INFLUENCE OF NON-METALLIC INCLUSIONS ON THE CORROSION RESISTANCE OF STAINLESS STEELS IN ARC SURFACING

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The influence of non-metallic inclusions on the corrosion resistance of stainless steel of the austenitic-martensitic class, deposited in one layer with flux-cored wire under the flux by the arc method in the mode of stationary and pulsed arc burning, has been studied. In the latter case, the wire feed impulse was 0.5 and 0.7 Hz. The selected surfacing conditions, with both methods of the process, contribute to the formation of non-metallic inclusions of complex composition and structure in the deposited metal, including those containing calcium and its compounds with other elements, which are classified as corrosive, accelerating the occurrence of corrosion phenomena.

KEYWORDS

arc surfacing, stationary and pulsed surfacing, stainless steels, non-metallic inclusions, corrosion resistance

1 INTRODUCTION

In recent years, surface engineering has gained more importance in the field of materials science, especially in the aerospace, automotive, biomedical, and engineering fields [Mikhalev 2019, Pogrebjak 2016, Dyadyura 2017, Sukhodub 2019]. It is estimated [Prislupcak 2014] that material degradation due to wear and corrosion is very high. Surface treatment can improve the performance of materials at an affordable cost to bring about functional properties that would otherwise not be provided by the substrate material. Since surface engineering [Berndt 2020] can improve the performance of these materials at a lower cost, it has tremendous potential to provide technological solutions to various engineering problems.

Surfacing with hard wear-resistant materials [Kartsev 2020] is intended to increase the wear resistance of sealing, rubbing, and other surfaces of parts operating under conditions of metal-to-metal friction in corrosive media (acid, alkali, salt, and organic solvent) [Panda 2014, 2018a,b, 2019; Valicek 2016 & 2017, Macala 2009 & 2017, Pandova 2018, Krenicky 2012], high and low temperatures, and/or pressure [Bozek 2021]. The research [Sadeghi 2019] includes a substantive discussion of high-temperature corrosion mechanisms, new coating formulations, and a brief comparison of corrosion-resistant coatings obtained using various thermal surfacing techniques. It is suggested that with a careful and reasonable choice of technique and technology of surfacing, as well as compliance with a certain temperature range of pre-and associated heating of parts, it will be possible to obtain a high-quality connection of wear-resistant layers of alloys based on iron, nickel or cobalt [Miglierini 2004, Babinets 2020, Pollak 2020a,b]. Scientific researches [Zhu 2018, Pereplotchikov 2018] suggests that the best indicators of wear resistance are achieved due to the formation of microstructures in the surfacing material that corresponds to the Charpy principle: individual solid inclusions with a low coefficient of friction and a low tendency to scuffing are located in a plastic matrix.

The characteristics of the surface layer deposited with flux-cored wire are highly dependent on the flux. The research [Sharma 2019] is studied the physicochemical and thermophysical behavior of submerged arc surfacing slags developed by the $CaO-SiO_2-CaF_2$ and $CaO-SiO_2-Al_2O_3$ systems. Slag was obtained by submerged arc surfacing using agglomerated fluxes prepared in the laboratory. The research [Sharma 2020] is studied the thermophysical properties of twenty-one submerged arc surfacing slag. Submerged arc surfacing slags were analyzed using the disk and thermogravimetric methods. Non-metallic inclusions play an important role in determining the properties of the surface [Abdulsalam 2018]. Difficulties arise both with elements that are practically insoluble in steels (O, Ca, Mg, S) and with those whose content in the dissolved form is usually high (Si, Mn). The work [Bock 2019] provides a review of the literature on the determination of nonmetallic inclusions in metal alloys by atomic emission spectroscopy with single spark recording of spectra.

In the study [Kychkin 2019], the role of local austenite zones in the near-surface layers of austenite-martensitic metal of deposited beads on the development of corrosion processes along the boundaries of austenite grains was shown. Moreover, this effect was observed both in conventional methods of arc surfacing with flux-cored wire and in pulsed-arc surfacing, although in the latter case, the metal is formed with less structural micro-inhomogeneity. Many authors [Dwivedi 2018, Malushin 2020, Manjili 2018] believe that the reason for such a selective course of corrosion processes is the existing difference in the physical structure and chemical composition of the grain body and grain boundaries, on which secondary phases can precipitate, the accumulation of impurities, various types of non-metallic inclusions, etc. This physicochemical inhomogeneity, in turn, leads to the appearance of electrochemical inhomogeneity of the structure, when the boundaries or near-boundary zones of crystallites are more electrochemically negative in comparison with the body of the grain itself. As a result of the anodic process, the metal atoms are transferred to the solution in the form of hydrated ions.

This article is devoted to a more detailed study of this phenomenon, in particular, the effect of non-metallic inclusions on the corrosion resistance of the deposited metal.

The influence of non-metallic inclusions on the corrosion resistance of steels has long attracted the attention of researchers dealing with the problem of increasing the corrosion resistance of various structures, as well as parts and assemblies of machines and mechanisms operating in corrosive environments [Liu 2017, Webler 2020].

The least resistant areas of metal surfaces are, as a rule, areas directly adjacent to the NI. The activity of the metal in the places of its contact with non-metallic inclusions can be due to various reasons, such as: increased defectiveness of the crystal

lattice of the metal near the inclusion, depletion of this layer in corrosion-resistant alloying components of the alloy, activating the effect of the dissolution products of the NI itself, the presence of a gap between the inclusions and the metal. The most dangerous inclusions are complex oxysulfides [Lyu 2019, Prajapati 2017]. The authors of [Holly 2019, Sadenova 2019, Kazasidis 2021] note that not only the general level of contamination of the metal but also the chemical composition, shape, size, and distribution of inclusions have a significant effect on the corrosion properties of steel. At the same time, without denying the negative effect on the corrosion processes of oxysulfides, especially of large sizes, it is nevertheless considered that the most dangerous are inclusions of FeS and MnS. These inclusions, having a coefficient of linear expansion greater than that of the matrix, form a microvoid at the metal-inclusion boundary when the steel is cooled after solidification of the weld pool metal. While, silicates, oxides, and oxysulfides, which have low coefficients of linear expansion, remain in close contact with the matrix upon cooling. For this reason, sulfide inclusions, which, as a rule, have a line-like shape, are often the centers of destruction. In addition, in corrosive environments, sulfides play the role of microcathode regions leading to local destruction of the metal.

2 MATERIALS AND METHODS

2.1 Materials

Austenitic-martensitic steel (table 1) was selected as the material under study, deposited in one layer with 50% overlap of adjacent beads by arc surfacing under flux (table 2) with flux-cored wire (table 3).

Table 1. Chemical composition of austenitic-martensitic steel, %

C	Cr	Mn	Si	Ti	Ni	S	P	Cu	Fe
0.4	18	0.1	0.5	0.5	11	0.02	0.03	0.2	68

The chemical composition of welding flux for arc surfacing is given in table 2.

Table 2. Chemical composition of surfacing flux, %

C	MnO	CaO	MgO	Al_2O_3	CaF_2	SiO_2	Fe_2O_3	S	P
0.03	3.5	6	17	22	22	30	1.2	0.07	0.07

The chemical composition of powder for arc surfacing is given in table 3.

Table 3. Chemical composition of flux-cored wire, %

C	Cr	Mn	Si	Mo	Ni	S	P
0.4	20	0.5	0.6	1.5	0.8	0.02	0.02

The structure of the flux grains is pumiceous. Grain size, mm − 0.25−2.80. Bulk density, g/cm^3 − 0.8−1.1. Calcination mode before welding: (500-600) °C/2 hours.

For comparison, surfacing was also performed with the same wire in a pulsed-arc mode with a wire feed pulse during surfacing of 0.5 Hz (tim. = 1, tp = 1s) and 0.7 Hz (tim. = 0.7, tp = 0.7s). The metal obtained by surfacing with this wire has a martensitic-ferritic structure of metal and hardness HRC 42 ... 45.

2.2 Methodology of research

The size, shape and chemical composition of the remaining non-metallic inclusions have been investigated. Samples were prepared in accordance with the Standard Guide for Sample Preparation and Evaluation for the Automatic Steel Inclusion Evaluation Method (ASTM E 768-99), and the results were presented using standard test methods for determining the content of inclusions in steel (ASTM E 45-97).

For employed the properties samples to investigate the electron microscope (PEM-125K), scanning electron microscopy (SEO-SEM Jamp 9500F), electron-dispersive X-ray spectroscopy (X-MaxN20).

3 RESULTS AND DISCUSSION

Differences in composition and layers structure of the surfaced were studied. The least resistant areas of metal surfaces are, as a rule, areas directly adjacent to the NI. The activity of the metal in the places of its contact with non-metallic inclusions can be due to various reasons, such as: increased defectiveness of the crystal lattice of the metal near the inclusion, depletion of this layer in corrosion-resistant alloying components of the alloy, activating the effect of the dissolution products of the NI itself, the presence of a gap between the inclusions and the metal. The most dangerous inclusions are complex oxysulfides. The note that not only the general level of contamination of the metal but also the chemical composition, shape, size, and distribution of inclusions have a significant effect on the corrosion properties of steel.

At the same time, without denying the negative effect on the corrosion processes of oxysulfides, especially of large sizes, it is nevertheless considered that the most dangerous are inclusions of FeS and MnS. These inclusions, having a coefficient of linear expansion greater than that of the matrix, form a microvoid at the metal-inclusion boundary when the steel is cooled after solidification of the weld pool metal.

While, silicates, oxides, and oxysulfides, which have low coefficients of linear expansion, remain in close contact with the matrix upon cooling. For this reason, sulfide inclusions, which, as a rule, have a line-like shape, are often the centers of destruction. In addition, in corrosive environments, sulfides play the role of microcathode regions leading to local destruction of the metal.

The type of sulfide inclusions in steels is characterized by Gibbs free energy (ΔG) of their formation. In decreasing order of absolute values of Δ G, sulfides that may be present in steels are located in the following row: CaS>TiS>MnS>CrS>NiS>FeS (table 4).

Table 4. Standard Gibbs free energy of formation for some sulfides

Sulfide-forming element	Sulfide	- Δ G, kJ/mol
Fe	FeS	80-97,5
Cr	CrS	150-240
Ni	NiS	114
Mn	MnS	192-209
Mn	(Mn 0,9 Fe 0,1)S	184,8
Ca	CaS	476
Ti	TiS	More negative than MnS

According to Table 4, calcium sulfides are the most corrosive. However, their formation is possible only in carbon or low-alloy steels, smelted according to a certain technology. Most of the stainless steels produced are characterized by inclusions of manganese sulfides. Nevertheless, in the same works, one more fact is noted.

The main factor that determines the abnormally high corrosion rate of steel, regardless of its grade, is the presence of certain non-metallic inclusions in it, which are commonly called corrosive non-metallic inclusions (CNMI). These inclusions have a complex composition, and, as a rule, contain calcium.

As applied to oil pipelines, in the general case, a negative effect on the corrosion resistance of CNMI of two types has been established. Type 1 contains small (~3 ... 5 μm) non-metallic inclusions based on calcium aluminate, sometimes with additions of magnesium, silicon, and some other elements. Type 2 is characterized by the presence of complex inclusions with a core of calcium aluminate (with a different ratio of CaO and Al_2O_3), manganese sulfide, or another element, surrounded by a calcium sulfide shell.

It should be noted that when surfacing stainless steels in contrast to their smelting, small amounts of Ca can pass from the flux and charge of flux-cored wire (as well as side components charge in the case of grinding some ferroalloys, in particular, ferromanganese with the addition of finely ground calcium-containing inert material - marble, dolomite, etc. in the amount of 5-8%) into the metal of the weld pool. Thus, complex inclusions containing calcium can also form in it (at the stage of solidification of the deposited metal), which negatively affects the corrosion resistance of the stainless metal, as in carbon and low-alloy steels. A qualitative assessment of the rifling characteristics of the surfacing metal structure showed the presence of non-metallic inclusions of a round shape along the boundaries of the austenite phase (Fig. 1).

The elemental composition of these inclusions is indicated in the tables (Figure 1, a, b, c). The chemical composition of the observed inclusions is variable. As expected, in the composition of many non-metallic inclusions', there is Ca in addition to Mn, Si, Al. Analysis of Fig. 1 indicates that the Ca content in them can reach ~ 25%. They can be classified as inclusions of complex composition, which is confirmed by the results of elemental analysis (Table 1, a, b, c). The non-metallic inclusions' negative effect of the course of corrosion processes is clearly seen from Fig. 2, which shows the onset of corrosion in the local near-surface austenitic zone and its further development grain boundaries of the metal, passing through individual or several non-metallic inclusions. The process last stage is especially clearly demonstrated in Fig. 3, where the grain boundary with traces of corrosion forms, as it were, a loop (Fig. 3a) or a half-loop (Fig. 3b), passing through several non-metallic inclusions.

In connection with the presence of calcium-containing inclusions in the deposited metal (austenitic-martensitic stainless steel), apparently, of the first type, their influence on the course of corrosion processes should be considered in more detail. Moreover, they have recently received much attention in connection with pipelines' long-term operation for various purposes, made of carbon and low-alloy steels.

It should also be noted that the tendency of steels to local corrosion does not always correlate with the amount of non-metallic inclusions, estimated by the standard method. Steel can be very pure in terms of NI but subject to localized corrosion. As mentioned earlier, this is largely determined by the presence of calcium-containing inclusions, which are difficult to identify. Moreover, aluminate non-metallic inclusions are outwardly indistinguishable from non-metallic inclusions', which are always present in steels, for example, oxides of the same dimensions.

It should be noted that the mechanisms of the influence of the corrosive non-metallic inclusions on the acceleration of corrosion processes have not yet been fully disclosed and are the subject of research and discussion. There is an opinion that the corrosiveness of the first type corrosive non-metallic inclusions is associated with an increased level of internal stresses arising from the difference in the temperature coefficients of linear expansion of the inclusion and the surrounding matrix. According to calculations, the stress arising

at the boundary between the corrosive non-metallic inclusions of the first type and the matrix is quite significant and can exceed steel's yield strength. Thus, in a very narrow (several micrometers) region around the first-type corrosive non-metallic inclusions, significant elastic tensile stresses can appear, or the matrix's plastic deformation can occur [Mascenik 2020]. All these are factors that contribute to the preferential local dissolution of the metal.

Simultaneously, based on the preliminary results of the studies of steel samples made of corrosive non-metallic inclusions of the first type carried out at present, the zone around the inclusion may have a chemical composition that differs from the rest of the matrix particular, in the oxygen content.
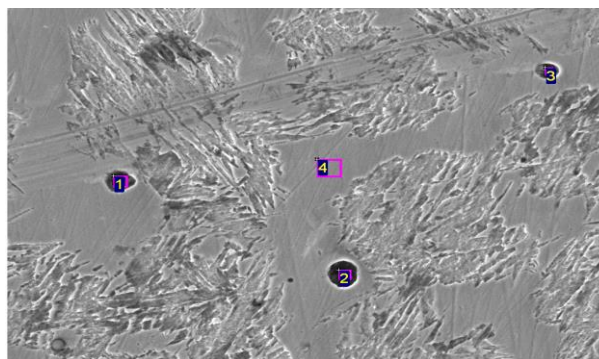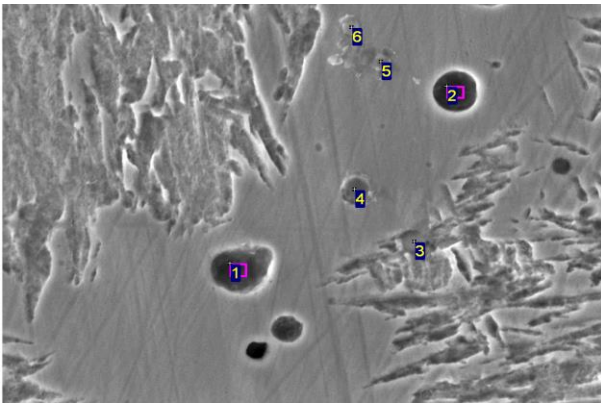

Spectrum	N	O	Al	Si	P	Ca	Ti	Cr	Mn	Fe	Mo
1	1.93	7.07	0.17	0.57	0.33	5.91	0.64	10.09	1.11	basic	2.74
2	3.06	9.74	1.45	0.54	0.29	5.91	0.43	8.34	0.12	basic	1.64
3	0.35	5.09	1.90	0.49	0.17	1.07	0.08	17.29	1.10	basic	1.06
4	0.00	0.55	0.04	0.74	0.05	0.00	0.00	16.88	0.99	basic	0.95
Average	1.34	5.61	0.89	0.58	0.21	3.22	0.29	13.15	0.83	basic	1.60
Std. deviation	1.43	3.88	0.93	0.11	0.13	3.13	0.30	4.60	0.48		0.82
Max.	3.06	9.74	1.90	0.74	0.33	5.91	0.64	17.29	1.11		2.74
Min.	0.00	0.55	0.04	0.49	0.05	0.00	0.00	8.34	0.12		0.95

a)
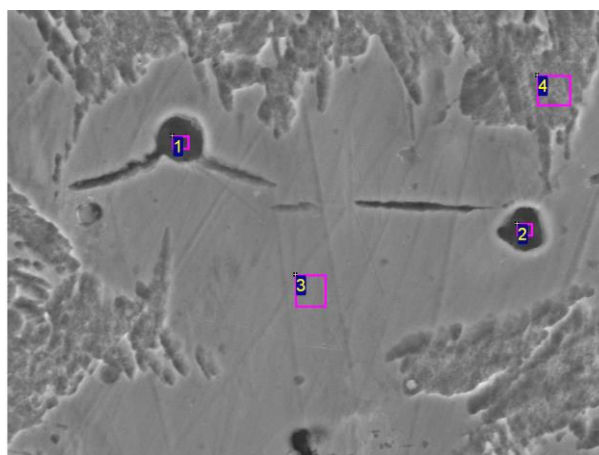
Spectrum	N	O	Na	Al	Si	Ca	Cr	Mn	Fe	Mo
1	6.51	7.61	0.32	0.23	0.23	9.17	8.94	0.19	basic	1.38
2	6.81	23.59	0.25	0.21	0.68	24.61	5.93	0.00	basic	1.19
3	0.00	0.00	0.01	0.10	0.60	0.00	15.04	0.91	basic	0.29
4	0.13	0.88	0.00	0.02	0.55	0.00	12.88	0.92	basic	0.39
Average	3.36	8.02	0.14	0.14	0.52	8.45	10.70	0.50	basic	0.81
Std. deviation	3.81	10.92	0.16	0.10	0.20	11.61	4.06	0.48		0.55
Max.	6.81	23.59	0.32	0.23	0.68	24.61	15.04	0.92		1.38
Min.	0.00	0.00	0.00	0.02	0.23	0.00	5.93	0.00		0.29

b)

Spectrum	N	O	Al	Si	Ca	Ti	Cr	Mn	Fe	Mo
1	0.00	10.49	1.93	3.45	1.07	0.10	15.38	0.66	basic	1.55
2	2.28	3.28	0.07	0.39	1.17	0.00	17.08	1.15	basic	0.48
3	0.00	40.89	8.60	15.13	0.04	1.25	4.20	17.88	basic	1.39
4	0.00	32.88	6.90	12.64	0.19	1.03	8.26	18.08	basic	1.17
5	0.00	12.72	2.44	4.89	0.23	0.24	13.19	7.55	basic	2.03
6	0.00	6.51	1.43	2.71	0.26	0.39	13.42	3.46	basic	0.87
Average	0.38	17.79	3.56	6.54	0.49	0.50	11.92	8.13	basic	1.25
Std. deviation	0.93	15.35	3.38	5.93	0.49	0.52	4.81	8.01		0.54
Max.	2.28	40.89	8.60	15.13	1.17	1.25	17.08	18.08		2.03
Min.	0.00	3.28	0.07	0.39	0.04	0.00	4.20	0.66		0.48

c)

Figure 1. Non-metallic inclusions in the structure of metal deposited with flux-cored wire (zoom x1000)

Thus, the reason for the abnormal acceleration of corrosion processes in corrosive non-metallic inclusions' presence may not be physical but chemical in nature. Research on this issue is ongoing. The presence of calcium in non-metallic inclusions of the deposited metal poses the task of assessing their detection methods, the effect on the corrosion rate of stainless steels, and understanding the phenomena occurring in this case.
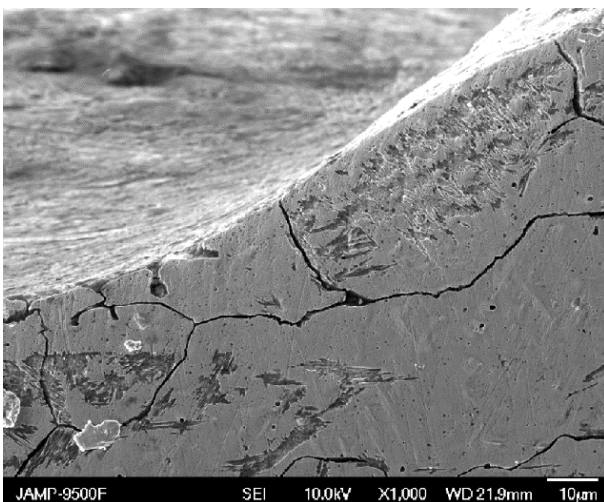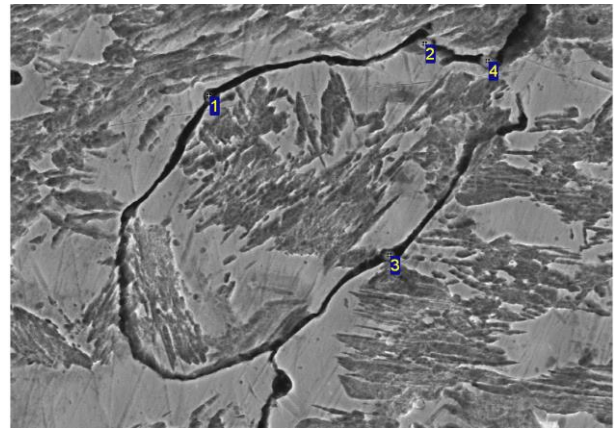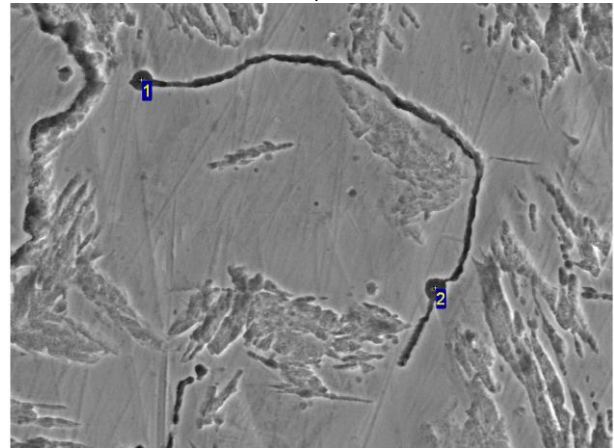

Figure 2. The beginning of corrosion in the local near-surface austenitic zone

a)

b)

Figure 3. Intergranular boundary with traces of corrosion, forming a "loop", passing through non-metallic inclusions 1-4 (a) or "half-loop", passing through non-metallic inclusions 1 and 2 (b) (zoom x1000)

4 CONCLUSIONS

It has been established that in contrast to cast stainless steels, steels deposited with flux-cored wires by arc method under a layer of flux contain non-metallic inclusions, calcium content, and its compounds with other elements. Calcium-containing non-metallic inclusions are deposited stainless steels that can be corrosive active, significantly accelerating the corrosion process, as it happens in cast carbon and low-alloy steels. It is necessary to carry out additional studies to assess the identification methods and the effect of calcium-containing non-metallic inclusions on deposited stainless steels' corrosion rate. For a more thorough consideration of the presence of chromium carbides along the grain boundaries and their possible influence on the course of corrosion processes, it seems that more precise methods of studying the microstructure with high resolution should be involved in the research. However, it can already be argued that such precipitates of carbides for this type of deposited steels are not all-encompassing.

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