

CLASSIFICATION OF METHODS OF MODIFICATION AND MICROALLOYING OF DEPOSITED METAL (Review)

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A literature review of the main methods of modification and microalloying of deposited metal was made. It was shown that, in contrast to the «microalloying» term, the «modification» term should mean not only a method of introducing small additives of chemical elements or their compounds into the deposited metal but also various physical effects or technological processes aimed at achieving the same goal – refinement of micro- and macrostructure of metal, cleaning of grain boundaries and near-boundary zones, improving the complex of technological, mechanical and operational properties of steels and alloys. Examples of such physical effects are application of vibrations to a product in the process of surfacing; input of energy from a pulsed heat source; pulsed electrode or filler wire feed; application of sources with modulation of welding current; external electromagnetic effect, etc. The classification of modification methods, depending on the scheme of their implementation and impact on the properties of the deposited metal is given. The main advantages and disadvantages of methods of introducing modifying and microalloying additives into the deposited metal are shown. It was determined that from the considered methods of modification, the chemical method is the simplest and the most rational, which implies introducing modifiers into the deposited metal directly through the charge of flux-cored electrode wires. This method is technologically and economically simple and versatile, and can be used with minor changes in electric arc, electroslog and plasma surfacing. 33 Ref., 1 Table, 3 Figures.

Keywords: arc surfacing, deposited metal, flux-cored wire, modification, microalloying, metal structure, service properties

It is well-known that «microcontent» of chemical elements or their compounds in the metal or alloy can significantly influence the nature of metal solidification, shape and composition of nonmetallic inclusions, structure of grain boundaries and near-boundary regions, weldability, hardenability, thermal and wear resistance, etc., i.e. a whole range of technological and service properties [1].

One of the technologically and economically effective methods of controlling the metal «microcontent» at electric arc surfacing is its modification and microalloying. Investigation of this problem is rather widely described in literature as regards modification of steels and alloys, produced by casting methods, as well as welds. At the same time, information on application of different methods of modification and microalloying at surfacing is rather scattered. Therefore, the objective of this work, which consists in analysis of the main methods of modification and microalloying, which can be applied at different surfacing processes, as well as their main advantages and disadvantages, apparently, is quite urgent.

Despite the fact that the «modification» and «microalloying» terms often mean similar processes, which consist in addition of small amounts of certain elements to the metal, which change the metal structure and properties, this idea is not entirely accurate [2].

The process of metal modification has been known for a rather long time. In keeping with the data of [3], this process was first used to obtain high-quality cast iron with application of silicocalcium and ferrosilicon as modifiers. At the same time, there exist several thoughts at present about what exactly should be understood by «modification» term. So, in keeping with [2, 4], modification is a process of active regulation of initial crystallization and/or change of the degree of dispersity of the crystallizing phases, by adding to the melt small amounts of individual elements or their compounds. In work [5], modification means something more, namely any technological process of producing alloys with a fine-grained structure with the purpose of improving their mechanical, technological and service properties. By such processes the authors mean any chemical or physical methods of influencing the structure and properties of metals or alloys, namely addition of modifiers; application of different physical impacts (vibration, ultrasonic or electromagnetic stirring) or a combination of the above.

Unlike modification, microalloying is usually understood as a method of adding individual elements or their compounds with not higher than 0.1 % residual content, which make a significant impact on the processes running in the solid phase (on phase composition of the alloys, size of secondary austenitic grains, structure

and purity of the boundaries and near-boundary zones, etc.) [4]. In the opinion of the author of work [2], the «microalloying» term often includes the modification and deoxidation processes, differing by the mechanism of their impact on the structure and properties of steel and alloys. The role of small additives at microalloying is manifested predominantly as a result of their influence on the metal solid state (formation of interstitial or substitutional solid solution; sizes of secondary grains; dispersity, shape and distribution of nonmetallic inclusions; boundary structure and fine structure of the grains; lowering the negative impact of impurities, etc.).

Thus, the «modification» term should be understood as a totality of any chemical, physical or complex processes influencing the final structure and properties of the metal (alloy). The term «microalloying» means the method of influencing the structure and properties of the metal (alloy) only due to adding to its composition small amounts ($\leq 0.1\%$) of elements or their compounds, which have a considerable influence on the processes running in the solid phase.

Classification of the modification methods. The methods of modification of the deposited metal can be divided into three main groups: chemical (adding modifiers); physical (application of external physical impact) and combined ones.

1. Chemical methods of modification. In this case a certain amount of chemical elements or their compounds are added to the deposited metal to achieve favourable changes of the metal structure and properties. Depending on their impact, the modifiers can be divided into four classes [5, 6]:

- modifiers of the 1st kind that improve wettability of one component of the alloy by another one, i.e. which lower the surface tension on their interface, and thus facilitate formation of the solid phase contacting the liquid one;

- modifiers of the 2nd kind that are the direct crystallization centers. They, however, can be so rather conditionally in the case when the melt temperature is so close to that of modifier solidification that it will be insufficient for melting of the modifiers added to the pool;

- modifiers of the 3rd kind (inoculators) that change the structure due to reduction of overheating of the crystallizing liquid metal. Higher cooling rate promotes increase of the crystallization rate and reduction of development of the liquation processes that is favorable for the structure;

- complex modifiers, which are a complex of two and more modifiers from one or different element classes from those given above. Depending on their type, the impact of such modifiers is usually manifested in a more intensive and complex manner, simultaneously affecting the mechanical, technological and service properties of the metal.

It should be also noted that the nature of action of modifiers of the 1st kind can change at formation

of modifier chemical compounds with other elements. The new chemical compound will in the long run have the role of an independent modifier already of the 2nd kind. These compounds can be surface-active under some conditions, and under other conditions they can be inactive, contrarily. So, for instance, boron, which usually is a modifier of the 1st kind, can form a more stable chemical compound with iron — FeB_2 , which is the crystallization center as a modifier of the 2nd kind [5]. The names of some chemical elements, or their compounds, which are often used for modification and microalloying, as well as their main physical properties are given in the Table [7, 8].

1.1. Modifiers of the 1st kind. Modifiers of such a type include additives, unlimitedly soluble in the liquid phase and slightly soluble in the solid phase (0.001–0.1 %). In their turn, these additives can be divided into two types: those, which do not change the surface properties of the crystallizing phase — «a», and those, which change the surface tension on melt-crystal interface — «b». Soluble impurities of type «a» can slow down the growth of the solid phase only due to the concentration barrier on the crystal-melt interface with no change taking place in the process energy characteristics [5].

Additives of «b» type are usually called surface-active. They selectively concentrate on the surface of

Physical properties of modifying chemical elements (compounds) [7, 8]

Modifier	Melting temperature, K	Boiling temperature, K	Density, g/cm ³
Mg	923	1363	1.738
Ba	1002	2170	3.500
Sr	1042	1657	2.540
Ce	1071	3740	6.757
Ca	1115	1757	1.550
La	1194	3730	6.150
Pr	1204	3785	6.773
Y	1795	3611	4.470
SiO_2	1983	2503	2.650
Zr	2125	4650	6.506
Al_2O_3	2345	3250	3.950
B	2349	4200	2.340
SiC	3003	3103	3.210
VC	3083	4173	5.770
WC	3143	6273	15.630
TiN	3203	N/D	5.400
ZrN	3253	Same	7.090
TiCN	3400	»	5.120
TiC	3533	4573	4.930
NbC	3763	4573	7.820
ZrC	3805	5373	6.730

crystals (dendrites). The surface-active substances can create a continuous adsorption layer. It means that with practical absence of solubility of the surface-active modifiers in the solid phase, a shell of the liquid enriched in modifiers forms around it [5].

Addition of modifiers of the 1st kind is accompanied by the change of surface tension and activation energy in opposite directions. It complicates their overall impact on crystallization and grain size. Macrograin refinement is the most characteristic effect for modifiers of the first kind. As increase of activation energy through additive adsorption on crystal faces promotes lowering of crystal growth rate, it causes coarsening of the grain dendrite structure. Thus, under the impact of modifiers of the 1st kind the macrograin is refined, but the micrograin is coarsened at the same time, i.e. an integrated impact on the macro- and microstructure is manifested [5].

1.2. Modifiers of the 2nd kind. Parameters of crystallization and macrostructure can be affected by hard particles added to the melt. Here, a number of researchers associate this influence exactly with the contact action on the process of nucleation of crystallization centers. It is accounted for by the fact that introduction to the melt of an insoluble additive with properties close to those of the crystallizing liquid, a significant lowering of the melt metastability range takes place. Such additives are called isomorphous with the crystallizing liquid, they have the crystalline lattice parameters close to those of this liquid, and it is believed that, similar to modifiers of the 1st kind, they ensure macrograin refinement. The content of this type of modifiers is usually limited to 0.1 % [5].

Proceeding from generalization of different works, the following conditions were formulated for selection of insoluble particles with the greatest modifying ability at surfacing [5]: use of refractory insoluble compounds; use of disperse particles with the large total interface; it is desirable for the particles to have metal properties (by the type of chemical bond); the most effective are particles of stable chemical compounds, which will form in the weld pool, as a result of interaction with one of the components or the alloy base.

1.3. Modifiers of the 3rd kind (inoculators). In order to increase the quality of the deposited metal at surfacing and welding, specially prepared powders of micro- and nanosized particles from refractory compounds of different elements are often used. When added to the weld pool, they are evenly distributed in the liquid metal volume and act as effective centers of metal pool crystallization. In keeping with the data of [5], addition of inoculants to the crystallizing melt, ensures an increase of homogeneity and dispersity of the cast structure, optimization of the shape and distribution of the nonmetallic inclusions, and reduction of the quantity of some defects that essentially increases the level and isotropy of the metal properties.

However, despite improvement of the macrostructure, application, for instance, of metal powder, cast shot and other similar additives as inoculators at electroslag surfacing can lead to greater contamination of steel by nonmetallic inclusions, mainly oxides. This is related to insufficiently mastered technology of introducing the inoculators, complexity of producing and storing them, as they require protection from oxidation during storage, transportation and addition to the liquid pool [5].

1.4. Complex modifiers. The main advantage of application of complex additives-modifiers is the fact that simultaneous action of two and more modifiers enhances the effect obtained at application of one modifier. This is related to the above process of crystallization center nucleation on insoluble impurities in the layer of the liquid phase with diffusion overcooling, due to addition of a soluble additive (especially surface-active) [5]. There are three types of complex modifiers: refining; strengthening and refining-strengthening. A feature of steel modification by complex modifiers is the fact that parallel to structure refinement, the nature and shape of nonmetallic inclusions also changes, level of grain boundary contamination by nonmetallic inclusions decreases, uniformity of distribution of structural components becomes higher, etc. [5].

2. Physical methods of modification. The main methods applied at surfacing, which can be regarded as this type of modification, and are aimed at metal structure refinement, are rather well known and covered in technical literature. This is, in particular, application of vibrations to the product during surfacing; input of energy from a pulsed heat source; pulsed feed of electrode or filler wire; application of sources with welding current modulation, and in a number of cases addition of separate heat sources which affect the HAZ near the line of the weld (deposited bead); external electromagnetic impact, etc. It is not rational to conduct a detailed review of each of the above-mentioned methods in this paper, so we will briefly focus on several processes, which allow making a rather significant influence on the structural state of steels and alloys and on their properties, respectively, and which can be used at surfacing.

2.1. Low-frequency vibrations and mechanical stirring of the liquid pool. Vibration is a method of forced mixing of the liquid pool. Vibration frequency usually is 1–30 Hz at variable amplitude, resulting in breaking up of large dendrites during solidification. Their fragments promote nucleation of new crystallization centers, leading to microstructure refinement and intensification of the processes of liquid pool degassing [9, 10].

2.2. Electromagnetic stirring. This method first became applied at continuous pouring of steel ingots, and then it began to be used in welding and surfacing. This method ensures breaking up of dendrites on the crystallization front and distribution of their fragments over the entire liquid pool volume, leads to lowering

and leveling of temperature in the pool volume and widening of the region of overcooled liquid metal. The number of crystallization centers grows in direct dependence on power, applied to the inductor, and, hence, on the speed of liquid phase movement [11, 12].

One of the examples of application of electromagnetic stirring is use of current-conducting mould (CCM) at electroslag surfacing [13]. In this case, slag pool rotation in the horizontal plane takes place in the mould, which is due to interaction of magnetic fields of the slag pool and the current-conducting section of the mould. Slag pool rotation leads to that the filler material particles fed into the slag pool, melt completely and the metal is better refined. In addition, pool rotation ensures a more fine-grained structure of the deposited metal.

2.3. Ultrasonic treatment. This method envisages an impact of high-frequency mechanical vibrations on the liquid pool. An alternating pressure is applied to the liquid, resulting in its rupture with formation of fine voids – ultrasonic cavitation. It may lead to breaking up of primary dendrites and phase components. Its fragmentation and even distribution of the fragments create additional crystallization centers and lead to macrograin refinement. The grain size decreases with increase of power applied to the pool, particularly after the cavitation threshold has been reached [14]. By some data [5], unlike electromagnetic stirring, at ultrasonic treatment not only volume crystallization and the associated undesirable phenomena do not develop, but, contrarily, the size of dendrite clusters decreases with grain refinement, as a result of shortening of the transition zone.

3. Combined modification methods. In this case, chemical additives-modifiers are applied simultaneously with physical impact and technological measures. It leads to enhancement of the effect of the modifiers and enables producing superfine and special structures. For instance, at addition of surface-active modifiers and application of ultrasonic treatment, the latter affects the nucleation energy in such a way that the modifying additive more is uniformly adsorbed on the nuclei of subcritical size and improves their resistance [15]. Selection of a rational method of physical impact and of the modifier in each specific case should be performed, proceeding from the objective of physical impact (increase of mechanical, technological, and service properties of the manufactured end products), as well as practical realization and cost-effectiveness of the method of impact.

Technologies of adding modifiers at chemical modification processes. At present exactly chemical methods of modification of the deposited metal and weld structure are quite widely used for surfacing and welding processes by adding powders of refractory chemical compounds to the weld pool. It ensures refinement of the metal structure and formation of new strengthening phases that promotes improvement of the metal service properties [16].

There exist a rather large number of methods of modifier addition to the weld pool: through electrode or filler flux-cored wires and strips; electrode coating; solid or flux-cored filler rods and wires, which are placed directly into the gap in welding; ceramic flux, etc.

Here, a partially unsolved problem still is preservation of ultradisperse particles in the weld pool, which actively dissociate during surfacing, as addition of modifiers directly into the weld pool through the flux or electrode wire is less effective than at casting, because of high temperature in the welding zone [17]. It is already widely known that the temperature in the arc column, through which the molten metal drop passes, may reach 5000–6000 K, the temperature of the drop proper reaching 2300–2700 K. Therefore, one of the methods to increase the effectiveness of deposited metal modification at arc surfacing is application of elements and compounds having a high melting temperature (2700 K) as modifiers.

Let us consider the main known methods of adding modifiers to the deposited metal, as well as their advantages and disadvantages.

1. Addition of modifiers contained in flux-cored wire charge, directly through the arc. Despite the above-mentioned disadvantages of the process of metal modification at passage of modifier additives directly through the welding arc, such a method is quite often applied, due to its relative simplicity and availability.

So, in [18] a complex master alloy was developed, which contains yttrium, cerium and rare-earth elements, and which was successfully applied in the charge of self-shielding flux-cored wires PP-AN155 and PP-AN156 for surfacing tools for hot deformation of metal (different forming and mill rolls, hot cutters, etc.) and flux-cored wire PP-AN163 for submerged-arc surfacing of propeller shafts and other parts. As shown by production testing, modification and microalloying of the deposited metal allowed increasing the tool resistance by 20–30 %.

In work [19] metal modification to improve its wear resistance was performed with success by adding potassium fluorozirconate to flux-cored wire charge. Steel modification by zirconium by adding to electrode wire composition powders of ferroalloy of Fe–Si–Zr alloying system was also applied with success in [20].

In the opinion of the authors of [21], when boron is used as modifier, its addition to the charge of flux-cored electrode wires looks the most promising, as application of solid wire requires increase of its boron and titanium content, which results in increase of the wire hardness and rigidity. Addition of boron microadditives to the deposited metal through the flux seems to be insufficiently reliable, because of low metallurgical activity of boron.

In works [22, 23] deposited metal modification was performed by adding to the charge flux-cored

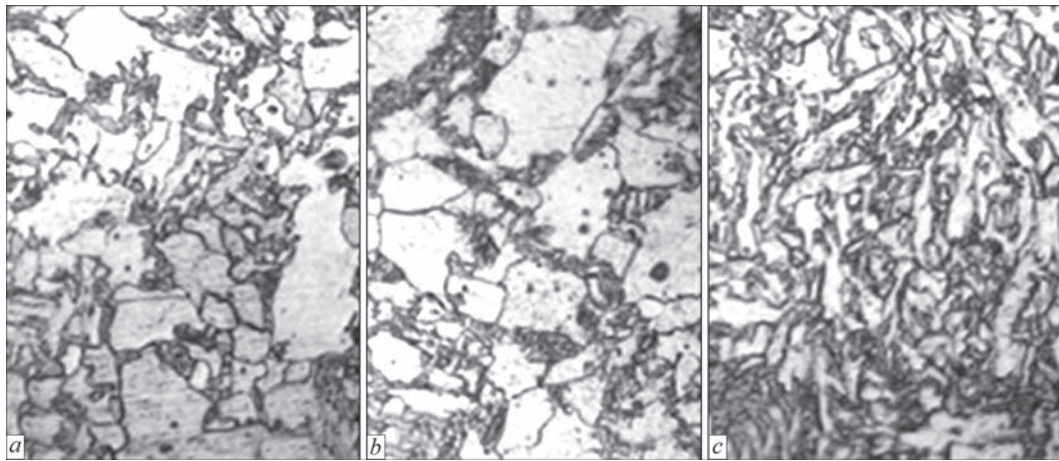


Figure 1. Microstructure ($\times 400$) of the metal of welds made [17]: *a* — without modifier addition; *b* — with TiO_2 addition, $d_{av} = 8 \mu\text{m}$; *c* — with TiO_2 addition, $d_{av} = 4 \mu\text{m}$

electrode wires of titanium and tungsten carbide and boron nitride powders. In these cases, so-called microcoolers were used (see below), in order to enhance the modification effect.

As we can see, despite the negative influence of high temperatures in the arc gap, the method of modification by introducing modifiers into the charge of flux-cored electrode wires is quite successfully used. By the data of [24], the sizes of disperse particles of the refractory compounds should be equal to 50–300 μm , in order to achieve a more tangible effect from addition of modifiers to the weld pool through the flux-cored wire charge. More over, in the opinion of the authors of [25], in order to guarantee the transition of modifier components from electrode materials into the deposited metal, such processes should be used, in which a low-power and short-term thermal cycle is realized and a weld of a small volume forms.

2. Application of «microcoolers». In order to reduce overheating and melting of modifying additives, penetrating into the arc zone, in work [22] it was proposed to apply the additives-modifiers in a mixture with cold macroparticles (microcoolers) from wire chips of $2 \times 2 \text{ mm}$ size, which are fed into the mixture with modifiers in the form of filler material.

The same idea was realized in several works at nano-modification of welds [6], and deposited metal [26]. In order to preserve the activity of modifying nanoparticles and to ensure their attachment to macroparticles that have the role of microcoolers, mechanochemical treatment of the powder mixture of macro- and nanoparticles was performed in high-energy planetary mills. In particular, in work [26] nickel-based composite microgranules, containing nanoparticles (up to 100 nm size) of titanium carbonitride, were added to the charge of experimental flux-cored wires as a modifier.

More over, in keeping with the data of works [3, 17], reduction of the dimensions of additives-modifiers to the nanoscale allows improving their effectiveness. The smaller the sizes of the modifying particles,

and, hence, of the crystallization centers, the smaller is the weld metal grain, and the smaller are the stress intensity factors. Refinement of modifier particles increases the specific energy of the colliding particles and the probability of chemical interaction between them with formation of strong ties and more effective refinement of the metal structure (Figure 1).

3. Application of additional electrode or filler wire. The effectiveness of metal modification can be increased by introducing modifying additives to the low-temperature zone of the pool in the composition of additional electrode or filler flux-cored wire. Here, it is rational to use one flux-cored wire in multiarc welding (surfacing) process to change the produced joint quality characteristics [27].

So, in [16] it was demonstrated that metal modification was quite successfully performed using flux-cored filler wire, which consisted of low-carbon steel sheath and filler, containing granulated powder from nano- and microparticles of titanium nitride.

In work [28] a comparison was made of the results of titanium modification at two schemes of its addition in submerged-arc welding: standard, through the flux-cored wire charge through the arc column and with direct introduction of additional filler wire into the «cold» part of the weld pool. Investigations showed that as titanium is an effective deoxidizer, the probability of its oxidation is the highest in the arc column and in the drop, as well as in slag deoxidation reactions. Therefore, at titanium addition directly through the charge of flux-cored electrode wire a reduction of its content in the weld by 3.5 times was noted compared its content in the flux-cored wire. However, at application of wire of identical composition, but as a filler, titanium content in the weld decreased only by 1.3 times.

At the same time, application of additional filler wires and strips for deposited metal modification can be complicated by a number of factors. Introduction of electrode wires in front of the main electrode, as well as at a small distance from it, similar to addition-

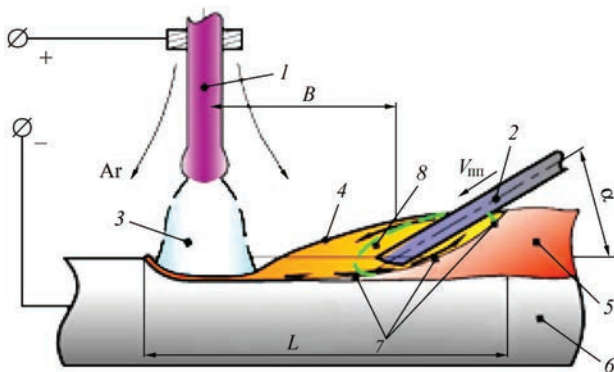


Figure 2. Scheme of adding modified flux-cored filler wire to the weld pool during electric arc surfacing [16]: 1, 2 — electrode and filler flux-cored wires; 3 — electric arc; 4 — weld pool; 5, 6 — deposited and base metals; 7 — deposited metal solidification front; 8 — overcooled zone of metal melt

al heating of the welding wire by current, still leads to considerable losses of the modifier, because of its melting and dissolution in the overheated melt. Application of electroneutral filler wire significantly limits its feed rate, exceeding which causes wire welding to the pool bottom, while application of several wires leads to pool overcooling and violation of the quality of deposited metal layer formation (Figure 2) [16].

In order to solve this problem, a comprehensive experimental study was performed in work [16], to determine the optimal parameters of introducing additional filler modifying wire into the weld pool: angle of inclination, feed rate, distance from electrode wire to filler wire edge, etc. Such an approach, however, requires considerable time and costs and needs to be improved.

Thus, the method of deposited metal modification through additional filler wire, fed into the weld pool tail part, allows increasing the content of modifiers in the deposited metal. At the same time, this method is complicated in terms of technology, involving considerable difficulties of ensuring a stable arc process and high quality of deposited metal formation and homogeneity of its structure.

4. Application of additional fillers, placed directly on the workpiece, being welded or surfaced. A method of laying filler rods or wires directly into the groove at welding is sometimes used to reduce the modifier losses. So, in [29, 30] cast rods or consumable electrodes of a specified length and diameter were used for weld modification, which were placed into the groove along the butt length directly before welding.

This method was upgraded in work [31], and a two-arc welding process was used for melting the modified master alloy and additional mixing of the weld pool. Such a schematic allows avoiding direct impact of the arc on modifying additives and shields them by a liquid interlayer (Figure 3).

In work [32] modifying additives (disperse powders of silicon carbide and aerosil) were used, which were located in a layer of alloying charge, applied di-

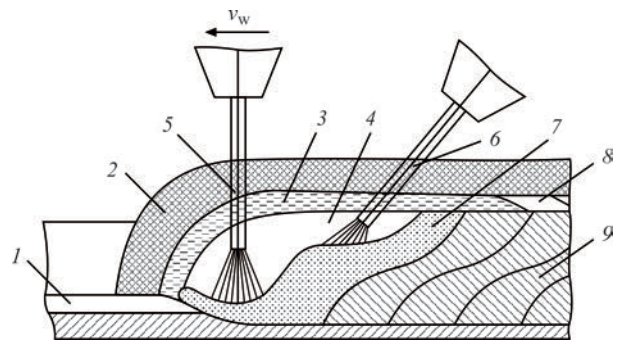


Figure 3. Scheme of two-wire submerged-arc welding with application of modified master alloy [31]: 1 — master alloy; 2 — flux; 3 — liquid slag; 4 — gas bubble; 5, 6 — electrode wires; 7 — liquid pool; 8 — solidified slag; 9 — weld metal

rectly on the processed surface. However, this method has a significant disadvantage: at its application sound formation of the deposited metal can be achieved only in a narrow range of surfacing modes. In order to solve this problem, the authors of [33] suggested additional superposition of an external magnetic field. Such an approach, however, complicates the surfacing process even more in terms of technology.

As we can see, the above-listed deposition methods allow in a certain way avoiding direct impact of the welding arc on modified filler wires or rods. They, however, are technologically quite complicated, and often cannot be applied for surfacing.

Thus, the simplest and most rational of the above-described methods is addition of modifiers directly through the charge of flux-cored electrode wires. This method is quite versatile and can be used with certain modifications at different electric arc, electroslag, plasma and other surfacing processes.

Conclusions

1. Distinction between the terms «modification» and «microalloying» should be made. Modification is a set of any chemical or physical processes influencing the final structure and properties of metal (alloy), while microalloying is a method of influencing the structure and properties of the metal (alloy) only due to introducing into its composition small additives ($\leq 0.1\%$) of elements or their compounds, having a significant impact on the processes running in the solid phase.

2. Modification and microalloying of steels and alloys are mostly used for refining their micro- and macrostructure; reduction of chemical, physical and structural inhomogeneity; favourable changes in the nature and shape of nonmetallic inclusions; enhancement of the set of technological, mechanical and service properties of steels.

3. In terms of technology and economy, the simplest and most rational method of metal modification at surfacing is addition of modifiers directly through the charge of flux-cored electrode wires. In order to increase the modification effectiveness in this case, it

is recommended to use modifiers with melting temperature above 2700 K and (or) additionally use special particles-microcoolers.

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