

EFFECT OF SURFACE-ACTIVE ELEMENTS ON THE FORMATION OF SOLIDIFICATION CRACKS

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Based on analysis of physicochemical properties of harmful impurities, the concepts of the mechanism of their effect on formation of solidification cracks in different metals are summarized, which consist, firstly, in enrichment of crystallite boundaries with them (residual liquid) at the last stages of solidification, and, secondly, in revealing the effect of adsorption-induced decrease of strength and ductility. The difference in the nature of solidification of single-phase and two-phase (with eutectic) alloys was established as regards enrichment of crystallite boundaries with harmful impurities. It is shown that the specific surface energy at solid metal – melt interface and on grain boundaries has the greatest effect on crack formation. It was established that also the surface-active alloying elements, characterized by the same physicochemical properties as harmful impurities, can influence the increased tendency to solidification cracking. The formula for subcritical crack growth, proposed by E.E. Glikman et al., can serve as a criterion for evaluation of the effect of surface-active elements on the formation of solidification cracks. 36 Ref., 2 Tables, 9 Figures.

Keywords: *solidification cracks, harmful impurities, physicochemical properties, effect of adsorption-induced decrease of ductility and strength, distribution coefficient, enrichment of grain boundaries, state diagram, solidification nature, eutectic*

Solidification cracks are one of the most common defects in welding and casting of various metals and alloys. Generalized postulates of the current theory of solidification cracking (technological strength of metals) were discussed already in 1960s [1].

According to this theory technological strength of metals depends on:

- brittleness temperature range (BTR);
- deformability in BTR;
- deformation rate (intensity of elastoplastic deformation increase with temperature lowering), i.e. solidification cracking is affected by two factors: metallurgical (BTR and deformability in it) and force (deformation rate).

A necessary condition for solidification cracking is presence of solid and liquid phases in metal. However, there are still differences in the views on the amount and form of the liquid phase, leading to cracking. A fairly comprehensive review of this subject was made by V.V. Podgaetsky [2].

At the same time, there is no unambiguous opinion about the mechanism of the influence of intercrystalline liquid interlayers (residual liquid phase) on solidification cracking.

N.N. Prokhorov [3], as a result of theoretical analysis of the process of alloy deformation in the solid-liquid state came to the conclusion that deformability depends mainly on the sizes of crystallites and thickness of the liquid phase, i.e. it does not take into

account the physicochemical interaction of the solid and liquid phases. A number of authors (B.I. Medovar, V.V. Podgaetsky, I.I. Novikov, A.E. Runov, I.R. Patskevich) have suggested that the liquid phase can influence solidification cracking, as a result of manifestation of the effect of adsorption-induced lowering of ductility and strength (ALDS).

As noted by V.V. Podgaetsky [2], the influence of surface properties of the residual liquid on solidification cracking was taken into account by J. Borland in his generalized theory of hot cracking. He, however, makes the following comments on this theory [2]: «The statement of the significant influence of the area of contact between the crystallites on the possibility of cracking is questionable... in the presence of the liquid phase on the crystallite boundaries initiation of intercrystalline crack filled with the melt is more beneficial in terms of energy than in other regions, even at higher angle θ (as noted in the work by I.I. Novikov [5]). Conditions of initiation of cracks of sulphide origin are not consistent with the above theory, either. Even though the chemical composition of intercrystalline sulphide films differs greatly from that of the weld metal, this does not prevent solidification cracking». However, I.I. Novikov [5] believes that the change of the composition of commercial alloys by additional alloying should not change the brittleness, resulting from ALDS effect, as the value of surface energy on the boundary of the solid and liquid phases is so small in the majority of alloys that be the energy

Table 1. Some physical properties of the considered metals and impurities [10]

Physical properties	Alloying elements				Impurities			
	Fe	Ni	Cu	Al	P	SS	Bi	TNa
Surface tension, σ_m , MJ/cm ²	1840	1810	1350	914	69.7	42	90	91
Specific heat of sublimation P , MJ/cm ³	46.0	55.7	43.9	27.8	18.5	4.4	8.8	4.3
Melting temperature, K	1811	1728	1357	932	317.4 (for white) 870 (for red)	392	545	371

somewhat larger or smaller is practically of no importance for the mechanical properties of the alloy in the solid-liquid state. We believe, this is a mistaken opinion. Such a difference in the views is indicative of insufficient theoretical and experimental studies of the process of solidification crack formation.

Considering that for real structures the range of force factor regulation is small (structure rigidity is preset in most of the cases, and is determined by service requirements, and the ranges of welding mode parameter variation for specific products are also limited), the most effective impact on cracking can be produced by metallurgical methods. Therefore, let us consider the influence of metallurgical factor on cracking.

As is known, the influence of metallurgical factor on solidification cracking at a constant welding mode is determined mainly by chemical composition of the metal, and, primarily, presence of harmful impurities [2, 5–9]. These impurities are:

- sulphur, phosphorus in iron and its alloys;
- sulphur, phosphorus in nickel and its alloys;
- bismuth, lead, sulphur, phosphorus in copper and its alloys;
- sodium in aluminium and its alloys.

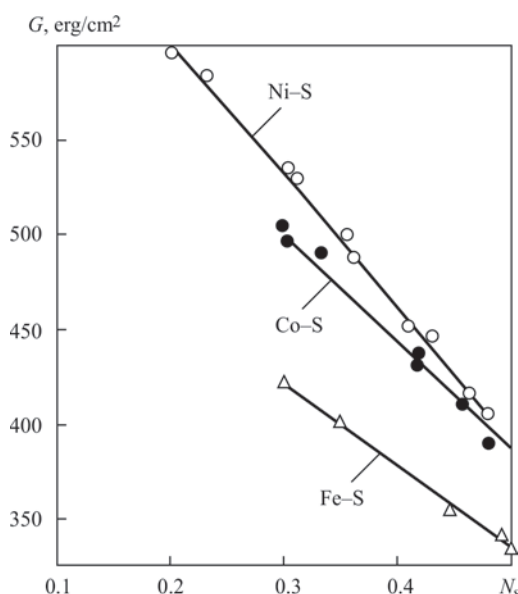


Figure 1. Sulphur effect on surface tension of iron, nickel and cobalt [11]

The above-mentioned elements, apparently, feature some common physicochemical properties, determining their similar specific influence on solidification cracking.

As is known, the physicochemical properties of impurities are largely determined by the state diagram of metal–impurity system. Analysis of these diagrams shows that all of them belong predominantly to the eutectic type with limited (low) solubility of the impurity in the solid metal, and the melting temperature of the eutectic or chemical compound in metal–impurity system is lower than the melting temperature of metal–solvent. All the considered impurities are characterized by high surface (adsorption) activity. Its theoretical evaluation is usually performed by the totality of several criteria [10]:

$$\Delta\sigma = \sigma_{\text{solv}} - \sigma_{\text{imp}} > 0; \tag{1}$$

$$\Delta P = P_{\text{solv}} - P_{\text{imp}} > 0 \text{ where } P = \frac{L}{V}; \tag{2}$$

$$\Delta T_m = T_{\text{solv}}^m - T_{\text{imp}}^m, \tag{3}$$

where σ_{solv} , σ_{imp} are the surface tensions of the solvent and the impurity, respectively; P_{solv} , P_{imp} is the specific heat of sublimation of the solvent and impurity, respectively; L is the heat of sublimation in the melting point (J/g·atom); V is the atomic volume at melting temperature (cm³/g·atom); T_{solv}^m , T_{imp}^m is the melting

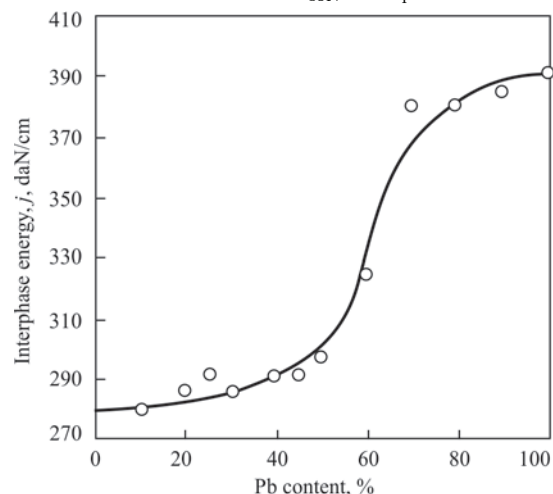


Figure 2. Interphase energy j on the boundary of copper with Bi–Pb melts at 350 °C [12]

Table 2. Equilibrium coefficient of distribution K_0 of impurities in the considered metals

Fe		Ni		Cu			Al
S	P	S	P	Bi	Pb, S	P	Na
0.003–0.03	0.13	≈ 0.0001	0.02	0.0001	0.01	0.11	0.0014

temperature of the solvent and the impurity, respectively.

If inequalities (1)–(3) are satisfied, the impurity is surface active relative to this solvent. As one can see from Table 1, evaluation of the considered impurities by the above criteria is indicative of their surface activity relative to iron, nickel, copper and aluminium, respectively (Figures 1–4).

Another common property of the above impurities is their small coefficient of distribution (Table 2), that promotes an increase of their concentration along the crystallite boundaries, and enhances their harmful impact even at small concentrations of the impurity in the metal.

Numerical values of the coefficient of distribution in Table 2, are given by the data of works [14, 15], and were also calculated by us by the respective binary state diagrams [16].

Thus, all the considered impurities have the following common specific physicochemical and thermodynamic properties:

- state diagram of metal-impurity system of eutectic type with limited (small) mutual solubility;
- melting temperature of the impurity, eutectic or chemical compound in metal-impurity system is below the melting temperature of metal-solvent;
- high surface (adsorption) activity of the impurity;
- small coefficient of impurity distribution.

As is known from physicochemical mechanics of metals [17], the above-mentioned specific properties of impurities are indicative of the fact that in the system of solid metal–liquid phase, containing surface-active elements, the effect of adsorption-induced lowering of ductility and strength (special case of Re-binder effect) or, as it is sometimes called, the effect of liquid metal brittleness (LMB) can be manifested.

And indeed, solidification cracks form during deformation of weld metal, which is in solid-liquid state. Therefore, the specifics of physicochemical interaction of the solid body with the liquid phase (manifested in different adsorption activity of the latter), should have a certain influence on weld metal deformability.

In terms of physicochemical mechanics of metals [14] lowering of metal deformability can be presented as follows:

- in thermodynamic aspect: surface-active elements reduce the free surface energy of the solid and thus facilitate crack initiation;
- in molecular-kinetic aspect: during physicochemical interaction of the solid with atoms of surface-active elements restructuring and breaking of interatomic bonds are facilitated.

E.E. Glikman with associates [18] conducted a cycle of experimental and theoretical studies to clarify the micromechanism of embrittling action of surface-active melts (Rebinder effect). As a result, mo-

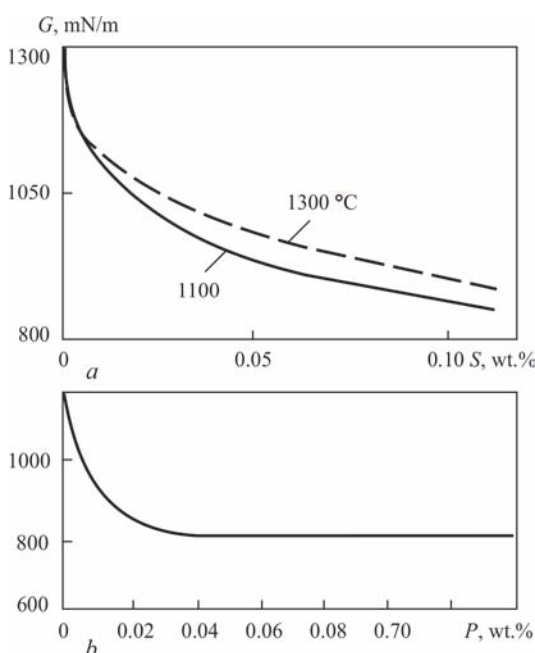


Figure 3. Effect of sulphur (a) and phosphorus (b) on surface tension of copper [13]

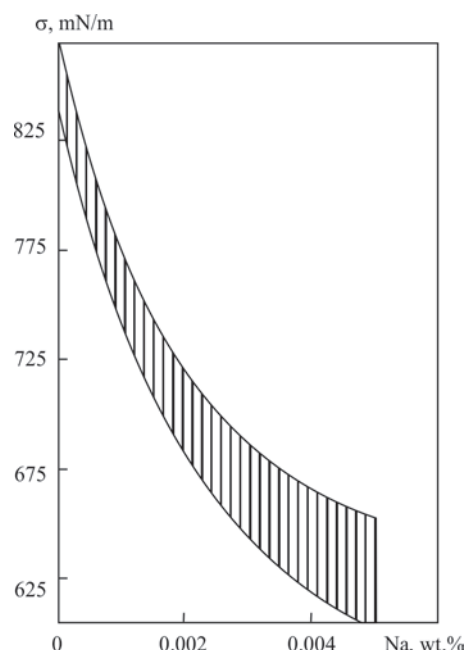


Figure 4. Effect of sodium on surface tension of aluminium [13]

lecular-kinetic model of fracture under the impact of surface-active melt was proposed.

The controlling stage of fracture is that of subcritical crack growth. Its duration determines the time to fracture at creep and limit deformation to rupture (under tension). After the crack has reached a certain length, the subcritical growth stops and rapid fracture takes place. An abrupt acceleration of crack growth is related to achievement of a certain critical deformation in the plastically deformed region at the tip, the measure of which is the so-called critical crack opening displacement δ_c .

Quantitative expression of the rate of subcritical crack growth in the proposed mechanism of embrittling action of surface-active melts is given by the following formula [18]:

$$V = A \frac{(\sigma - \sigma^*) \exp\left(\frac{Q}{KT}\right)}{T},$$

where Q is the activation energy of subcritical crack growth

$$Q = (S_{SL} + S_c)\gamma_{SL} - S_b\gamma_b + H_l - S_c\gamma_{OSL},$$

where σ is the deforming stress; σ^* is the threshold stress, at achievement of which the embryo microcrack, filled with the melt, starts growing; γ_{SL} is the specific surface energy on solid metal–melt interface; γ_b is the specific surface energy of grain boundaries; A is the constant: $A = (\omega^{5/3}D_0C_0)/(R^2hK)$ (for designations see work [18]).

The greatest influence on the rate of subcritical crack growth is produced by value Q and, in particular, specific surface energy on solid metal–melt boundary (γ_{SL}) and specific surface energy of grain boundaries (γ_b). With reduction of γ_{SL} and increase of γ_b the crack growth rate increases exponentially. So, γ_{SL} decrease by 25 % leads to 30 times increase of crack growth rate [18]. Such an abrupt dependence of fracture rate on the specific fracture energy on «crystal–melt» boundary is an important experimental result, which confirms on the microscopic level the main postulate of the hypothesis of adsorption-induced lowering of strength (ALDS) about the determinant role of surface energy lowering in facilitating restructuring and breaking of interatomic bonds at the crack tip.

In our opinion, the proposed molecular-kinetic model, explaining the embrittling impact of surface-active melts, the most fully reflects the available experimental data on the influence of various physico-chemical conditions on manifestation of ALDS effect.

Summarizing the above-said, the mechanism of the influence of surface-active elements on cracking susceptibility, in our opinion, can be presented as follows:

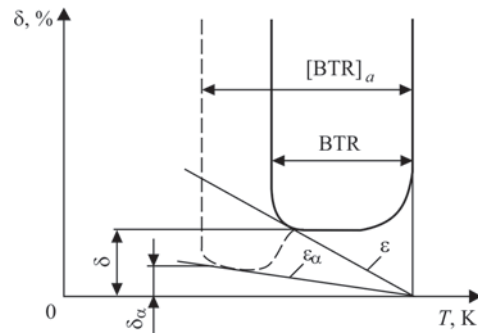


Figure 5. Dependence of relative elongation on solidification temperature (scheme)

- during weld metal solidification, the composition of the liquid phase along the crystallite boundaries will change in the direction of its enrichment by surface-active elements, as a result of their low coefficient of distribution. Maximum achievable concentration of the surface-active element in the liquid interlayer will be equal to value C_0/K_0 , where C_0 is the average concentration of the element in the weld pool; K_0 is the equilibrium coefficient of distribution;
- as a result of manifestation of the effect of adsorption-induced ductility and strength lowering, the surface-active elements lower the deformability of the solidifying metal, and also expand its brittleness temperature range.

This is graphically presented in Figure 5, and the experimental data are given in Figure 6.

B.A. Movchan also notes that at solidification «an exceptionally high concentration of impurity forms on the crystallite boundary» [19].

Available experimental data confirm the above concepts. So, by the data of Auger spectroscopy [20], in copper containing ≈ 0.003 % Bi, its concentration on grain boundaries reaches ≈ 50 %. By the data of work [21], sulphur content on the crack surface in ingots of oxygen-free copper was equal to ≈ 0.75 % at its concentration in the metal equal to 0.002 %. Sulphur concentration in welds on steel is equal to

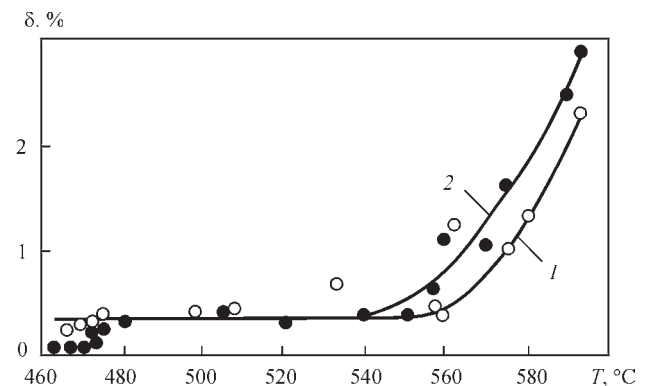


Figure 6. Effect of sodium content in aluminium alloy V95 on relative elongation in solidification temperature range [5]: 1 — alloy V95; 2 — alloy V95 + 0.0034 wt.% Na

10–12 % along crystallite boundaries at its average content of ≈ 0.03 % [2].

Investigation of the morphology and composition of surface layer of hot cracks forming in monel welding, which was performed by the methods of electron fractographic and X-ray emission analyses, also confirms the above-said: the crack surface is mainly enriched in sulphur (27 times) [22].

Experimental data on manifestation of ALDS effect, caused by various surface-active elements in welding, were presented for the first time in works [23, 24]. These data are indicative of the fact that abrupt embrittling action on metals can be produced not only by impurities, but also by alloying elements at certain concentrations, which are applied as effective deoxidizers, desulphurizers, modifiers, etc., and which are characterized by the above physicochemical properties.

Such an alloying element is cerium, causing solidification cracking in monel welding [25].

Proceeding from the above mechanism of the impact of surface-active elements on solidification cracking, the main metallurgical measures for their prevention are as follows:

1. Lowering of surface activity of the solidifying liquid phase (γ_{SL} increase) by:

- reduction of impurity concentration;
- binding the impurities into refractory or low surface-inactive compounds.

2. Reduction of surface energy of grain boundaries by:

- alloying by surface-active elements;
- changing the strength properties and crystalline structure of the solid phase.

It should be noted that the above factors (item 1) not only increase the surface energy of the liquid phase γ_{SL} , but also reduce the BTR.

Let us consider the examples of application of the above-mentioned measures of solidification cracking prevention in welding various metals.

1. γ_{SL} increase

Concentration of impurities along the crystallite boundaries can be reduced both through application of refining fluxes (that is not always effective), and by changing the nature of alloy solidification. So, the amount of the liquid phase at solidification of single-phase alloys tends to zero, when approaching the solidus temperature (by the main components) (Figure 7), and the concentration of the impurity along the crystallite boundaries tends to value C_0^{imp}/K_0 [26] (where C_0^{imp} is the initial impurity concentration in the metal, and K_0 is the equilibrium coefficient of distribution). Impurity concentration in this case can be high even at a low initial content in the metal that, in its turn, can lead to solidification cracking, as a result of ALDS effect manifestation. At solidification of two-phase alloys (of eutectic type), when approaching the solidus temperature (eutectic), the amount of the liquid phase is not equal to zero, but has finite values (Figure 7). As a result, impurity concentration on crystallite boundaries at the final stage of solidification will be equal to the initial value (C_0^{imp}), i.e. enrichment of crystallite boundaries will practically be absent, and, thus, solidification cracking resistance will be enhanced.

This, in our opinion, can account for higher technological strength of two-phase aluminium bronzes and brasses, as well as chromium-nickel bronze, compared to single-phase alloys and metals.

Binding impurities into even though low-melting, but low surface-active compounds also allows increasing the resistance to solidification cracking. So, for instance, presence of oxygen in low-carbon steel reduces surface activity of sulphur as a result of its binding into oxysulphides, and, although the melting temperature of oxysulphide phase is below the iron sulphide melting temperature, the embrittling action of sulphur decreases, and metal cracking resistance increases, respectively. V.V. Podgaetsky et al. [2], experimentally showed an increase of interphase tension

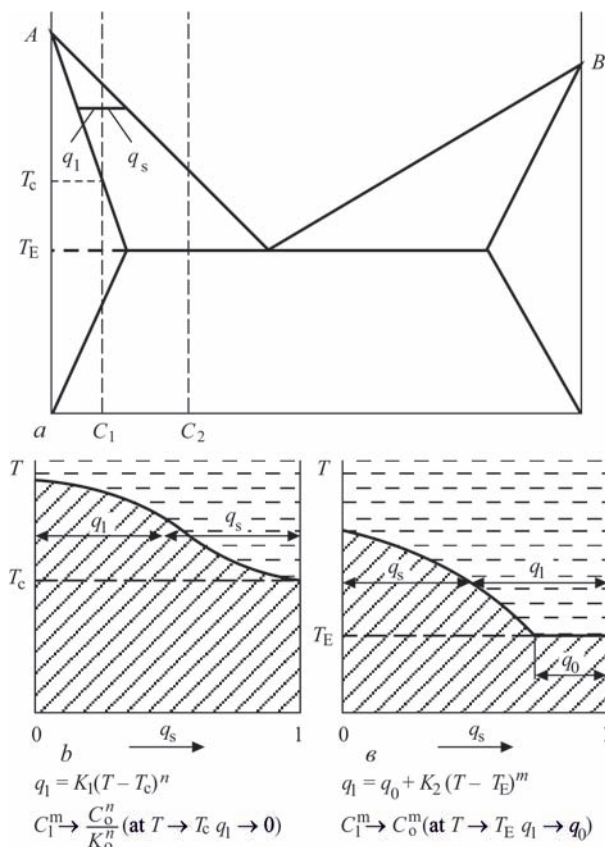


Figure 7. Schematic of the state diagram of two-component alloy (a) and features of solidification of single-phase C_1 (b) and two-phase C_2 (c) alloys

of oxysulphide phase (lowering of surface activity), which is expressed in changing of the film form of the sulphide phase to globular form at oxygen addition to the weld.

In low-carbon steel manganese is also used for binding sulphur into a refractory compound (MnS), due to its low equilibrium coefficient of distribution K_{0Mn}^{Fe} [14].

As was noted in [27], L. Riv, when studying the influence of the composition of metal of welds made by electrodes, determined that electrodes the least susceptible to cracking produce the most oxidized weld metal (total oxygen content of 0.17 to 0.20 %). The electrodes the most susceptible to cracking produced welds with minimum oxygen content (0.05–0.06 %). This can be explained by the fact that at oxygen content of 0.05 % the surface activity of sulphur increases 5 to 10 times [28].

2. Reduction of grain boundary energy (γ_b)

Metal alloying by elements, lowering the surface energy of grain boundaries, at a certain concentration will reduce the probability of solidification cracking. So, for instance, monel alloying by surface-active element — cerium reduces γ_b , and, thus, increases the cracking resistance of welds.

Considering the relation between grain boundary surface energy γ_b and solid surface energy γ_s ($\gamma_b \approx 0.27\gamma_s$ [29]), as well as the correlation dependence between γ_s of a solid and its mechanical properties (ultimate strength, hardness) [30], one can note that metal alloying by strengthening elements will increase and, other conditions being equal, will strengthen the harmful effect of the impurities, and, the probability of cracking, respectively. This is also noted in works [12, 17].

The experiments performed by us on copper-nickel alloys confirm the above-said [31]. Increase of aluminium content in the alloy led to increase of its hardness and of solidification cracking susceptibility, accordingly, both in the weld metal and in the HAZ.

3. BTR reduction

The most effective reduction of BTR will be observed at metal alloying with elements, firstly, increasing the liquid phase solidus temperature, and, secondly, having a small coefficient of distribution that will ensure an increase of the solidus temperature at small concentrations of the element and at practically unchanged liquidus temperature, i.e. BTR is reduced.

Presence of a region of solid solutions of base metal and alloying element and surface inactivity of the latter will be indicative of absence of liquid metal brittleness in this system.

It should be also noted that elements with a small coefficient of distribution have the metal-alloying el-

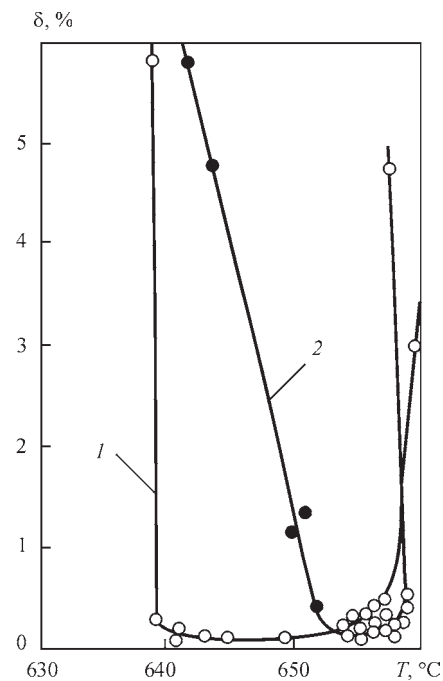


Figure 8. Effect of iron content on relative elongation of aluminium alloy (Al + 1.5 wt.% Mn) in the solidification range ([5]): 1 — Al + 1.5 % Mn; 2 — Al + 1.5 % Mn + 0.25 % Fe

ement state diagram of eutectic type, i.e. at metal alloying with such an element, at concentration higher than its maximum solubility at T_{eut} (when the eutectic starts appearing), impurity concentration along the crystallite boundaries will be also lowered, and surface activity of the residual liquid phase will decrease, respectively. Optimum concentration of the alloying element is determined experimentally.

Such elements, for instance, in aluminium alloys and commercial aluminium, are iron (Figure 8), as well as nickel, cerium and scandium.

State diagrams of aluminium with these elements are given in works [32, 33].

Thus, analyzing the influence of various alloying elements on the change of surface energies of the liquid phase γ_{SL} and grain boundaries γ_b , including also

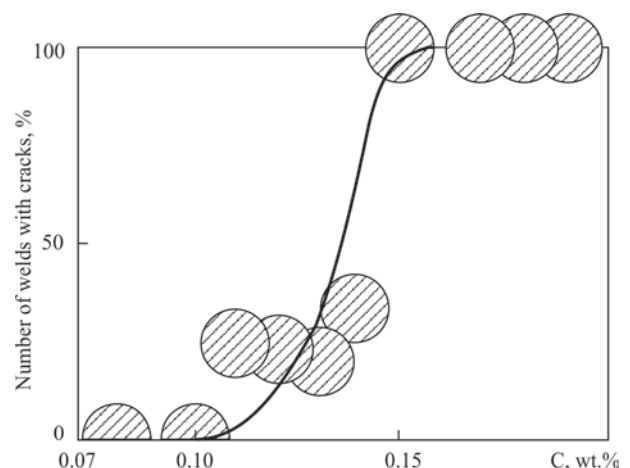


Figure 9. Dependence of frequency of solidification cracking on carbon content for fillet welds of tee joints of low-carbon steel [2]

the nature of metal or alloy solidification, we can assess their influence on the probability of solidification cracking.

So, for instance, the harmful influence of carbon in welding low-carbon steel can be explained as follows:

- firstly, carbon increases the surface activity of sulphur in iron (lowers γ_{SL}), both as a result of its physicochemical properties, and due to steel deoxidation, i.e. lowering of positive influence of oxygen;
- secondly, carbon enhances the strength properties of the solid phase, and increases its surface energy, accordingly.

An abrupt increase of cracking susceptibility at increase of carbon content in steel above 0.16 % (Figure 9, *a*) [2] is associated, in our opinion with the change of the nature of solidification, namely: at carbon concentration of ≈ 0.10 –0.16 % the amount of the liquid phase at the last stage of solidification has a final value, and no significant enrichment in sulphur occurs along the crystallite boundaries, and solidification is over at $T = 1499$ °C (Figure 9, *b*) [34]. At the content of C > 0.16 % the amount of the liquid phase at the final stage of solidification tends to zero, and enrichment of the crystallite boundaries with sulphur become markedly greater (γ_{SL} decreases), and BTR increases, respectively. More over, appearance of austenite grains leads to increase of surface energy of grain boundaries (γ_b). All that is exactly what causes an abrupt increase of cracking susceptibility of low-carbon steel at carbon concentration higher than 0.16 %.

The given arguments provide an explanation also for the following fact. Dr. Vera, Hungarian metal scientist, studied the hot brittleness of Al–Si alloy [5]. He established that maximum hot brittleness is observed at 1.6 % Si content. At the same time, at 1.88 % Si content the hot brittleness is practically zero. Vera explains this fact by crack healing by the melt. This is possible at achievement of a critical amount of the eutectic (12–13 %) [5]. At 1.88 % Si concentration, only of ≈ 2 % eutectic can form, according to Al–Si state diagram. Therefore, this explanation is unconvincing, the more so that Vera did not have any direct proof of crack healing by the melt [4]. In our opinion, this fact can be explained as follows: proceeding from Figure 7, as well as Al–Si state diagram at 1.6 % Si content, Na saturation will be maximum Na ($K_0^{Na} = 0.0014$), and Na concentration will be equal to C_0/K_0^{Na} , and at 1.88 % Si content the eutectic already is in place (according to Al–Si state diagram [32] the eutectic starts appearing already at 1.65 % Si), and no enrichment occurs (Na concentration is equal to the initial one C_0).

It should be noted that sodium is the main cause for solidification cracking in aluminium and its alloys, because of the small coefficient of distribution ($K_0^{Na} = 0.0014$).

In [35] it is noted that 100 % rejection of ingots because of cracks is observed at the Na content > 0.001 % in aluminium alloy 1920. In our opinion in aluminium and its alloys, particularly high-strength ones, sodium content should be limited and controlled. At the same time, it is not controlled in GOST 4784–97 (Aluminium and wrought aluminium alloys), although its content in aluminium alloys can be within 0.001–0.03 % [36].

Summarizing the above said, one can note that the formula of subcritical crack growth, in our opinion, can be the criterion of evaluation of the element effect on solidification cracking, and taking it into account can reduce the time and metal consumption in development of new welding and welded materials.

Conclusions

1. Based on analysis of physicochemical properties of impurities, the concepts of the mechanism of their effect on solidification crack formation were summarized. They consist in the following: firstly, enrichment of crystallite boundaries with them at the last stages of solidification, secondly, manifestation of the effect of adsorption-induced lowering of ductility and strength. The graphic interpretation of this mechanism was proposed, illustrating the lowering of deformability of the solidifying metal and increase of BTR under the impact of surface-active impurities.

2. Difference in the nature of solidification of single- and two-phase (with eutectic) alloys was established as regards enrichment of crystallite boundaries with impurities.

3. Application of molecular-kinetic model of solid fracture under the impact of a surface-active melt (proposed by E.E. Glikman et al.) enables explaining from a single point of view the available experimental data on the influence of various metallurgical factors on solidification cracking. It is shown that specific surface energies on solid metal–melt boundary (γ_{SL}) and on grain boundaries (γ_b) have the strongest impact on crack formation.

4. It is also shown that surface-active alloying elements having the same physicochemical properties as those of the impurities can also influence the higher susceptibility to solidification cracking.

5. The formula of subcritical crack growth can be a criterion for evaluation of the influence of surface-active elements on deformability of the solidifying metal (on solidification cracking).

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