CONTRIBUTION TO MATHEMATICAL MODELLING OF MANGANESE DIFFUSION IN A WELDED JOINT OF TWO IRON-BASED MATERIALS

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PŘÍSPĚVEK K MATEMATICKÉMU MODELOVÁNÍ DIFÚZE MANGANU VE SVAROVÉM SPOJI DVOU MATERIÁLŮ NA BÁZI ŽELEZA

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Abstrakt

Tato práce vychází z programu výzkumných prací zabývajících se studiem nových materiálů. Součástí tohoto studia jsou pokusy o využití numerického modelu tuhnutí předlitků. Existují určité rozdíly mezi experimentálními výsledky a výsledky modelu. Ke zpřesnění těchto výsledků by mohly přispět informace o intervalu krystalizace. Předpokládá se, že při rozmezích chemického složení vzorků, které byly k dispozici, může mít na teplotu krystalizace největší vliv obsah uhlíku.

Z hlediska etapy projektu, která se věnuje transportním dějům v materiálech se předpokládá provedení experimentu tak, že jako „protikus“ ke každému vzorku se vybere vzorek čistého materiálu a obě části budou elektrošokově svařeny. Po tepelném zpracování se uvažuje vliv uhlíku a manganu na gradient chemického složení v obou částech vzorku. Předkládaná práce se zabývá difúzí manganu.

Článek vychází z již dříve vyvinuté a aplikované původní metodiky adaptace Levenberg – Marquardtova algoritmu pro model redistribuce substitučních prvků ve svarovém spoji dvou materiálů na bázi železa. Postup je numericky i graficky demonstrován na manganu.

Teoretickým základem vyhodnocení je řešení 2. Fickova zákona jednorozměrové difúze s příslušnými okrajovými podmínkami. Okrajové podmínky předpokládají stejné hodnoty difuzivit prvku v obou materiálech. Zároveň předpokládají, že ve velké vzdálenosti od roviny svarového spoje jsou koncentrace totožné s výchozí chemickou analýzou. Parametry difúzní rovnice se optimalizovaly metodou nelineární regrese.

Výstup programu zahrnuje statistické výsledky, vyjadřující míru použitelnosti modelu a strukturní i chemickou heterogenitu. Výsledné hodnoty optimalizačních parametrů závisejí na metodě zpracování.

Abstract

The work is based on the program of research works dealing with investigation of new materials. This investigation comprises attempts of use of numerical model of son of cast blanks. There are some differences between experimental results and results obtained from the model. Information about crystallization interval could contribute to higher precision of these results. It is assumed that within the interval of chemical composition of the available samples it is specifically the carbon contents that could have had the biggest influence of crystallization temperature.

From the viewpoint of the given stage of the project, which deals with transport phenomena in materials it is planned to make an experiment in the following manner: each

sample will get its “counterpart” made form pure material and both parts will be welded by electric shock. After heat treatment an influence of carbon and manganese on chemical composition gradient will be studied in both parts of the sample. The presented work deals with manganese diffusion.

The paper is based on already previously developed and applied methodology of adaptation of Levenberg–Marquardt algorithm for the model of re-distribution of substitution elements in a welded joint of two iron-based materials. The procedure is illustrated numerically and graphically on the example of manganese.

Theoretical basis for evaluation is solution of the Fick’s second law of one-dimensional diffusion with the relevant boundary conditions. The boundary conditions assume the same values of the element diffusivities in both materials. They also presume that at great distance from the plane of the welded joint the concentrations are identical with initial chemical analysis. Parameters of diffusion equation were optimized by method of non-linear regression.

Program outputs comprise statistical results expressing the extent of usability of the model and structural and chemical heterogeneity. Final values of optimization parameters depend on the method of processing.

Keywords: welded joint, diffusion, manganese re-distribution

1. Introduction

Knowledge of re-distribution of elements in welded joints made of various iron-based materials have both theoretical and practical significance particularly for piping subjected to long-term thermal loads, e.g. in power engineering.

These issues have been for a long time already in the centre of interest of the works oriented on interstitial and substitution elements [1-5].

Presented methodology leading to obtaining of the relevant results consists of acquisition of the necessary experimental data, theoretical solution of the problem, its evaluation and interpretation of results.

Experience gained with use of this methodology is demonstrated in the presented work on manganese re-distribution.

2. Experiment

Measurement of the manganese re-distribution was realized on two welded joints, which were formed by electrolytic iron of the purity 3N and by Mn steels with chemical composition specified in the Table 1.

Table 1 Chemical composition of steels [wt %]

Steel	C	Mn	Si	P	S	Cr	Ti	Cu	Mo	V	Al	Co	Nb
Sample 25	0.39	0.97	0.29	0.011	0.006	0.05	0.01	0.13	0.01	0.01	0.01	0.01	-
Sample 31	0.08	1.29	0.23	0.011	0.005	0.03	0.02	0.11	0.01	0.07	0.03	0.01	0.04

Initial material – electrolytic iron of the purity 3N was re-melted twice in a plasma furnace in order to increase the purity to 4N. Afterwards it was melted in a vacuum furnace and cast into a graphite mould with three cylindrical holes with diameter 10 mm and depth 100 mm under presence of argon.

Samples of pure iron and of Mn steels were cut in transversal direction; frontal faces were polished and prepared metallographically to high cleanness. Welded joints were prepared at the Institute of Physics of Materials at the Academy of Sciences of the Czech Republic in Brno (ÚFM AV ČR Brno) by electric shock welding under protective argon atmosphere, while metallographic polished sections of the samples were mutually pushed together by mechanical pressure. Dimensions of each cylindrical sample were the following: diameter 10 mm, height 10 to 15 mm.

Welded diffusion pairs were subjected to isothermal annealing in electric resistance furnace at the temperature of 900 °C for 48 hours. The samples were after annealing cut in the middle perpendicularly to the weld boundary and metallographic polished sections were prepared on the surface of cut areas. Interfacial boundary was determined on microscope and a mark was made here by micro-hardometer. Distribution of selected elements over the welded interface was studied by the WDX method (wavelength dispersive X-ray analysis) – perpendicularly to the interface. This analysis was made in individual points with a step of 5 μm (altogether 26 measurements, the 13th measurement point was exactly at the interface, see the Fig. 1).

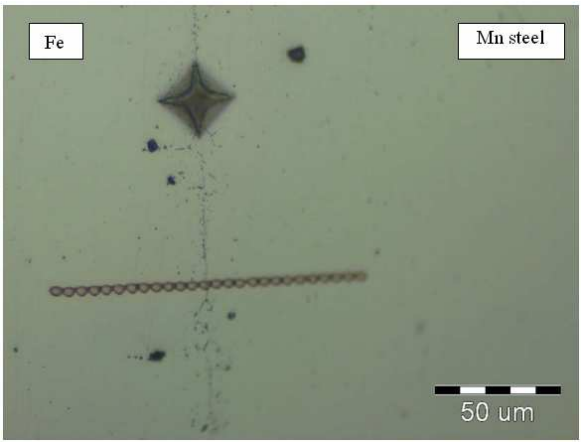

Fig.1 Structure of the welded joint (sample 31, 900°C / 48 h)

3. Theoretical basis

3.1 Diffusion

Theoretical basis for evaluation is solution of the Fick's second law of one-dimensional diffusion with the relevant boundary conditions. The boundary conditions assume the same values of the element diffusivities in both materials. They also presume that at great distance from the plane of the welded joint the concentrations are identical with initial chemical analysis. Parameters of diffusion equation were optimized by method of non-linear regression.

Solution of diffusion in the system that is formed by two media is based the concept of one-dimensional diffusion for semi-infinite areas: $(0\langle x\langle\infty)$, $(-\infty\langle x\langle 0)$. The basis equation can be found in the Ref. [6], or with respect to analogy between diffusion and heat conduction also in Ref. [7].

$$c_1 = A_1 + B_1 erf\,\frac{x}{2\sqrt{D_1 t}}, \quad x > 0 \tag{1}$$

$$c_2 = A_2 + B_2 erf \frac{|x|}{2\sqrt{D_2 t}}, \quad x < 0 \tag{2}$$

where c_1 is concentration in the area $x > 0$, c_2 is concentration in the area $x < 0$, A_1, B_1, A_2, B_2 are constants, which are obtained after insertion of initial and boundary conditions, D_1 is diffusion coefficient in the area $x > 0$, D_2 is diffusion coefficient in the area $x < 0$.

Diffusion of elements in the welded joint is solved by the equations (1), (2) with the following initial and boundary conditions:

Initial conditions:

1.) $c_1 = c_0$, $t \leq 0$, $0 < x < \infty$ (3)

2.) $c_2 = d_0$, $t \leq 0$, $0 > x > -\infty$ (4)

Initial conditions assume constant concentrations of diffusing component, different in both diffusion domains (diffusion areas).

Boundary conditions:

3.) $\frac{c_2}{c_1} = 1$, $x = 0$, $t > 0$ (5)

4.) $D_1 \frac{\partial c_1}{\partial x} = D_2 \frac{\partial c_2}{\partial x}$, x = 0, $t > 0$ (6)

Boundary conditions (5), (6) are valid in the time $t > 0$ on the boundary of both diffusion domains at $x = 0$. The boundary condition (5) assumes equality of concentrations on the boundary, the boundary condition (6) assumes equality of densities of diffusion flows on the boundary. It is assumed that diffusivity in both areas is independent on concentration. If the equality $D_1 = D_2$ is true, the concentration profile at the time $t > 0$ at $x = 0$ is continuous. The next calculation comprises this assumption.

Solution leads to the result that is valid for both parts of the welded joint - in the following form:

$$N_i(x,t) = N_1 + 0{,}5\left(N_2 - N_1\right) erfc\left(\frac{x}{2\sqrt{D_i t}}\right). \tag{7}$$

This form is cited in the work [2], where the values N, N_1, N_2 substitute generally expressed concentration by weight percents: $N(x,t) \equiv c_1 \equiv c_2$, $N_1 \equiv c_0$, $N_2 \equiv d_0$. The equation (7) satisfies the initial and boundary conditions. It can be easily proven that at big distance on both sides of the welded joint the following is valid: $N(\infty,t) = N_1$, $N(-\infty,t) = N_2$, i.e. that these concentrations are therefore identical with initial concentrations at the time $t = 0$.

3.2 Evaluation of experimental data

A modified form of the equation (7) from the work [9] was used for evaluation of experimental data:

$$N_i(x,t) = N_1 + 0{,}5\left(N_2 - N_1\right) erfc\left(\frac{x - x_0}{2\sqrt{D_i t}}\right) \tag{8}$$

From the measured values $N_i(x,t)$ and x, t the free optimization parameters N_1, N_2, D, x_0 were evaluated by method of non-linear regression. The parameter x_0 represents a correction of inaccuracy of deduction of (zero) position of the welded joint; from mathematical viewpoint it is a transformation of the coordinate x.

The attempt of evaluation of parameters from the equation (7) by the user program Polymath 5.1 [10] was unsuccessful. That's why there was developed a proprietary optimization program of non-linear regression. The Levenberg–Marquardt's method, which uses an algorithm according to the Ref. [11], is the core of the program. The program was at first tested on simulated data with small scatter. When real experimental data with greater scatter were used, it was found that the pursued special function minimizing sum of squares of deviations can have several local minimums. That's why the basic algorithm was applied only on limited part of multi-dimensional space (from the viewpoint of optimization parameters). The limitation did not permit e.g. negative values of the parameters N_1, N_2, D and too big deviations of the pars N_1, N_2 from the relevant chemical composition. It chose from the found local minimums the most suitable one, i.e. the one with the minimum residual sum of squares. In case of close magnitudes of these values it preferred a minimum with the smallest error of the value of diffusivity.

Scatters of real data cannot be attributed to a systematic error of the experiment, but rather to processes embedded already in steelmaking technology, which result in structural and chemical non-homogeneity in the area of the welded joint.

4. Results

Measurement results and their evaluation are contained in the Table 2 and in the Figures 2. and 3.

Table 2 Concentration of manganese and its diffusion coefficients

Sample No. (T[K], t[s])	$10^2 N_1$ [wt %]	N_2 [wt %]	$10^{12} D_{Mn}$ [cm^2/s]
sample 25 (900°C, 48h)	96.2 ± 1.2	$2.5\times10^{-6} \pm 1.4\times10^{-2}$	4.3 ± 0.6
sample 31 (900°C, 48h)	147.3 ± 1.7	$5.5\times10^{-8} \pm 1.7\times10^{-2}$	6.8 ± 0.7

Figure 2 corresponds to the manganese re-distribution in the sample 25, Fig. 3 corresponds to the same situation in the sample 31.

Computing program has made in both cases a correction of inaccuracy of determination of zero position of the interface of both materials. Shift in the Fig. 3 is quite considerable and changes due to oxidation at the zone of contact of both materials cannot be excluded. Number of concentration data in the areas corresponding to initial conditions according to the equations (3) and (4) is not big. This may be the cause of errors in the values N_1, N_2. These parameters and their statistics are determined by computing program. Diffusivity errors are permissible.

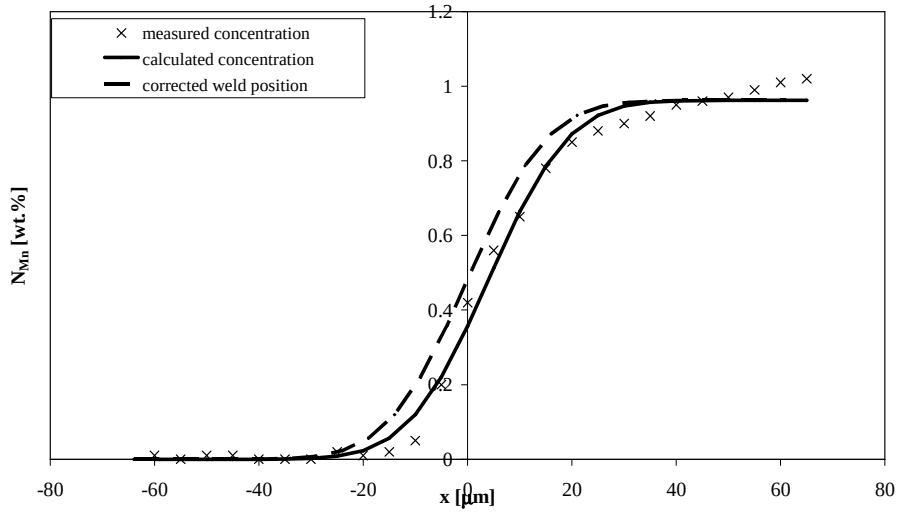

Fig.2 Manganese re-distribution (sample 25, 900°C, 48h)

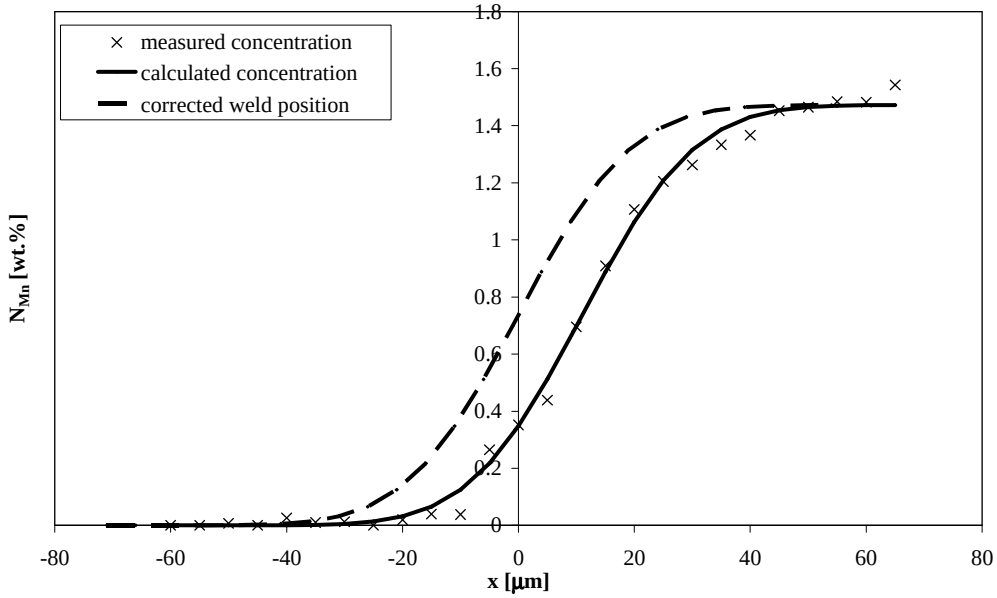

Fig.3 Manganese re-distribution (sample 31, 900°C, 48h)

5. Conclusion

Results of the manganese diffusion that were obtained by original method based on adaptation of the Levenberg–Marquardt algorithm can be considered as the first orientational information, since diffusivity was measured only at one temperature and in one time interval. Both these quantities must be put into their context and further developed. The values of the manganese diffusivity are acceptable, possible comparison with other authors for a similar system of pairs is not available.

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