

APPLICATION POSSIBILITIES OF THE DIFFUSION BONDING FOR MATERIAL 10GN2MFA USED IN THE NUCLEAR POWER INDUSTRY

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The aim of this paper is to demonstrate the application possibilities at using diffusion bonding for creation the homogenous joints regarding their strength properties in comparison with the fusion MMAW method and with the parent material. Connection was carried out for low-alloyed bainitic steel 10GN2MFA that is designed for production shells and bodies of the primary steam-generators collectors which are used in the nuclear power industry. In the paper are theoretically described the basics of diffusion. Moreover, there is described also the proposal and optimization of the diffusion bonding process parameters and optimization of the thermal treatment procedure. Experiments were carried out by means of the thermal-mechanical simulator Gleeble 3500. Welded joints were subsequently evaluated in light of mechanical properties by hardness measurement and metallography.

KEYWORDS

Diffusion Bonding, Gleeble 3500, 10GN2MFA Steel, Mechanical Properties, Process Parameters

1 INTRODUCTION

Production facility for thermal and nuclear power stations can be characterized by creation of non-demountable joints which are supposed to transfer high loading under high temperatures at dynamic stresses. Great majority of these joints is created by the arc welding methods, whereas in light of used technology prevail methods 111, 121 a 141 acc. to ISO 4063.

During the application of fusion welding methods significant heterogeneity occurs which influences changes of the mechanical properties throughout the weld. When creating heterogeneous joint, the structural heterogeneity of joint is much more influenced by the type of additional used material and the level of the parent and filler material mixture. Inhomogeneity in the joint area can be intensively decreased by using special welding methods, such as application of the diffusion bonding. In that case the area of structural heterogeneity is restricted to approximately tens (maximally hundreds) of micrometers. Therefore, degradation of material properties in the welding area is much smaller and also preferable due to dynamic stress.

In the following chapters is described the procedure at proposal and execution of diffusion joint for bainitic steel 10GN2MFA. As the major criterion there was achieving the same or similar joint mechanical properties as has the non-heat affected

parent material. Strength of joint was determined by means of the static tensile test for diffusion welded samples.

2 DIFFUSION THEORY

Diffusion in solid matters can be generally described by two aspects. The first one is called as phenomenological diffusion and takes into account only macroscopic perspective. In light of the second approach, microscopic perspective is considered in the atomic diffusion theory.

This theory respects internal composition of material including crystal lattice interferences. In case of non-existing motive power on atoms, their jumps are random. However, these jumps occur in specific crystallographic equivalent directions. Their type depends on the crystal lattice type and also the on the diffusion mechanism. Motive powers which cause the movement of atoms can be of physical or thermodynamic source. Physical motive powers act on single particles directly, whereas thermodynamic motive powers act volumetrically and depend on temperature, pressure and chemical potential. The explanation of the atomic diffusion theory principals is introduced for example in works [Kazakov 1985].

General phenomenological diffusion concept is described as thermodynamic non-returnable events given by a flow of thermodynamic quantities. This is defined by their amount which passes through the surface unit by time unit from one stage to the other or to a system. According to Onsanger, the flow J_i of thermodynamic value i is linearly dependent on each thermodynamic motion power X_i which depends on specific system according to the Eq. (1), [Pluhar 1987]

$$J_i = \sum_j L_{ij} \cdot X_j \quad (1)$$

where, L_{ij} are Onsanger's kinetic coefficients and X_i are thermodynamic motion powers (e.g. self-diffusion by vacant mechanism when the only motive powers are gradients of primary element chemical potential). According to Fick, diffusion occurs due to the gradient of concentration. It is a special case of general understanding of diffusion which is only valid when simplified assumptions are taken into account and when it is impossible to describe complicated transferable effects. In spite of that, Fick's explanation is technically important due to its simplicity and high number of gathered values of the overall diffusion coefficient D in different thermodynamic systems.

According to the first Fick's law (2), the diffusion flow J_A of element A atoms throughout given time and in the direction of axis x and throughout the surface unit depends on the gradient of element concentration and is proportional to diffusion coefficient D . Simply stated, the first Fick's law expresses speed and amount of atoms which diffuse through material.

$$J_A = -D \frac{\partial C_A}{\partial x} \quad (2)$$

In case of variable diffusion flow, the change of concentration is connected with time. These are termed as the so-called non-stationary diffusion which is described according to the second Fick's law (3),

$$\frac{\partial C_A}{\partial t} = \frac{\partial}{\partial x} \left(D \frac{\partial C_A}{\partial x} \right) \quad (3)$$

where, C_A is the concentration of element A, x is the direction of diffusion concentration change, t is amount concentration change time and D is the diffusion coefficient.

3 BAINITICSTEEL 10GN2MFA

Steel 10GN2MFA is low-alloyed steel with the bainitic structure that is used as a material for production energetics units of nuclear power plants as can be e.g. shells and bodies of primary steam-generators collectors [Kander 2007] – mainly for piles of WWER-1000 type. For such material is characteristic quite high heat-resistance together with very good plasticity.

Production of this steel is a little bit complicated. Company Vítkovice Heavy Machinery uses for its production electric arc furnaces with subsequent refining in ladle, vacuum degassing and uphill casting with the protection of casting flow by the inert gas. Such method provides content of phosphorus below 0,01% and sulphur below 0,005%. Steel that is produced by such method isn't sensitive to stress corrosion cracking (SCC). Its brittle-fracture properties can be improved by means of the inter-critical annealing that seems to be a good alternative for expensive vacuum production of this steel [Kander 2007]. Chemical composition of steel 10GN2MFA determined by the spectrometer Q4 Tasman is given in Table 1. In Fig. 1 is then shown the structure of parent material.

Table 1. Chemical composition of steel 10GN2MFA

Chemical composition	C	Mn	Si	P
wt. %	0.12	0.95	0.22	0.07
Chemical composition	S	Cr	Mo	Ni
wt. %	0.03	0.04	0.58	1.20

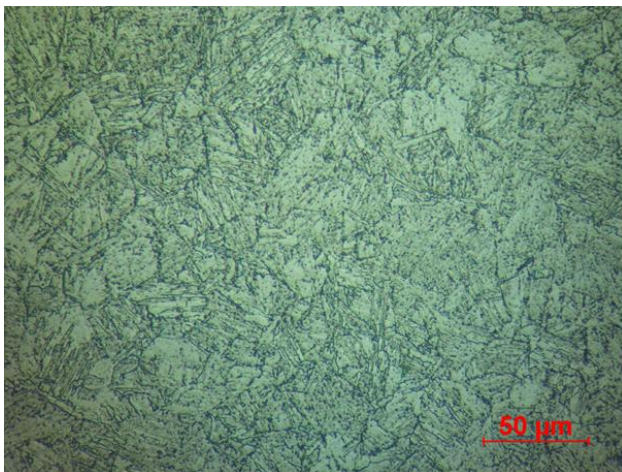


Figure 1. Initial structure of steel 10GN2MFA (magnification 500x)

Table 2. Mechanical properties of steel 10GN2MFA

Material	Proof Yield Str. $R_{p0,2}$ (MPa)	Ultimate Strength R_m (MPa)	Uniform Ductility A_g (%)	Total Ductility A_5 (%)
10GN2MFA	519	711	8.07	17.3

Steel 10GN2MFA can be welded by many common welding methods. If the welded material has pearlitic structure, there isn't required pre-heating. At welding material of bainitic structure, pre-heating is used and after cooling there is bainitic structure. Only under the cooling rates over $100^\circ\text{C}\cdot\text{s}^{-1}$, martensitic structure can be achieved [Moravec 2016]. Table 2 summarizes mechanical properties of steel 10GN2MFA for RT.

4 PROPOSAL AND EXECUTION OF THE EXPERIMENT

The thermal-mechanical simulator Gleeble 3500 was used to carry out the diffusion bonding experiments. Advantage of this system is the possibility of a very precise supervision and regulation of the major technological parameters of the diffusion bonding (temperature, pressure force and time). Heating up of the sample is there realised by the resistive heat which occurs in the sample due to the current passage.

As another advantage of this system there is possibility to use very precious vacuum (10^{-2} – 10^{-1} Pa), eventually inert or reducing shielding gases. In Fig. 2 are subsequently shown samples clamped by the high-temperature jaws of the Gleeble system before the own diffusion bonding.

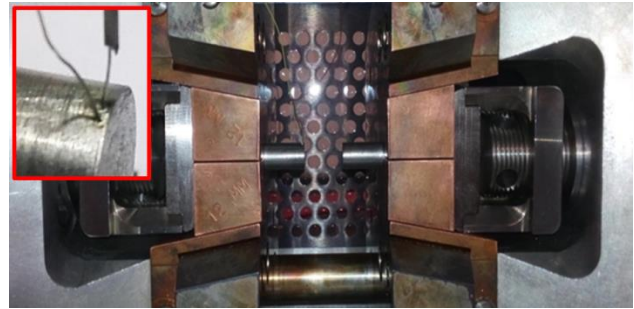


Figure 2. Samples clamped by the high-temperature jaws of the Gleeble system before the diffusion bonding

The controlling thermocouple type K (diameter 0,25 mm), which is used for the temperature supervision on contact area of both materials, is placed 0.3 mm from front side of sample on the material. Close is not possible connect TC with help of capacitor welding.

The largest generating of resistive heat occurs in the place of the largest transitional resistance which is placed directly on the touching surfaces of welding samples. For this reason unequal thermal field occurs in the welding samples with the maximum in the place of contact between both samples (zone where the controlling thermocouple is placed).

To realize the experiment there were used samples having the cylindrical shape, length $l = 50$ mm and diameter $d = 12$ mm. Front contact surfaces of all testing samples were machined for roughness $R_a = 1.2$ µm. Proposal of the diffusion bonding basic parameters was done based upon the recommendations from references and by realization the so-called "cascade" test. Reference [Kazakov 1985] recommended to select exposure temperature as 0.6 till 0.9 of melting temperature T_m . For steel 10GN2MFA it was finally chosen the exposure temperature as $0.75 \cdot T_m$ (so 1125°C). It is temperature under which occurs a diffusion of high intensive and simultaneously there isn't intensive growing of austenitic grain in tested material.

Holding force which results as pressure on the contact areas was chosen for the relevant temperature by means of so-called "cascade" test. During this test is sample firstly heated up to exposure temperature and after that is applied holding fore. Such force acts for certain time (mostly 5 min) and during such time is monitored the sample deformation by means of strain gauge extensometer. In the case that there isn't deformation, holding force is increased by the given magnitude up to the moment when the sample starts to deform. After that, such test is terminated. There should occur micro-deformations on the contact areas at diffusion bonding, because it increases the area for diffusion passage of atoms. However, there should not be deformation of whole testing sample.

Based upon the cascade test was for the all experiments chosen holding force as 1.2 kN, which corresponds to the pressure on the contact areas as 10.6 MPa.

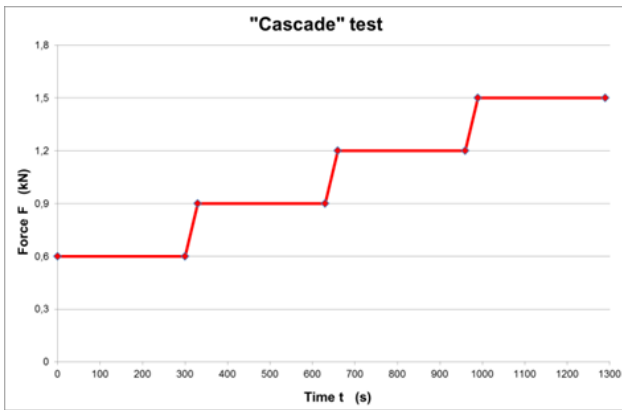


Figure 3. Evolution of "cascade" test for determination the holding force under the exposure temperature 1125°C

So for the first experimental phase there was only one variable as exposure time (holding time) on the given temperature. Such time varies from 600; 1200 up to 2400 sec. Heating rate to the temperature 1125°C was as 10°C·s⁻¹ and when the exposure time was out, welded sample was cooled down by cooling rate as 10°C·s⁻¹. During the diffusion jointing was in the working chamber vacuum of 1.8·10⁻¹ Pa. In Fig. 4 is shown such diffusion jointing of sample under the temperature 1125°C.

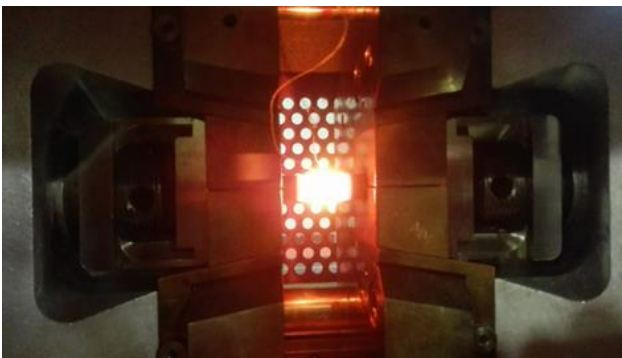


Figure 4. Diffusion jointing of sample at temperature 1125°C

For every exposure time there were carried out three testing measurements, whereas two samples were used for static tensile test and one sample for metallographic evaluation and hardness measurement. Unfortunately there is no place in this article to show figure of microstructure. After creation of diffusion joint (Fig. 5), from samples were prepared testing sample on which were subsequently carried out static tensile test acc. to the standard ČSN EN ISO 6892-1. Results of this test are given in table 3. All samples revealed fracture beyond the diffusion weld and HAZ, how can be clearly seen from Fig. 6.



Figure 5. Diffusion jointed sample at temperature 1125°C; holding force 1.2 kN and exposure time 1200 sec



Figure 6. Diffusion jointed samples after static tensile test revealed fracture beyond the diffusion joint area

In all tested cases, results from this first experimental phase revealed very good strength properties in comparison to the parent material. However, ductility values (A_g and A_5) were much lower than parent material (8,07 and 17,3 %). That is why

there was needed to realize second experimental phase to increase ductility of joints by welding parameters modification.

Table 3. Mechanical properties of diffusion joints

Sample No. / Time	Proof Yield Str. $R_{p0,2}$ (MPa)	Ultimate Strength R_m (MPa)	Uniform Ductility A_g (%)	Total Ductility A_5 (%)
No. 1 (600 s)	782	854	0.77	1.02
No. 2 (600 s)	775	861	0.81	0.98
No. 3 (1200 s)	742	926	0.99	1.74
No. 4 (1200 s)	747	933	1.06	1.92
No. 5 (2400 s)	711	893	0.92	1.13
No. 6 (2400 s)	704	888	0.89	1.07

4.1 Experiments leading to increase ductility

Second experimental phase arose from the diffusion joint process parameters of the highest strength and ductility values (samples No. 3 and No. 4). As an exposure temperature there was 1125°C, holding time 1200 sec and holding force 1.2 kN. Achieved values of yield strength $R_{p0,2}$ and ultimate strength R_m for the joint were higher approx. by 20% than for the basic material (at very low ductility). From the structural analysis and hardness HV10 measurement was observed the pure bainitic structure of hardness 311 HV. As a presumption to achieve higher ductility with keeping the sufficient strength, there was lowering of cooling rate, even. to lower exposure temperature. Experiments were divided into two stages. At the first stage were samples jointed at 1125°C, holding force 1.2kN for time 1200 sec and heating rate 10°C·s⁻¹. Cooling rates were as 1°C·s⁻¹ and 0,1°C·s⁻¹. Such cooling rates were proposed acc. to CCT diagram (see Fig. 7) so that achieved structure was on the boundary of bainitic and perlite area (hardness cca. 235 HV), even. bainitic-perlite structure with hardness about 210 HV. The 2nd stage used same values, only exposure temperatures were lowered on 0.7 T_M (1050°C) and 0,65 T_M (975°C).

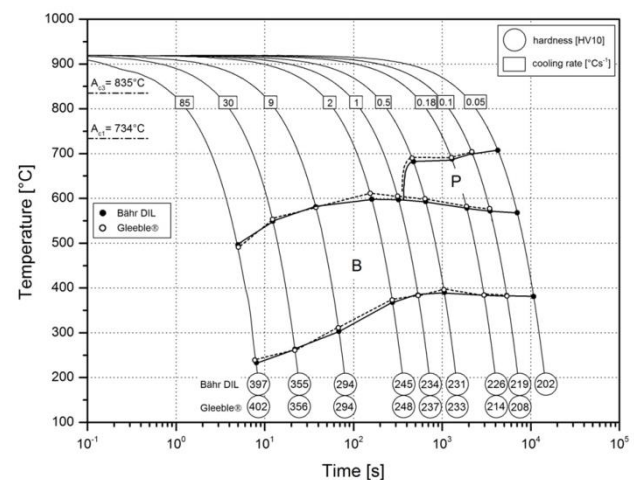


Figure 7. CCT-diagram of 10GN2MFA steel constructed for two different heating methods [Moravec 2015]

Also here were for every testing configuration carried out three measurements, whereas two samples were used for static tensile test and one sample for metallographic evaluation and hardness measurement. As it is obvious from Table 4, by lower

cooling rate there was rapid increase in ductility of joint and whole sample. At samples No. 7 and No. 8 was cooling rate $0.1^{\circ}\text{C}\cdot\text{s}^{-1}$ and for No. 9 and No. 10 it was $1^{\circ}\text{C}\cdot\text{s}^{-1}$.

Table 4. Mechanical properties of diffusion joints with lower cooling rate ($T = 1125^{\circ}\text{C}$; $F = 1.2 \text{ kN}$; $t = 1200 \text{ sec}$)

Sample No. / Time	Proof Yield Str. $R_{p0,2}$ (MPa)	Ultimate Strength R_m (MPa)	Uniform Ductility A_g (%)	Total Ductility A_5 (%)
No. 7 (CR $0,1^{\circ}\text{C}\cdot\text{s}^{-1}$)	491	688	7.52	19.88
No. 8 (CR $0,1^{\circ}\text{C}\cdot\text{s}^{-1}$)	496	691	7.48	19.56
No. 9 (CR $1^{\circ}\text{C}\cdot\text{s}^{-1}$)	530	721	8.15	17.21
No. 10 (CR $1^{\circ}\text{C}\cdot\text{s}^{-1}$)	527	728	8.09	17.34

That is why for experiments with lower exposure temperature there was hold holding force 1.2 kN, exposure time 1200 sec, heating rate $10^{\circ}\text{C}\cdot\text{s}^{-1}$ and cooling rate was chosen based upon the previous results as $1^{\circ}\text{C}\cdot\text{s}^{-1}$. In Table 5 are summarized the measured values of mechanical properties. From these results it is obvious that samples jointed under the lower temperature revealed the lower strength values approx. by 10% as well as the lower values of ductility.

Table 5. Mechanical properties of diffusion joints with lower exposure temperature.

Sample No. / Time	Proof Yield Str. $R_{p0,2}$ (MPa)	Ultimate Strength R_m (MPa)	Uniform Ductility A_g (%)	Total Ductility A_5 (%)
No. 11 (T = 1050°C)	498	692	7.11	16.81
No. 12 (T = 1050°C)	492	696	7.04	16.93
No. 13 (T = 975°C)	478	674	6.78	15.93
No. 14 (T = 975°C)	483	669	6.83	15.86

5 CONCLUSIONS

As it was shown in this paper, by application of diffusion bonding it is possible to achieve the same values of mechanical properties as has the parent non-heat affected material. Own procedure to optimize process parameters was applied for steel 10GN2MFA that is used at production of primary nuclear power plants collectors. Nowadays, this steel is welded exclusively by arc welding methods (mainly 111, 121, 141acc. to ISO 4063). However, by their utilization there is degradation of mechanical properties – mainly in HAZ. That is why these joints need subsequent thermal treatment, but already after its application the total strength of joint achieves cca. 85 up to 90% of parent material strength and toughness correspond to cca. 80% of parent material toughness [Moravec 2013].

In the paper was presented the process leading to the adjustment of optimal process parameters – in light of exposure temperature and holding time, as well as in light of proper holding force that causes micro-deformations only on

the boundary between jointed materials. Moreover, there was proved that at diffusion jointing is necessary (as one of the basic parameter) to take into account also cooling rate from the exposure temperature.

The best results of diffusion joint on steel 10GN2MFA was achieved at exposure temperature 1125°C for time 1200 sec, holding force 1.2 kN with subsequent cooling rate $1^{\circ}\text{C}\cdot\text{s}^{-1}$. The whole process was performed in vacuum of $1.8\cdot 10^{-1} \text{ Pa}$. Due to such procedure there was obtain diffusion joint of high quality with ductility comparable with the parent material and strength properties actually a little bit higher than of parent material. Cooling rate $1^{\circ}\text{C}\cdot\text{s}^{-1}$ prolongs the whole process by approximately 30%. On the other hand, the PWHT (which is required during the fusion-welding process) is eliminated. It might seem that in light of productivity is time of process over 1200 sec quite uneconomical for such small cross-section of sample. Nevertheless, it is necessary to take into account that time of process will be the same for samples of diameter 12 mm as well as for samples of diameter 120 mm and in this case it would be highly economical process. The total holding time can be decreased e.g. by application of higher exposure temperature under which diffusion will have higher intensity. On the other hand, it will also lead to grain growth which would subsequently influenced also mechanical properties of joint. Finally, as the only one limiting factor at diffusion bonding there is requirement to perform whole process in vacuum, eventually in the shielding gas.

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