Thermodynamic Aspects of Cr₂O₃ Reduction by Carbon

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Received June 3, 2019; revised September 11, 2019; accepted September 20, 2019

Abstract—In order to save chromium resources, technology of flux-cored wire surfacing is of great practical interest. In this case, Cr₂O₃ chromium oxide and carbon as a reducing agent are used as fillers. The thermodynamic probability assessment of 16 reactions between them under standard conditions, as well as for certain reactions under different standard conditions, was carried out using tabulated thermodynamic data of reactants in the temperature range of 1500-3500 K. The following were considered as standard states for reactants: Cr(ref) (reference state, melting point 2130 K, boiling point 2952 K), Cr(liq), Cr(gas), Cr₂O₃(cr, liq), Cr₂O₃(gas), C(ref), and as possible reaction products and standard states for them CO(gas), CO₂(gas), $Cr_{23}C_6(cr)$, $Cr_7C_3(cr)$, $Cr_3C_2(cr)$. The reaction probability was estimated using standard Gibbs energy and was calculated using the Van Goff isotherm equation. The chromium dissolution in metal of surfacing bath or probable partial pressures of CO and CO₂ in gas phase was considered and then calculated from the equilibrium of carbon gasification reaction. The carbon presence in flux-cored wire with chromium oxide Cr_2O_3 as a reducing agent will necessarily lead to the occurrence of reduction reactions with chromium carbide generation, and possibly chromium itself. The generation of $Cr_7C_3(cr)$ carbide is likely. With a longer life span of chromium oxide and carbon at a temperature above 2500 K, chromium generation as a component of the surfacing bath is more thermodynamically probable than carbide generation. Chromium oxide has the highest reactivity in Cr₂O₃(liq) state. Direct reduction is preferential. The generation of CO(gas) as a carbon oxidation product is more probable. The chromium dissolution in metal increases the thermodynamic reaction probability with its generation, as well as further reduces the reaction probability in which chromium is the starting material.

Keywords: analysis, Gibbs energy of reaction, cored wire, chromium oxide, carbon, arc surfacing, reduction, chromium, chromium carbides

DOI: 10.3103/S0967091219120052

INTRODUCTION

This work is a logical continuation of a series of works [1-4], which laid the foundation for the preparation of a thermodynamic basis for developing a resource-saving technology to create an alloyed metal layer on the surface of metal products during electric arc surfacing. This is due to the formation of alloying elements directly in the surfacing process as a result of the interaction of oxide materials and reducing agents, purposefully introduced into the flux-cored wire. From the analysis of physical processes during surfacing [5-7], the following process lasts about 10 s: liquid droplet formation on the electrode's end surface, its detachment, transfer of a substance into a liquid weld pool on the weld pool metal. In this case, temperature on the drop-

let's surface during separation can reach 2950–3000 K. A characteristic feature of the process is that temperature in the arc's region can reach 10000-12000 K. Some electrode components during evaporation are located for a short time in the near-electrode space and near the arc shank at temperatures above 3000 K in the atomic, molecular, and ionized states (the arc can burn stably even in vacuum). In fact, the determination of what metal oxides are recognized as alloying is now being solved. With the use of certain reducing agents, the greatest thermodynamic probability of the reduction product formation in a short time in a hardened metal of the weld pool can be expected. It should also be noted that during current thermodynamic calculations, the possibility of participation in the reactions of ionized plasma particles is not considered.

THERMODYNAMIC EVALUATION

In this paper, thermodynamics of the interaction in the chromium oxide—carbon system is analyzed. A thermodynamic assessment of the occurrence probability of the following reactions is carried out:

$$\frac{2}{3}\operatorname{Cr}_2\operatorname{O}_3(\operatorname{cr}, \operatorname{l}) + 2\operatorname{C}(\operatorname{ref}) \to \frac{4}{3}\operatorname{Cr}(\operatorname{ref}) + 2\operatorname{CO}(\operatorname{g}); \quad (1)$$

$$\frac{2}{3}$$
Cr₂O₃(cr,l) + C(ref) $\rightarrow \frac{4}{3}$ Cr(ref) + 2CO₂(g); (2)

$$\frac{1}{3}\operatorname{Cr}_2\operatorname{O}_3(\operatorname{cr}, \mathbf{l}) + \operatorname{CO}(\mathbf{g}) \to \frac{2}{3}\operatorname{Cr}(\operatorname{ref}) + \operatorname{CO}_2(\mathbf{g}); \quad (3)$$

$$\frac{2}{3}Cr_2O_3(cr, l) + 2C(ref) \to \frac{4}{3}Cr(l) + 2CO(g); \quad (4)$$

$$\frac{2}{3}Cr_2O_3(cr, l) + 2C(ref) \to \frac{4}{3}Cr(g) + 2CO(g); \quad (5)$$

$$\frac{2}{3}\operatorname{Cr}_2\operatorname{O}_3(l) + 2\operatorname{C}(\operatorname{ref}) \to \frac{4}{3}\operatorname{Cr}(\operatorname{ref}) + 2\operatorname{CO}(g); \quad (6)$$

$$\frac{2}{3}Cr_2O_3(l) + \frac{54}{23}C(ref) \to \frac{4}{69}Cr_{23}C_6(cr) + 2CO(g); (7)$$

$$\frac{2}{3}Cr_2O_3(l) + \frac{54}{21}C(ref) \to \frac{4}{21}Cr_7C_3(cr) + 2CO(g); (8)$$

$$\frac{2}{3}Cr_2O_3(l) + \frac{26}{9}C(ref) \to \frac{4}{9}Cr_7C_3(cr) + 2CO(g); \quad (9)$$

$$\frac{4}{3}\mathrm{Cr(ref)} + \frac{23}{81}\mathrm{C(ref)} \to \frac{4}{69}\mathrm{Cr}_{23}\mathrm{C}_{6}(\mathrm{cr}); \qquad (10)$$

$$\frac{4}{3}\operatorname{Cr}(\operatorname{ref}) + \frac{4}{7}\operatorname{C}(\operatorname{ref}) \to \frac{4}{21}\operatorname{Cr}_7\operatorname{C}_3(\operatorname{cr}); \qquad (11)$$

$$\frac{4}{3}\operatorname{Cr}(\operatorname{ref}) + \frac{8}{9}\operatorname{C}(\operatorname{ref}) \to \frac{4}{9}\operatorname{Cr}_3\operatorname{C}_2(\operatorname{cr}); \tag{12}$$

$$\frac{1}{3}\operatorname{Cr}_{2}\operatorname{O}_{3}(l) + \operatorname{CO}(g) \to \frac{2}{3}\operatorname{Cr}(\operatorname{ref}) + \operatorname{CO}_{2}(g); \quad (13)$$

$$\frac{23}{81}Cr_2O_3(l) + \frac{93}{81}CO(g) \rightarrow \frac{2}{81}Cr_{23}C_6(cr) + CO_2(g);(14)$$

$$\frac{7}{27} Cr_2 O_3(l) + \frac{33}{27} CO(g) \rightarrow \frac{2}{27} Cr_7 C_3(cr) + CO_2(g); (15)$$

$$\frac{3}{13} Cr_2 O_3(l) + \frac{17}{13} CO(g) \rightarrow \frac{2}{13} Cr_3 C_2(cr) + CO_2(g). (16)$$

All reactions were recorded per 1 mol of oxygen. Thermodynamic characteristics of reactions (1)–(16) under standard conditions $[\Delta_r H^{\circ}(T), \Delta_r S^{\circ}(T), \Delta_r G^{\circ}(T)]$ were calculated by known methods [8] in the temperature range of 1500–3500 K by thermodynamic properties [[$H^{\circ}(T)$ — $H^{\circ}(298.15 \text{ K})$], $S^{\circ}(T)$, $\Delta_{f}H^{\circ}(298.15 \text{ K})$] of reagents Cr₂O₃, Cr, C, CO, CO₂, Cr₂₃C₆, Cr₇C₃, Cr₃C₂ [9].

The following ones were used as standard states for reagent substances in the range of 1500-3500 K: Cr(ref) (reference state) with a melting point of 2130 K and a boiling point of 2952 K; Cr(l) (1500-3500 K); Cr(g) (1500–3500 K); $Cr_2O_3(cr, g)$ with a melting point of 2603 K; Cr₂O₃(l) (1500-3500 K); C(ref) (1500–3500 K); CO(g) (1500–3500 K); CO₂(g) (1500–3500 K); Cr₂₃C₆(cr) (1500–2000 K); Cr₇C₃(cr) (1500–2500 K); Cr₃C₂(cr) (1500–2500 K). It follows from the list of standard states that the standard states in the calculations remained unchanged for some substances: C(ref), CO(g), CO₂(g), $Cr_{23}C_6(cr)$, $Cr_7C_3(cr)$, $Cr_3C_2(cr)$. The chemical composition of chromium carbides, in which there are tables of thermodynamic properties in [8], coincides with the data on the state diagram of the Cr-C system in [9].

The standard values of Gibbs energy for the considered reactions (1)–(16), obtained as a calculation result, are given in the table and in the figure. For analysis and discussion, all 16 reactions can be divided into six groups. When performing calculations according to reactions (1)–(3) (first group), the standard states for chromium oxide $Cr_2O_3(cr, g)$ and chromium Cr(ref) did not change. The thermodynamic probability of oxide reduction was evaluated chromium with carbon to produce chromium and carbon oxide (II)reaction (1), as well as chromium and carbon oxide (IV)-reaction (2) and the probability of producing chromium due to the reduction of chromium oxide with carbon oxide (II)) is reaction (3). As follows from the data in the table, reaction (1) differs in the greatest thermodynamic probability, and the reduction ability of solid carbon sharply increases with increasing temperature, as expected. The occurrence of reaction (3) is not thermodynamically probable under standard conditions, and reaction (2) becomes possible after 3100 K.

The second group includes reactions (4) and (5), in which CO₂ (g) is not considered a possible product of the direct reduction reaction of chromium oxide with carbon, but the standard state for chromium changes: $Cr(ref) \rightarrow Cr(l)$ —reaction (4) and $Cr(ref) \rightarrow Cr(g)$ reaction (5). It follows from the calculations that metastable states of chromium Cr(l) above the boiling point (superheated liquid chromium) and Cr(l) below the melting point (supercooled liquid) in the range of 2000–3500 K are stable in the reaction equilibrium, since the standard Gibbs energy of the reaction (4) differs little from the standard Gibbs energy of reaction (1).

The state of chromium in the form of Cr(g) vapor (reaction (5)) at temperatures lower than its boiling point is unlikely, especially at temperatures below 2000 K.

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Reaction	$\Delta_r G^{\circ}(T), \Delta_r G(T)$ (bottom row in line), kJ, at T, K							
	1500	2000	2130	2500	2603	2952	3000	3500
(1)	8.14	-161.06	-205.02	-334.20	-370.13	-478.78	-500.92	-730.50
(2)	164.99	115.84	103.39	64.50	53.84	31.19	20.91	-84.49
(3)	95.26	95.21	94.94	91.55	90.50	93.05	89.73	55.28
	4.06	-80.53	—	-167.10	_	_	-250.46	-365.25
(4)	17.72	-159.31	-204.99	-334.20	-369.97	-478.76	-493.58	-646.98
(5)	247.46	-10.80	-77.15	-264.40	-316.16	-478.76	-500.92	-730.50
(6)	-25.75	-180.32	-220.29	-337.60	-370.13	-478.78	-500.92	-730.50
(7)	-50.55	-206.56	_	—	—	—	—	_
(8)	-65.81	-223.42	-	-378.12	—	-	-	—
(9)	-34.85	-193.12	—	-348.49	—	—	—	—
(10)	-24.80	-26.25	—	—	—	—	—	—
(11)	-40.06	-43.10	-43.62	-40.52	—	—	—	—
(12)	-47.04	-50.74	-51.44	-48.82	—	—	—	—
(13)	78.32	85.58	87.30	89.85	90.56	93.05	89.73	55.28
	-12.88	-90.16	-	-168.80	—	—	-250.46	-365.25
(14)	69.66	87.75	-	-	-	-	-	—
	-21.54	-87.98	—	—	—	—	—	—
(15)	65.60	88.85	94.82	111.61	—	—	—	—
	-25.60	-86.88	-	-147.05	—	—	—	—
(16)	66.00	95.75	103.39	124.89	_	_	_	_
	-25.20	-79.98	_	-133.76	_	_	_	_

Table 1. Standard Gibbs energies, Gibbs energies of reactions (1)-(16) depending on temperature

Reaction (6) is the third logical position in the list of reaction groups. Here, Cr(ref) and C(ref) are the most preferred standard states and change the standard state for chromium oxide: $Cr_2O_3(cr, 1) \rightarrow Cr_2O_3(1)$. Metastable liquid chromium oxide at temperature of less than 2603 K has a higher chemical affinity for carbon during the formation of Cr(ref) and CO(g) than crystalline chromium oxide (Table 1).

The fourth group of reactions consists of reactions (7)–(9), in which the formation probability of chromium carbides is considered naturally upon the interaction of Cr_2O_3 (g), C(ref) and CO(g). All three reactions in the temperature range of stability of incongruently melting solid chromium carbides are thermodynamically more likely than reactions (1) and (6). Moreover, according to the available thermodynamic data for carbides, the carbide formation of the $Cr_7C_3(cr)$ composition is most likely.

In the fifth group of reactions, the thermodynamic probability formation of three chromium carbides was considered by direct interaction between chromium and carbon (reactions (10)-(12)). All three reactions are

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likely. However, chemical affinity of chromium to carbon is less than the affinity of chromium oxide to carbon with the formation of the same carbides and CO.

In the sixth group of reactions, the formation possibility of three chromium carbides by reduction of liquid chromium oxide $Cr_2O_3(l)$ with carbon monoxide (II) was considered. For comparison, the formation of Cr(ref) chromium from the same starting materials was also included here (reaction (13) differs from reaction (3) only by the standard state of chromium oxide). From the data in the table, it follows that chromium carbides also cannot be obtained under standard conditions by indirect CO reduction, as well as pure chromium.

Another feature of the electric arc surfacing is intense convective flows in molten phases: both on the electrode and in the surfacing bath. Thus, upon the thermodynamic assessment, it is necessary to consider that chromium formed as a result of the reactions (1)-(4), (6), (13). Also, chromium as the starting material for reactions (10)-(12) will probably be in the state of the dissolved component at temperature *T*.



Fig. 1. Standard Gibbs energies of reactions (1)-(16) depending on temperature.

Thermodynamic assessment of this state's influence on the reaction properties is carried out according to the equation of the Vant–Hoff isotherm:

$$\Delta_r G = \Delta_r G^\circ \pm \nu RT \ln \alpha_{\rm [Cr]},\tag{17}$$

where $\alpha_{[Cr]}$ is chromium activity in the molten metal relative to the standard state of "pure solid or liquid solid or liquid (depending on temperature) chromium at melt temperature T"; v is the stoichiometric coefficient in front of chromium in the reaction equation (the "+" sign is used for the reactions, in which chromium is the product, and the "-" sign is for the reactions, in which chromium is the starting material).

To assess the trend, the calculation of the second term in equation (17) at $v = \frac{4}{3}$ and constant tempera-

ture of 2500 K (average interval temperature) is enough to observe the influence degree at various probable activities. Thermodynamic contribution of the chromium solubility in the molten metal to the Gibbs energy of the reactions is given below:

$$\alpha_{[Cr]}$$
 0.001 0.01 0.1

$$\pm 2500\frac{4}{3} R \ln \alpha_{[Cr]}, kj$$
 191.437 127.63 62.8

Assessment of the trend shows that chromium dissolution increases the thermodynamic probability of reactions occurring with its formation: $\Delta_r G$ becomes more negative than $\Delta_r G$. On the contrary, it will further reduce the reaction probability (10)–(12). To a greater extent, the change in $\Delta_r G$ compared with $\Delta_r G$ occurs at the minimum values of chromium activity ($\approx \pm 200$ kJ).

Considering that the entire negative region of $\Delta_r G$ on the graph (see the figure) occupies an interval of 750 kJ, the contribution of 200 kJ looks very significant; in this case, the gap in the occurrence probability between some reactions increases significantly.

In the presence of solid carbon and oxygen, the content of CO and CO_2 gases and their partial pressures in the system cannot be arbitrary, but are determined by the equilibrium of the carbon gasification reaction:

$$C + CO_2 \rightarrow 2CO. \tag{18}$$

The peculiarity of the thermodynamics of this reaction is that, starting from temperature of 1500 K, the equilibrium gas phase consists practically of individual CO. Therefore, taking $P_{CO_2} = 1$ atm, as standard gas conditions require, and drawing conclusions on $\Delta_r G$ about the depth of reactions (3), (13)–(16) is not entirely correct. Therefore, $\Delta_r G$ was additionally calculated for these reactions by the Vant–Hoff isotherm equation considering the equilibrium partial pressures of P_{CO} and P_{CO_2} , according to the carbon gasification reaction, in which its equilibrium was calculated separately. For reactions (3), (13)–(16), the equation of Vant–Hoff isotherm can be written as follows:

$$\Delta_r G(3) = \Delta_r G^{\circ}(3) \pm RT \left(\ln P_{\rm CO_2} - \ln P_{\rm CO} \right); \quad (19)$$

$$\Delta_r G(13) = \Delta_r G^{\circ}(13) \pm RT \left(\ln P_{\rm CO_2} - \ln P_{\rm CO} \right); \quad (20)$$

$$\Delta_r G(14) = \Delta_r G^{\circ}(14) \pm RT \left(\ln P_{\rm CO_2} - \frac{93}{81} \ln P_{\rm CO} \right); \quad (21)$$

$$\Delta_r G(15) = \Delta_r G^{\circ}(15) \pm RT \left(\ln P_{\rm CO_2} - \frac{33}{27} \ln P_{\rm CO} \right); \quad (22)$$

$$\Delta_r G(16) = \Delta_r G^{\circ}(16) \pm RT \left(\ln P_{\rm CO_2} - \frac{17}{13} \ln P_{\rm CO} \right); \quad (23)$$

The Gibbs energies of the reactions calculated according to equations (19)-(23) are given in the table (lower value series for the corresponding reactions

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depending on temperature). Data analysis on $\Delta_r G$ shows that, considering the possible actual content of CO and CO₂ in the gas phase, reactions of indirect reduction of chromium oxide become possible and can complement the direct reduction process.

The calculations can be useful in the manufacturing of flux-cored wires containing chromium [11-21].

CONCLUSIONS

The thermodynamic analysis of 16 reactions under standard conditions and under different standard conditions shows that the presence of reducing carbon in the flux-cored wire used for surfacing, along with chromium oxide Cr_2O_3 as a reducing agent, necessarily leads to the flow of reduction reactions with the formation of chromium carbides, and possibly chromium itself. The longer the chromium oxide and carbon are in the electric arc process at temperatures above 2500 K, the more thermodynamically probable is the chromium formation process as a surfacing bath component. Chromium oxide has the highest reactivity, being in the state of $Cr_2O_3(g)$. The most probable way of carbon participation in the reduction process is direct reduction. As the product of carbon oxidation, the formation of CO(g) is most likely. The dissolution of chromium in the metal increases the thermodynamic probability of reactions occurring with its formation and, on the contrary, further reduces the reaction probability, in which chromium is the starting material. Considering the possible factual content of CO and CO_2 in the gas phase according to the equilibrium of the carbon gasification reaction, the indirect reduction reactions of chromium oxide become probable and can complement the direct reduction process.

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Translated by Sh. Galyaltdinov

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