work, the results of various methods (see the previous chapter) show relatively great differences of quantities, included in Eq. (1), in case of the diffusivities see Table 2. The reason is very high chemical and structural heterogeneity of the materials, forming the welded joint. Thus, if we measure the concentration profiles perpendicular to welded joint in different places, we get a somewhat different concentration profiles of the elements. Influenced diffusion layer is in close proximity to the welded joint. Diffusion characteristics calculation accuracy is influenced by the number of points measured in this area. For this reason, appear appropriate method A) (or B)), since it considers all points measured in the diffusion-affected zone.

Because all the methods were performed by an identical computation program, the results of all used methods are mutually comparable. Seen from this aspect the different results from the different authors and laboratories are fully apprehensible. The value of the diffusion coefficient of nickel in the pure α-Fe at 650 °C is 2.10^{-14} cm^2/s [16]. Obviously the local conditions determine the parameters of the diffusion equation. The rates of diffusion differing in the three levels of welds' depth may accelerate its unfavourable behaviour as far as the stability is concerned.

From the engineering viewpoint it would be useful to conduct a similar evaluation for all analysed elements, together with metallographic analysis and thermodynamic calculations of possible chemical processes, for complex comparison. Regrettably, such extensive appraisal is outside the frame of this paper.

The authors are fully aware of the fact that the presented model is simplified: it assumes only homogenous molecular diffusion in one direction. This limitation does not take into account the above mentioned secondary processes, which may run parallel or consequent with the basic process. In spite of these constraints the results obtained by the developed methodology are original and useful.

Knowledge and evaluation of heterogeneity of the welded joint is important in the first place for technical practice. Generally, the result and methodology can serve also as an incentive for the next theoretical and experimental investigation of this topic.

CONCLUSION

The paper presents original results of methodology for experimental concentration data evaluation and for expression of chemical heterogeneity of a welded joint on the basis of diffusion characteristics.

Adaptation of the Levenberg-Marquardt's algorithm for the solution of optimisation of parameters of a diffusion equation is also original. On the basis of application of this algorithm the different values of optimisation parameters of diffusion equation were determined for the concentration data sets measured in the individual areas. The different values of optimisation parameters (i.e. diffusion characteristics), determined for the individual concentration profiles, indicate the chemical heterogeneity of material.

Thus, in this work, diffusion is understood among others as the tool for expression of chemical heterogeneity of the system. This approach in diffusion as the tool for expression of chemical heterogeneity is original and new.

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Note: The responsible translator for English language is the lecturer from VŠB-TU Ostrava, Czech Republic