

Research Article

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Reactions at the molten flux-weld pool interface in submerged arc welding

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Abstract: In submerged arc welding (SAW) of chromium (Cr) containing steels, Cr is usually added to the weld metal from the weld wire, and not from the welding flux. Manufacturing of weld wires of specific compositions is expensive and time consuming and cannot closely match all the desired alloy compositions. Therefore, the weld wire chemistry is usually over matched to the base plate composition. Better matching between the weld metal and base plate is possible if the weld metal incorporates Cr from Cr containing metal powder, instead of sourcing Cr from weld wire of limited Cr content. Because Cr is easily oxidised, the oxygen partial pressure in SAW must be controlled. This work illustrates the control of the oxygen potential at the molten flux-weld pool interface by using aluminium (Al) powder addition. The controlled oxygen potential at the molten flux-weld pool interface ensures increased Cr powder transfer into the weld pool, without interfering with oxygen transfer from the plasma arc to the weld pool. The objective of this work is to use targeted powder additions to better control Cr reactions in SAW to improve Cr metal transfer to the weld metal and maintain an acceptable level of oxygen in the weld metal.

Keywords: oxygen, chromium, aluminium, powder, flux

1 Introduction

Submerged arc welding (SAW) is applied in the welding of thick steel plates in heavy engineering industries such as shipbuilding and pressure vessel construction [1]. In

the SAW process, electrical and chemical parameters are combined to form an arc between the weld wire tip and the steel base plate. The important electrical parameters in arc formation and arc stability are voltage, current, and the distance between the weld wire tip and the base plate. The important chemical parameters in arc formation and arc stability are the formation of volatilised fluoride compounds, vaporised metals, ions from oxide dissociation in the plasma arc, and the chemical interactions among these compounds, vapours, and ions. SAW process zones are illustrated in Figure 1. Raw unmelted flux and molten flux cover the arc to form the arc cavity. Weld wire and flux are continuously fed through the welding head arrangement as it moves along the weld. In the arc cavity, the molten weld wire metal droplets are transferred into the weld pool as complex physical and chemical interactions of heat and mass transfer occur [1]. Chemical reactions continue in the trailing molten steel weld pool and its molten flux covering, until the steel is solidified as weld metal [2,3].

Flux typically contains a mixture of various oxides and fluorides. Fluoride is usually added as CaF_2 . Fluoride compounds are added to serve a variety of functions. Fluoride is used to reduce diffusible weld metal hydrogen content as it reacts with hydrogen, released from moisture decomposition in the arc, to form insoluble hydrogen fluoride gas [4–6]. CaF_2 is added to control physico-chemical properties of the molten flux. For example, CaF_2 lowers slag viscosity, surface tension, and slag melting temperatures to control the weld bead profile shape. CaF_2 is added to control the slag solidification temperature which should be approximately 50°C below the solidification temperature of the welded steel plate to ensure increased time for element transfer reactions between the flux and weld pool [4,7]. This is also an important flux design parameter used to ensure that the slag layer remains molten for a longer period of time than the weld metal to ensure oxide inclusions incorporate into the slag layer at the slag-weld pool interface, thus ensuring weld metal cleanliness in terms of oxide inclusion content [8]. Chemical reactions between oxides and fluorides form volatilised fluorides which lower the partial pressure of

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oxygen and hydrogen in the arc cavity, which in turn lowers the levels of hydrogen and oxygen added into the weld metal [4,5].

Flux formulations are designed to increase element transfer of targeted elements from the molten flux into the weld metal. For example, SiO_2 and MnO addition to flux formulations are made to control Si and Mn levels in the weld metal of low alloy carbon steels. Manufacturer's user information documentation often refers to flux element transfer behaviour as high, moderate, or low alloying with respect to specific elements. For example, a flux described as high alloying in Si and/or Mn refers to the

$$\text{BI} = \frac{\% \text{CaF}_2 + \% \text{CaO} + \% \text{MgO} + \% \text{BaO} + \% \text{SrO} + \% \text{Na}_2\text{O} + \% \text{K}_2\text{O} + \% \text{Li}_2\text{O} + 0.5(\% \text{MnO} + \% \text{FeO})}{\% \text{SiO}_2 + 0.5(\% \text{Al}_2\text{O}_3 + \% \text{TiO}_2 + \% \text{ZrO}_2)}. \quad (1)$$

manufacturer's flux chemistry design to attain high Si (0.6–0.9%) and high Mn (1.4–1.8%) transfer from the flux to the weld metal [9].

Cr is added to low alloy steels to improve steel oxidation resistance and tensile strength at elevated service temperatures higher than 345°C. For example, Cr-molybdenum steels containing 9% Cr and 1% Mo are used to construct pressure vessels and pipelines for high temperature applications, such as thermal power plant applications [10,11]. Stainless steels contain more than 11% Cr to improve corrosion resistance. Most stainless steel grades may be welded by SAW [11]. Cr is added to the weld metal from the weld wire, and the weld wire chemistry is usually over matched to the base plate composition. Therefore, the fluxes used in SAW of Cr containing alloys are neutral or non-alloyed [9]. If any metallic powders of Cr, nickel, molybdenum, or niobium are added into the flux formulation, this is only done to compensate for arc losses of these elements [11]. Manufacturing of weld wires of specific compositions is expensive and time consuming and cannot closely match all desired

alloy compositions. Better matching between the weld metal and base plate will be possible if the weld metal incorporates metal powder to attain the desired composition. Improved understanding of Cr reactions in SAW will aid in the development of tailored Cr additions to the weld metal.

An important empirically determined flux composition guideline is to ensure that the flux Basicity Index (BI) is in excess of 1.5 to ensure low hydrogen and low total oxygen content in the weld metal [12,13]. Equation (1) illustrates the BI expression used for welding fluxes.

Weld metal oxygen must be controlled within a relatively narrow band to ensure high weld metal impact toughness. Both too low (<200 ppm) and too high (>500 ppm) weld metal oxygen contents were found to be detrimental to weld metal impact toughness [14]. Oxygen is carried from the plasma arc into the weld metal by the molten wire droplets, and this oxygen is sourced from the decomposition of less stable oxides at the high temperatures prevailing in the plasma arc. The flux chemistry is used to manage oxygen transfer to the weld metal from the plasma arc by diluting the oxides with fluorides and by adding more stable oxides to the flux [15]. The plasma arc stability of oxides was determined in welding tests made with binary CaF_2 -oxide flux mixtures, from high to low stability: CaO , K_2O , Na_2O and TiO_2 , Al_2O_3 , MgO , SiO_2 , and MnO [15]. Notably, CrO and Cr_2O_3 were not tested in this work and thus the arc stability of the Cr-oxides is not clear. It has been stated that the oxygen content in the weld metal of 9% Cr heat-resistant steels should be lowered to less than 300 ppm to maintain sufficient room temperature impact toughness [10]. Limited published studies are available on the element transfer behaviour of Cr from flux to weld metal and from metal powder to weld metal [16,17]. The objective of this work is to use targeted powder additions in SAW to better control Cr reactions in SAW to improve Cr transfer to the weld metal and maintain an acceptable level of oxygen in the weld metal.

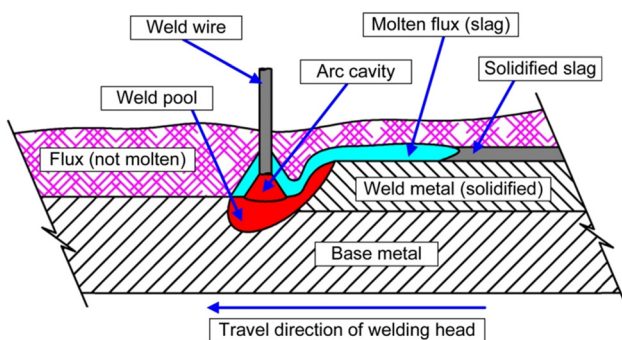


Figure 1: SAW process zone sketch.

2 Materials and methods

Welding experiments were conducted with and without metal powders (Al and Cr) to illustrate the process concept of oxygen potential control at the molten flux-weld

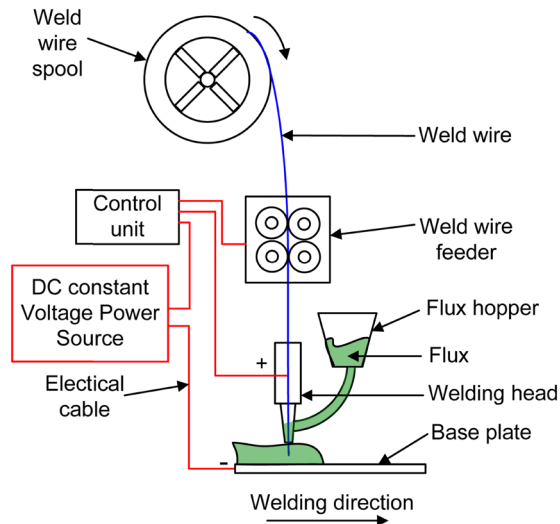


Figure 2: SAW equipment sketch (status: power switched off).

pool interface. SAW welding tests were made as bead-on-plate runs onto steel plates of 350 mm length, 12 mm plate thickness, and 300 mm plate width. The welding heat input of $2.0 \text{ kJ}\cdot\text{mm}^{-1}$ was attained with parameter inputs of 500 A, 28 V, and $42 \text{ cm}\cdot\text{min}^{-1}$ welding speed at (Direct Current Electrode Positive) with wire of 3.2 mm diameter. Figure 2 illustrates the SAW equipment set up used. This is the most commonly used SAW equipment set up [11]. The inputs of current (A), voltage (V), and welding head travel speed ($\text{cm}\cdot\text{min}^{-1}$) are selected on the control unit. The direct current (DC) constant-voltage power source is self-regulating as the power source controls the arc voltage for the arc current setting, which is in turn set by the wire diameter and wire feed speed [11]. Initially, when the weld run is started, the arc length is established by voltage adjustment. If the arc length changes during the weld run due to changes in welding conditions, the current is automatically adjusted to compensate for the change in arc length (arc voltage) [11].

Structural steel grade EN 10025-2 was used as base plate material. The weld wire major element levels are as per the supplier's specification sheet supplied by Afrox Ltd., South Africa. The rest of the element analyses in Table 1 were obtained from laboratory analyses. The base plate steel was analysed by optical emission spectroscopy (OES). The oxygen content in the base plate and weld wire was analysed by combustion method.

The welded plate was sectioned to remove cross section samples of the weld metal for major element analyses by OES and total oxygen content analyses by combustion method. The welded plate was sectioned at the middle position of the weld run to reveal the weld metal cross section. A band saw was used to make this cut, and the one half plate was sent to the laboratory for analyses. The OES analysis was made on the weld metal cross section face after surface preparation consisting of surface leveling and polishing, followed by 2% Nital etching to reveal the weld metal bead area. Following the OES analyses, the weld metal cross section surface preparation was repeated and a manual saw was used to remove a volume sample of the weld metal for oxygen content analysis by combustion method. This procedure was executed three times on different cross section cuts within 10 cm of the first section to obtain three different samples per weld run. A Scanning electron microscope (SEM)-Field emission gun (FEG, Zeiss crossbeam 540) was used in this study. Energy dispersive spectrometer (EDS) analyses were done at 20 kV and 5.6 mm working distance. Commercial agglomerated flux of composition listed in Table 2 was used in the welding experiments. This is an Aluminate Basic flux ($BI = 1.4$) and was extensively analysed as reported previously [18]. Al (99.7%) as supplied by Sigma-Aldrich and Cr (99.0%) as supplied by Alfa Aesar were used as metal powder additions.

3 Results

The weld metal compositions for the two scenarios, the Base Case welding experiment without metal powder addition (BC), and the welding experiment with metal powder addition (MP) are summarised in Table 3. The MP analyses are the average of three different sections made along the weld bead length.

Comparison of the BC and MP weld metal analyses in Table 3 clearly shows increased levels of Si, Mn, Al, and Cr in the MP weld metal due to added metal powders used in SAW. The SEM micrograph in Figure 3 and associated EDS analyses in Table 4 are used to confirm increased Cr content in the weld metal. The EDS analyses in Table 4 confirm the increased Si, Mn, Al, and Cr in the bulk weld

Table 1: Plate and wire compositions (mass%)

	C	O	Si	Mn	Al	P	S	Ti	Cu	Cr
Plate	0.120	0.007	0.155	1.340	0.067	0.019	0.007	0.005	0.030	0.160
Wire	0.110	0.003	0.137	0.990		0.009	0.023		0.140	

Table 2: Flux composition (mass%)

	MnO	CaO	SiO ₂	Al ₂ O ₃	CaF ₂	MgO	FeO	TiO ₂	Na ₂ O	K ₂ O
Flux	6.8	0.1	19.6	24.9	17.9	22.2	2.4	1.0	1.6	0.2

Table 3: Weld metal compositions (mass%)

	C	O*	Si	Mn	Al	Cr	P	S	Ti
Base case (BC)	0.110	499	0.260	1.300	0.032	0.110	0.022	0.011	0.005
Metal powder (MP)	0.081	299	0.980	1.700	6.690	7.790	0.014	0.010	0.065

*ppm.

metal analyses shown in Table 3 as compared to the BC weld metal analysis. For comparison purposes, a SEM micrograph of the BC weld metal is displayed in Figure 4, and the EDS analyses in the marked areas are summarised in Table 5.

4 Discussion

Increased Si and Mn contents in the MP weld metal, compared to the BC weld metal, confirm that sufficient contact time was established between Al powder and molten flux to enable reactions (2) and (3) to proceed forward. The Al₂O₃ products formed in these two reactions most likely ended up in the molten flux, since the reactions occurred at the molten flux-weld pool interface. The unreacted flux already contains a large quantity of Al₂O₃ at 24.9 mass%. An increase in Al₂O₃ content in the molten

flux due to Al reduction reactions will lower the slag viscosity since Al₂O₃ is present as solid spinel (MgO·Al₂O₃) in these types of fluxes, even at the high temperatures found at the molten flux-weld pool interface [18–20]. Furthermore, slag detachment from the weld bead deteriorates with higher Al₂O₃ content as the slag liquid phase percentage is too small at the weld pool steel solidus temperature [19,20]. Therefore, the flux formulation should be adjusted with increased CaF₂ additions to ensure that a sufficient quantity of liquid phase is present in the slag despite the

Table 4: SEM-EDS analyses of marked areas in the MP weld metal in Figure 3 (mass%)

Position	Si	Mn	Al	Cr	Fe
a	0.93	1.76	4.31	9.00	84.0
b	0.96	1.75	4.35	9.13	83.8
c	0.94	1.76	4.30	9.32	83.7

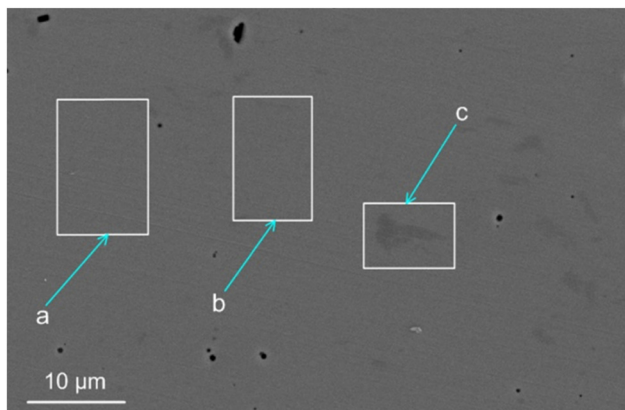
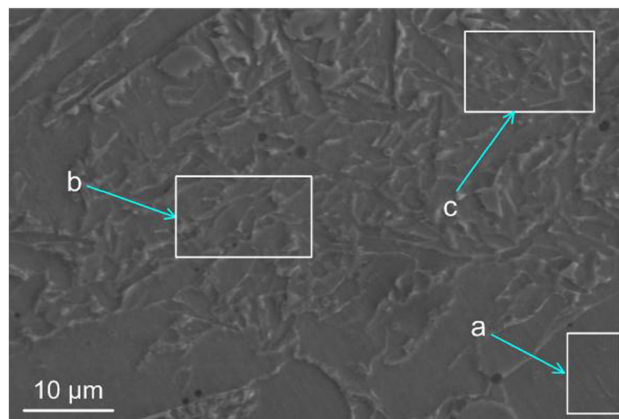
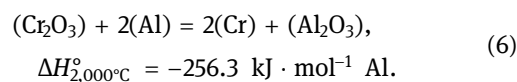
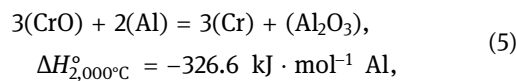
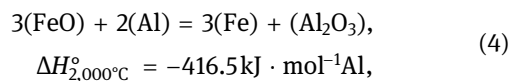
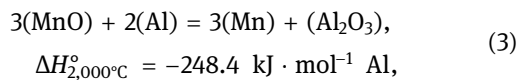
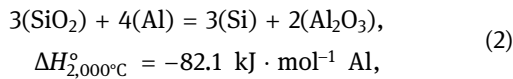
**Figure 3:** SEM micrograph of MP weld metal with analysed areas a, b and c (×4,530).**Figure 4:** SEM micrograph of BC weld metal with analysed areas a, b and c (×4,500).

Table 5: SEM-EDS analyses of marked areas in the BC weld metal in Figure 4 (mass%)

Position	Si	Mn	Al	Cr	Fe
a	0.29	1.42	nd*	nd	98.29
b	0.33	1.43	nd	nd	98.24
c	0.56	1.36	nd	nd	98.08

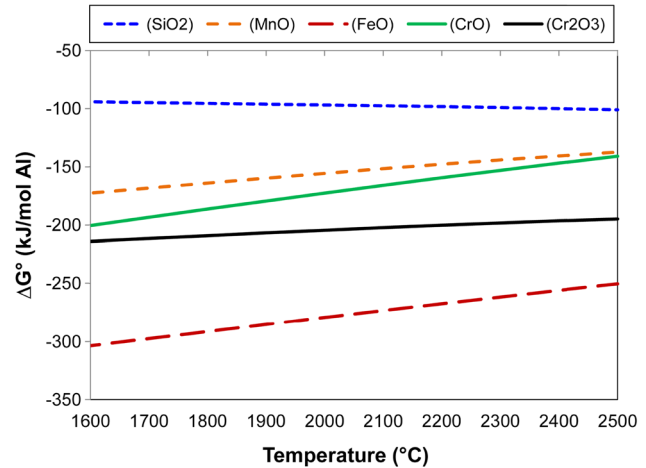
*nd = not detected.

increased solids (spinel) added to the slag from the Al reduction reactions. Most importantly, sufficient reducing conditions were maintained in the presence of Al powder to dissolve large quantities of Cr powder into the weld pool, and not oxidise the Cr powder to Cr-oxides at the molten flux-weld pool interface. The ppm O in the MP weld metal is lower at 299 ppm O compared to the BC weld metal ppm O of 499 ppm. Weld metal oxygen content is set by controlling the oxygen potential at the molten flux-weld pool interface. Several previous studies confirmed that excess oxygen transferred from the arc cavity reacts with the molten steel at the plasma-weld pool interface to form FeO. This FeO ends up in the molten flux. The correlation of increased FeO in the molten flux with increased weld metal ppm O is well known [4,16,21–24]. The oxygen potential prevailing at the molten flux-weld pool interface, as presented by the quantity of FeO, is reduced in this work by adding Al powder as in reaction (4).



(): liquid

Figure 5 illustrates the thermodynamic order of the possible Al-oxide reactions (2)–(6), at the molten flux-weld pool interface according to data from FactSage 7.3. It is seen that FeO will react first with Al, then Cr_2O_3 , CrO, MnO, and SiO_2 at 2,000°C, the accepted typical weld pool temperature [2]. All the reduction reactions, reactions (2)–(6), are exothermic and will add heat into the weld

**Figure 5:** Gibbs free energy plot for reactions (2)–(6).

pool. Reduction of FeO by Al serves to create sufficiently reducing conditions at the molten flux-weld pool interface to ensure Cr remains in the metallic state and is not oxidised from the weld pool to CrO and Cr_2O_3 . Adding Cr-oxides into the flux for transfer of large quantities of Cr from the molten flux to the weld pool, in the absence of de-oxidisers such as Al, is likely not thermodynamically viable. This is due to the high oxygen potential at the molten flux-weld pool interface as evidenced by high FeO content analysed in molten flux from SAW work [4,16,21–24].

The reaction flow diagram in Figure 6 illustrates the reaction sites and reaction sequence in the SAW process. Figure 6 corresponds largely to our diagram for Ti-Al powder additions in SAW [25]. Reaction steps A to E1 are as presented from previous works. It is well established that an excessive quantity of oxygen is initially added to the molten weld wire droplets from the arc cavity gas phase, up to 2,000–3,000 ppm O [26,27]. This initial oxygen quantity in the metal droplets is sourced from the decomposition of less stable oxides at the high temperatures prevailing in the plasma arc, and so flux chemistry is used to manage oxygen transfer to the weld metal from the plasma arc [15]. The plasma arc stability of oxides was determined in welding tests made with binary CaF_2 -oxide flux mixtures, from high to low stability: CaO , K_2O , Na_2O and TiO_2 , Al_2O_3 , MgO , SiO_2 , and MnO [15]. Accordingly, from the flux composition in Table 2, the flux oxides of MnO, SiO_2 , MgO , and Al_2O_3 may all dissociate in the plasma arc to release oxygen in the arc cavity for adsorption onto the molten weld wire droplets, reactions A to C in Figure 6. Interestingly, in spite of the added Al and Cr powders in the weld pool, the weld metal oxygen content was lowered but remained significant at 299 ppm O.

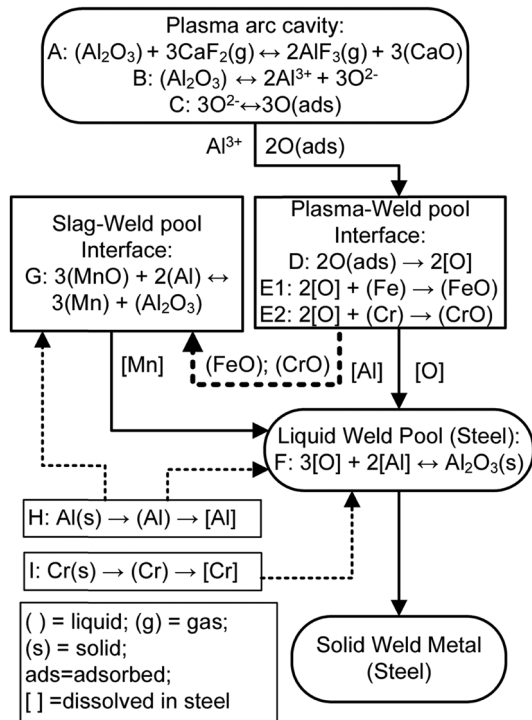


Figure 6: SAW reaction flow diagram with Al and Cr powder additions.

From this relatively high ppm O level of 299 ppm, it appears that the weld metal oxygen content is not set by equilibrium de-oxidation reactions when Al and Cr metal powders are added to the flux-weld pool interface. Because of the high Al and Cr contents in the weld metal as displayed in Table 3, the equilibrium dissolved ppm O in the weld metal should be much lower as referenced for data reported for similar chromium containing alloys at less than 30 ppm O for 1% Al in the alloy at 1,600°C [28]. Gas-slag-metal equilibrium has been illustrated to hold for SAW in the absence of metal powders as discussed elsewhere [24,29]. The same mechanism of oxygen transfer from the arc cavity to the weld pool appears to hold for the instance of Al and Cr metal powder addition since relatively high ppm O in the MP weld metal is attained, despite high Al and Cr contents in the weld metal.

Because Cr has a higher affinity for oxygen, compared to Fe, it is expected that CrO will form at the plasma-weld pool interface where excess oxygen is added into the weld pool to react with dissolved elements in the weld pool. This reaction of CrO formation, reaction (E2), is similar to the FeO formation reaction (E1), in which excess oxygen, transferred from the arc cavity, reacts with the molten steel at the plasma-weld pool interface to form FeO. This FeO ends up in the molten flux. The correlation of increased

FeO in the molten flux with increased weld metal ppm O is well known [4,16,21–24].

The oxygen potential prevailing at the molten flux-weld pool interface, as presented by the quantity of FeO and/or CrO, is reduced in this work by adding Al powder. Similar to reaction G for reduction of MnO from the molten flux, FeO is also reduced by Al, reaction (4). If any CrO or Cr_2O_3 is formed at the plasma arc-weld pool interface, these oxides would be reduced to Cr metal according to reactions (5) and (6). Therefore, the reduced oxygen potential at the molten flux-weld pool interface prevents oxidation of Cr powder to Cr-oxides, and ensures that the Cr is melted and dissolved into the weld pool, reaction I in Figure 6. The reduction of the oxides with Al via reactions (2)–(6) is exothermic. The exothermic heat released from reactions (2)–(6) at the molten flux-weld pool interface facilitates melting and dissolution of Cr powder into the weld pool. Since there is an excess of Al added, some of the Al also dissolves into the weld pool, reaction H in Figure 6. The results presented here confirm that the oxygen potential at the molten flux-weld pool interface may be lowered by Al powder addition to increase Cr powder transfer into the weld pool, without interfering with oxygen transfer from the plasma arc to the weld pool.

5 Conclusion

- The molten flux-weld pool interface oxygen potential may be lowered by Al powder addition to the weld pool to improve Cr powder transfer to the weld pool, without interfering with oxygen transfer from the plasma arc to the weld pool.
- The application of Al powder to the SAW process does not interfere with oxygen transfer from the plasma arc to the weld pool, but rather modulates the weld pool total oxygen content to lower levels of ppm O as compared to standard SAW without Al powder addition.

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Conflict of interest: Authors state no conflict of interest.

Data availability statement: The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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