

THE INFLUENCE OF INTRODUCING REFRACTORY COMPOUNDS INTO THE WELD POOL ON THE WELD METAL DENDRITIC STRUCTURE

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A study was conducted in order to expand the knowledge base on the mechanisms of the influence of refractory compounds in the weld pool on modification of weld metal structure. Results of experiments on inoculation of TiN, SiC, NbC, VC, TiC, TiO₂, Al₂O₃, MgO, ZrO₂ dispersed particles to the liquid metal of the weld pool are given. Possibility of the influence of refractory inoculants as the melt «microcoolants», centers for epitaxial initiation of a new phase in the liquid metal and surface-active compounds on modification of the dendritic structure of the welds was analyzed. It is shown that under the conditions of welding low-alloyed steels by flux-cored wire of «metal core» type in shielding gas atmosphere, the refractory compounds, inoculated to the weld pool, the most actively influence modifying of the primary structure, as surface-active compounds. It is established that the change of dendrite morphology in the weld metal influences the temperature range of the recrystallization process, formation of secondary microstructure and mechanical properties of the welds. 10 Ref., 5 Tables, 8 Figures.

Keywords: low-alloyed steel, weld, refractory inclusions, dispersed particles, dendritic structure, modification, austenite grains

At present, structural steels make up the main base of welded metal structures. The range of low-alloyed rolled stock is characterized by a wide variety, and development of new grades is going on, which are capable of providing further improvement of service properties of welded metal structures. Improvement of the characteristics of higher and high strength low-alloyed steels is achieved due to complication of their production technology, comprehensive alloying by such elements as nickel, molybdenum, niobium and vanadium. The high cost of these elements in Ukraine makes urgent the investigations, aimed at finding alternative possibilities of metallurgical influence on metal structure formation. Such work is performed both for low-alloyed rolled stock, and metal of welds produced during its welding. It should be noted that investigations in the welding field are characterized by additional difficulties, compared to traditional metallurgy, which are associated with a high speed of the processes, presence of gas-plasma phase, increased gradient of contacting phase composition, and a number of other factors.

The current concepts of formation of the primary weld metal structure are based predominantly on investigations of the processes of solidification of large metal volumes in steel ingot production. The identity of metallurgical processes, which are in place in these

two cases, allows using the investigation results in the field of ferrous metallurgy, but a significant difference in the parameters of flow kinetics, temperature and concentration gradients, necessitates performance of special investigations as regards the features of formation of primary structure of weld metal.

In welding low-alloyed steels with carbon content above 0.18 %, in keeping with the state diagram of Fe–C system, melt solidification begins at temperatures below 1500 °C. It should be noted, however, that the changes in alloying element content in the liquid metal on the solidification front, which are caused by concentrational overcooling, influence the lowering of this temperature. From practical experience it is known that such a lowering of temperature is equal to approximately 10 °C.

A feature of formation of weld metal primary structure is predominantly epitaxial crystal nucleation on partially melted grains of the base metal. Base metal grains are of a nonuniform orientation, so the crystals, nucleating in the weld pool, will also have different orientation, and those of them, the axes of which coincide with the direction of the largest heat flow, will grow, the most intensively absorbing smaller neighbours. Such crystals, divided by high-energy boundaries, form the structure of primary dendrites. The curvature of the solidification front, i.e. the cur-

vature of the solid/liquid surface in the weld pool that changes, depending on the energy source movement, leads to instability of the front and formation of the structure in the form of columnar dendrites. During cooling, δ -, γ -transformation occurs with formation of low-angle boundaries, inside the primary dendrites. Presence of a liquation interlayer of liquid metal, and of surface-active elements, segregation of refractory inclusions in the zone between the growing dendrites, determine the morphology of primary austenite grains, that further on will affect the formation of final microstructure of the metal of welds and their mechanical properties.

One of the promising directions of improvement of the weld mechanical properties is refinement of its structure grains. The methods of metal modification are widely used with this purpose. At present, there is no single description of the mechanism of iron alloy modification. However, the wide application practice is indicative of the effectiveness of such processes. It is known that addition of refractory particles to the metal melt facilitates nucleation of equiaxed crystals in the concentrational overcooling zone, and increase of the number of nucleation centers influences the lowering of the dendrite growth rate. This is also promoted by presence of surface-active compounds, which lower the dendrite surface energy that leads to reduction of primary structure dimensions. To increase the effectiveness, it is rational to combine the impact of refractory particles and surface-active compounds [1]. In addition, in any case, the features of weld pool solidification should be taken into account at selection of modifiers.

This work is devoted to investigation of the features of modifying impact of refractory compound particles, which differed by their physico-chemical indices, with the purpose of substantiation of selection of metal structure modifiers for welds of low-alloyed high-strength steels.

1. Influence of inoculants on the dendritic structure. Formation of the metal alloy structure begins from appearance and growth of dendrites. At weld pool solidification, these processes occur much faster, than in the case of large metal ingots, but, nonetheless, formation of weld metal microstructure, determining the entire complex of its mechanical properties, begins exactly at this stage. Unfortunately, investigations of the features of formation of the dendritic structure of weld metal are not given too much attention in scientific-technical publications. This section gives the results of investigation of the influence of refractory compound particles inoculation into the weld pool on the morphology of dendrites in the weld metal.

1.1 Experimental procedure. By the nature of their action, the modifying additives can be conditionally divided into three types: «microcoolants», epitaxial centers of new phase nucleation, and surface-active compounds. Refractory compounds were selected for investigations. These compounds allow making a conclusion about the effectiveness of each of these directions at formation of the structure of metal of welds of low-alloyed steels with the yield limit of up to 600 MPa. The list of joints selected for experiments and their physico-chemical parameters are given in Table 1.

In order to reveal the nature of the refractory compound action, it is rational to store it in the form of a crystalline phase in the temperature range of metal melt existence. Performed calculations showed that at addition of the selected compounds to the weld pool, the size of the refractory particles should be not less than 200 μm . At addition of particles of 200–500 μm size, they can participate in the process of melt solidification as crystalline inclusions of exogenic type of 50–100 nm size.

Investigations of the influence of refractory particles on modifying the metal structural components were performed on samples of deposited metal, which

Table 1. Physico-chemical characteristics of compounds used in experiments

Compound	$T_m, ^\circ\text{C}$	Crystalline lattice type	Lattice parameter, nm	Mismatch to δFe size, %	Interphase energy, mJ/m^2
ZrO ₂	2715	Tetragonal	a-3.640c-5.152	1	2863
VC	2810	FCC	a-4.159	13	1559
MgO	2825	FCC	a-4.213	14	2226
TiN	2930	FCC	a-4.260	16	393
TiC	3160	FCC	a-4.336	18	884
NbC	3608	FCC	a-4.469	21	548
TiO ₂	1843	Tetragonal	a-4.593c-2.959	25	2444
Al ₂ O ₃	2044	Rhombohedral	a-5.120 α -55.25°	39	972

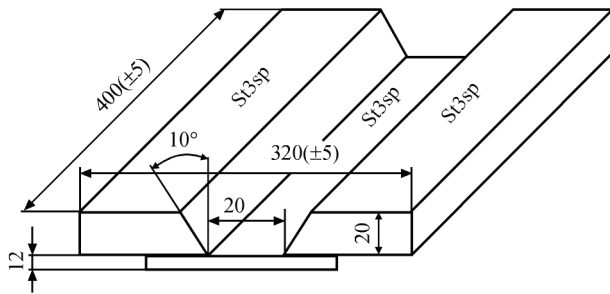


Figure 1. Butt joint preparation for cutting out weld metal samples in keeping with ISO 14171 requirements

were cut out of the last metal layer of a butt weld that was produced using experimental flux-cored wire of «metal core» type of 1.6 (±0.1) mm in M21 shielding gas atmosphere to ISO 14175 standard [2]. Butt joints of 1.4 type to ISO 9692-1 standard [3] (Figure 1) were welded at reverse polarity direct current of 230 (±10) A at arc voltage of 28 (±2) V and welding speed of 18 ((±0.5) cm/min. After each pass the butt joint was cooled in air up to the temperature not higher than 120 °C. The process heat input was equal to 26–28 J/cm.

Particles of refractory compounds of 200–500 μm size were added (inoculated) to the molten metal through the core of flux-cored wire, which was fed to the weld pool in the form of «cold» filler. Schemes of preparation of the butt, welded joint and areas of cutting out the samples for investigations are given in Figures 1–3. The inoculated filler content was equal to 0.1 % of the weld pool mass. The composition of the studied samples and mechanical properties of weld metal are given in Tables 2 and 3, respectively. Figure 2 gives the scheme of the butt joint filling, according to which the passes from the first to the ninth were made by flux-cored wire of the base alloying system, and passes from the tenth to the eighteenth were made using «cold» filler, to the core of which refractory compound particles were added.

The welds had a conditional designation that corresponded to a refractory compound, which was inoculated into the weld pool. Samples of weld metal, with base alloying system (conditional designation BA), were also used in the investigations. In order

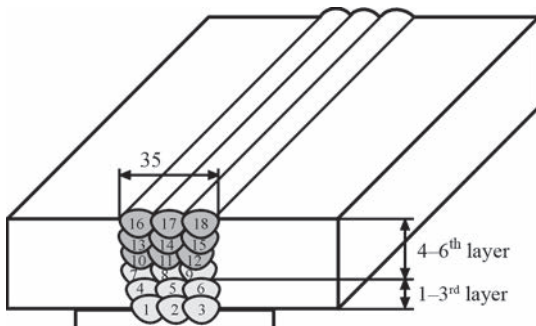


Figure 2. Scheme of bead layout at filling of the butt joint

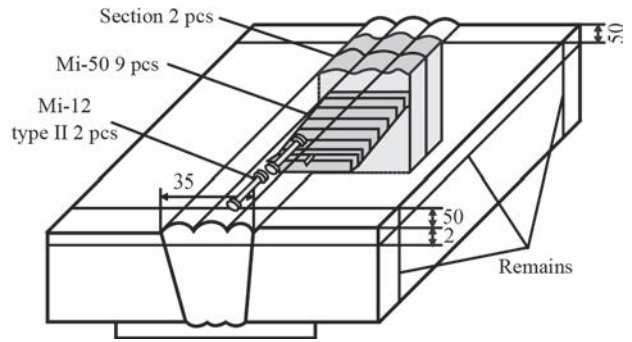


Figure 3. Scheme of sample cutting out to determine the chemical composition, mechanical properties and structure of weld metal

to separate the impact of titanium-containing compounds from increase of titanium content, weld metal samples, additionally alloyed by titanium (conditional designation FeTi), compared to the base system, were also included into investigation.

Transverse samples were cut out of the welded joints in order to study the structure and phase composition of the weld metal, as well as mechanical properties of the welded joints by the scheme, given in Figure 3.

At metallographic investigations the fraction of structural components in the metal and distribution of nonmetallic inclusions were determined. The microstructure was studied by the methods of optical metallography, using light microscope «Neophot-30» with image displaying in the computer screen.

Metallographic investigations of the last pass were conducted in optical microscope «Neophot-30» on transverse microsections of the weld metal, polished and etched in 2 % nital solution. The same sections were used for investigations in the scanning electron microscope. Microstructural analysis was conducted in keeping with the requirements of ISO 17639 standard [4] and IIW recommendations [5].

The primary weld metal structure was studied on polished samples, etched in a boiling saturated solution of sodium picrate ($C_6H_2(NO_2)_3ONa$) in water. Samples were cut out in the direction normal to the longitudinal axis of the weld, so that dendrites were visible on the section surface. They grew towards the largest thermal gradient in the weld pool. When studying the primary structure by the images, obtained by optical microscopy, the dimensions of columnar dendrites (dimensions λ_d in Figure 4) were determined.

The nature of distribution of nonmetallic inclusions in the weld metal structure was analyzed, using the images of the surface of polished unetched sections, obtained in optical microscope «Neophot-30». The processed images were identified by special software. Each inclusion in the approximation was regarded as a certain polyhedron, the area of which is measured and recorded in the Table for further math-

emational processing. Thus, each inclusion visible in the micrograph and taking up more than 3 pixels, was analyzed. This way a statistical sample was formed by the dimensions of nonmetallic inclusions in the weld metal. Further processing was performed using proprietary software, which ensures a complete cycle of processing the obtained data and deriving the results in the form of a spreadsheet. Then, the obtained sample was used to plot a histogram of frequencies with preset size ranges. Such a histogram of frequencies represents the size distribution of inclusions in the metal matrix.

Mechanical properties of metal of the studied welds were determined by the procedures, given in ISO 15792 standard [6].

1.2. Investigation results. The results given in Table 2 show that by its chemical composition the met-

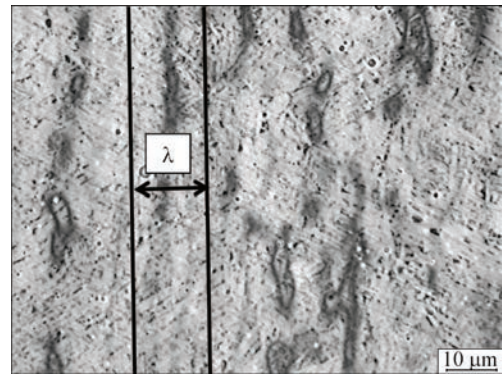


Figure 4. Example of dendrite structure ($\times 1000$) of the studied samples (λ is the distance between primary dendrite axes)

al of the studied welds corresponds to low-alloyed steels. Addition of certain inoculants to the weld pool affected only the content of aluminium and titanium in the weld metal that is attributable to the features of

Table 2. Chemical composition of the studied weld metal, %

Weld	C	Si	Mn	S	P	Cr	Ni	Mo	Al	Ti	Zr
BA	0.034	0.340	1.21	0.021	0.020	0.12	2.13	0.28	0.028	0.013	N/D
FeTi	0.036	0.335	1.22	0.022	0.021	0.14	2.14	0.26	0.038	0.029	»
TiN	0.035	0.317	1.24	0.019	0.009	0.14	2.15	0.26	0.036	0.021	»
VC	0.052	0.227	1.21	0.022	0.021	0.14	2.13	0.25	0.027	0.004	»
NbC	0.049	0.253	1.19	0.021	0.020	0.13	2.15	0.27	0.029	0.003	»
SiC	0.053	0.351	1.20	0.020	0.025	0.12	2.12	0.26	0.025	0.004	»
TiC	0.046	0.340	1.25	0.021	0.019	0.13	2.15	0.24	0.023	0.021	»
TiO ₂	0.035	0.405	1.24	0.018	0.021	0.11	2.17	0.27	0.031	0.027	»
Al ₂ O ₃	0.034	0.424	1.26	0.019	0.023	0.11	2.15	0.29	0.042	0.015	»
MgO	0.031	0.227	1.21	0.025	0.024	0.14	2.15	0.29	0.023	0.013	»
ZrO ₂	0.033	0.223	1.25	0.024	0.024	0.12	2.14	0.30	0.024	0.013	0.06

Table 3. Mechanical properties of the studied weld metal

Weld	σ_p , MPa	$\sigma_{0.2}$, MPa	δ , %	ψ , %	KCV, J/cm ² at T, °C			
					+20	0	-20	-40
BA	685	610	15	54	97	87	75	53
FeTi	747	690	19	60	74	69	63	61
TiC	716	644	19	63	110	97	85	73
TiN	712	580	5.3	14.7	55	47	40	–
SiC	726	650	21	62	85	72	65	61
VC	780	706	14	56	57	55	52	–
NbC	820	757	18	57	45	39	31	–
TiO ₂	709	636	19	57	85	72	60	50
Al ₂ O ₃	728	621	17	54	82	58	50	36
MgO	644	586	19	60	103	85	69	60
ZrO ₂	649	592	21	64	97	91	84	76

Table 4. Results of analysis of nonmetallic inclusion content in the weld metal

Weld	Inclusion fraction (%) in weld metal	Inclusion fraction (%) in the size range			
		<0.31 μm	0.37–0.61 μm	0.67–0.97 μm	1.03–1.57 μm
BA	0.47	29.63	46.67	17.65	6.05
FeTi	0.58	27.36	52.01	14.77	4.99
TiC	0.62	24.51	45.80	22.69	7.00
TiN	0.77	31.11	47.97	16.32	4.60
VC	0.61	26.56	46.48	19.24	7.72
NbC	0.58	29.77	45.74	18.97	5.52
SiC	0.68	27.16	44.89	21.59	6.36
TiO ₂	0.81	30.89	40.96	20.37	7.78
Al ₂ O ₃	0.74	30.48	43.95	18.50	7.07
MgO	0.62	33.09	44.69	14.73	6.40
ZrO ₂	0.55	35.53	45.43	14.21	4.82

the processes of deoxidation under the impact of the added compounds.

Results given in Table 3 are indicative of the fact that addition of refractory compound particles to the weld pool, makes a noticeable impact on the mechanical properties of the weld metal. Such an impact is associated with the changes in the metal structure, the consideration of which is the subject of this study.

1.3. Size distribution of nonmetallic inclusions. One cubic millimeter of the weld metal can contain up to 10¹⁰ nonmetallic inclusions of 30 nm to 3 μm size. However, only about 30% of them can have an active impact on new phase nucleation. Nonmetallic inclusions in the studied weld metal differed by their morphology. Very fine inclusions of up to 0.3 μm size had single-phase structure in the form of refractory

oxides, carbides or nitrides. Coarser inclusions of up to 1.0 mm size consisted of two or three phases — refractory inclusion and oxide phase based on manganese, silicon or titanium compounds. Inclusions of more than 1.0 μm size had multilayer morphology with presence of low-melting sulphides on the outer surface. It is believed that such inclusions form in the solid-phase metal solution and cannot affect formation of the primary structure [7]. Inclusions of the second type make the most noticeable impact on new phase appearance at formation of the secondary microstructure [8], but their participation in the processes of primary structure nucleation and growth still has not been widely covered. Without doubt the finest refractory inclusions were present in the liquid metal, contacted the growing dendrites and could affect their development. Proceeding from the above considerations, Table 4 gives the results of determination of both the total content of inclusions in the weld metal, and their distribution according to the three specified dimensional ranges.

1.4. Morphology of primary structure grains. Used methods of etching weld metal samples allowed forming data bases on the dimensions of both the dendrites and primary austenite grains. Obtained results are given in Table 5.

Figure 5 gives the images of weld metal samples after etching to reveal the primary structure.

1.5. Discussion of the obtained results. Influence of inoculants on primary structure morphology. Dendrites, which form in the weld metal, initiate on base metal grains, and grow mainly along the maximum thermal gradient to the weld center that leads to columnar dendrite shape. The width of columnar dendrites is associated with the size of base metal grains on the fusion line. However, as growing of some unfavourable oriented dendrites was suppressed by those

Table 5. Dendrite interaxial spaces λ and primary austenite grain size d

Size, μm	BA	FeTi	TiC	TiN	VC	NbC	SiC	TiO ₂	Al ₂ O ₃	MgO	ZrO ₂
Dendrites	80±17	57±13	100±20	55±12	70±15	50±12	100±21	100±19	57±13	152±23	158±22
Austenite	70±15	45±10	80±16	40±9	65±13	35±8	80±18	80±17	45±10	70±13	75±15

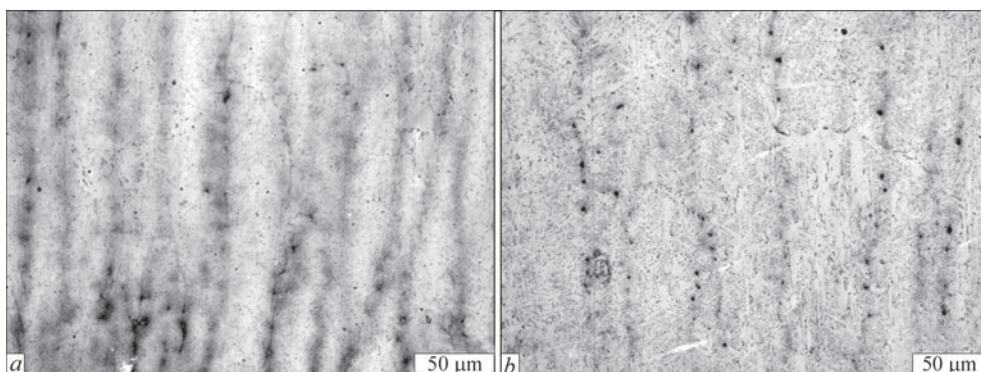


Figure 5. Primary structure of weld metal BA (a) and ZrO₂ (b)

that grew faster, than certain dendrites increased in size, and, as was expected, were wider than the initial base metal grains.

As is seen from the results, given in Table 5, inoculation of refractory compound particles to the weld pool affects the morphology of dendrites and primary austenite grains. The composition of the refractory compounds for the experiments was selected with the purpose of revealing one of the possible mechanisms of their impact on the primary structure. According to the mechanism of inclusion action as «microcoolants», their effectiveness is determined by the level of latent solidification energy. The process of inclusion melting is accompanied by consumption of this energy and respective lowering of the temperature of the metal melt, in which they are located [8]. The higher the melting temperature of the compound, the larger the value of latent energy of melting and the greater the impact, which such inclusions will have on melt overcooling. Increase of overcooling level should affect the solidification rate, i.e. the dendrite size. Figure 6 shows the histograms, which were plotted by the results, given in Tables 1 and 5.

As one can see from the given results, there is no unambiguous dependence between the melting temperature of particles inoculated into the weld pool and the width of dendrites in the weld metal. The largest width of the dendrites is characteristic for the metal of welds, into the composition of which inclusions with melting temperature of 2715 and 2825 °C were inoculated, whereas dendrites in the metal of welds, inoculated by inclusions with a higher temperature, have much smaller dimensions.

Another mechanism of modification of the metal structure is associated with the possibility of epitaxial nucleation of the new phase in metal melts on the surface of nonmetallic inclusions, having crystalline lattice parameters, close to the dimensions of the lattice of δ - or γ -iron. The smaller the difference in the lattice dimensions, the higher should be the intensity of nucleation of the hard phase in the melt, and the smaller should be the dimensions of this phase (i.e. of dendrites) [9]. Figure 7 shows the histograms, plotted by the results, given in Tables 1 and 5.

Mismatch parameter D was calculated by the following formula

$$D = \frac{\gamma_{\delta} - \gamma_i}{\gamma_{\delta}},$$

where γ_{δ} and γ_i are the parameters of δ Fe crystalline lattice and inclusion, respectively.

Results of metallographic analysis showed that the greatest width is found in dendrites of the met-

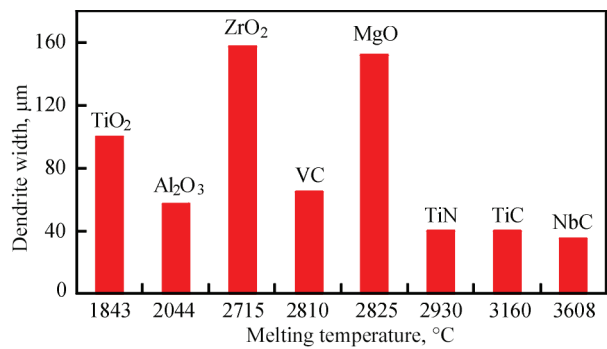


Figure 6. Dependence between the melting temperature of particles inoculated into the weld pool and width of dendrites in the weld metal

al of welds, into the composition of which particles of zirconium and magnesium oxide were inoculated, which have the lowest parameters of mismatch to δ Fe lattice. Here, the weld metal dendrites, to the composition of which vanadium carbide particles with the same mismatch parameter as that of magnesium oxide were added, have noticeably smaller size. It can be assumed that the mechanism of dendrite nucleation on nonmetallic inclusions with crystalline lattice parameters, close to δ Fe dimensions in the metal of the studied welds, did not have a leading role.

It is one of the possible mechanisms of modifying the metal structure, which is related to the impact of surface-active nonmetallic inclusions on interphase energy on the melt solidification front. The value of adsorption of the surface-active compound (Γ) is determined by its concentration in the surface layer, and the connection between adsorption and change of surface-tension value (σ) is described by Gibbs equation

$$\Gamma = -\frac{Q}{kT} \frac{d}{dQ}$$

where Q is the volume activity of the compound.

Compounds that lower the surface tension ($\frac{d\sigma}{dQ} \leq 0$), should accumulate on the interface, and

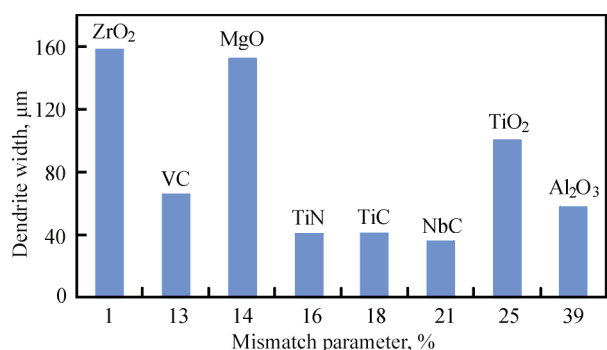


Figure 7. Dependence of the lattice parameter of mismatch of particles inoculated into the weld pool to δ Fe lattice and width of dendrites in the weld metal

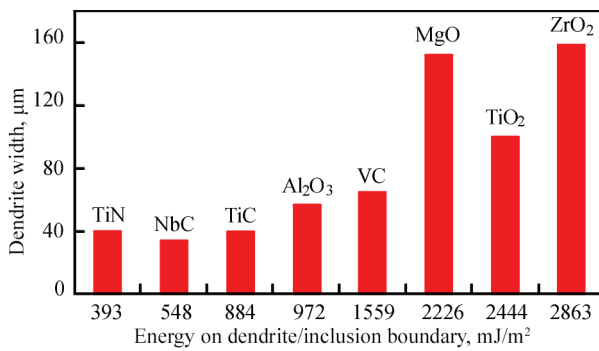


Figure 8. Dependence between the energy on dendrite boundary and particles inoculated to the weld pool and width of dendrites in the weld metal

compounds increasing surface tension $\left(\frac{d\sigma}{dQ} \geq 0\right)$ must move away from it [10].

Reduction of such energy in the zones of inclusion contact with the growing dendrite is accompanied by lowering of the required degree of overcooling and intensification of dendrite growth. Figure 8 shows a histogram, plotted by the results given in Tables 1 and 5.

Results given in Figure 8, are indicative of the fact that the change of surface energy on dendrite boundaries due to inoculation of refractory compound particles into the weld pool, affects the size of the dendrites. The case of adding TiO₂ particles somewhat falls out of this series that may be related to formation of more stable titanium compounds at high temperatures. Obtained results correspond to the dependence, which is described by the above Gibb's formula. Presence of inoculants in the metal melt, which lower the energy of the dendrite boundaries, promotes intensification of their growth in the direction of maximum temperature gradient. Dendrites with a large length-to-width ratio form in the metal of these welds, while inoculation of compounds that increase the dendrite grain boundary, leads to formation of wider dendrites with low length-to-width ratio.

As was already noted, the processes of formation of the weld structure occur at much higher rates than at solidification of large metal ingots. This can serve as an explanation of the fact that the activity of refractory particles in the weld pool melt as «microcoolants» or centers of epitaxial nucleation of the new phase is not as high as the manifestation of their surface-active impact.

During cooling a substructure appears in the columnar dendrite body, which is divided into grains with low-angle boundaries. This substructure can develop in different ways, depending on solidification

parameters G/R , where G is the thermal gradient, and R is the solidification front speed. For a dendritic structure with a wider dimension of the dendrites, the solidification front speed is lower, so that they form primary austenite structures with coarser grains (Table 5). Thus, the weld metal forms a primary structure, in which the austenite grains are divided by both the high-energy and low-energy boundaries. The topography of these boundaries affects the nature of further development of weld metal structure during cooling.

Conclusions

Investigations of the impact of dispersed particles of refractory compounds inoculated into the weld pool on the features of formation of weld metal dendrite structure were studied. The connection between physico-chemical properties of the inoculants and morphology of the dendrites was analyzed. It is found that the inoculated compounds the most effectively exert their influence as surface-active substances at solidification of weld pool metal. It is noted that the refractory inoculants do not have any systematic approach on the solidification process as «microcoolants» or centers of epitaxial nucleation of the new phase. It is established that the most massive grains of primary austenite form in the body of the widest dendrites.

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Received 26.10.2020