WELDING RESEARCH

SUPPLEMENT TO THE *WELDING JOURNAL*, MARCH 1989

Sponsored by the American Welding Society and the Welding Research Council

Submerged Arc Welding: Evidence for Electrochemical Effects on the Weld Pool

It is hypothesized that weld metal chemistry is significantly affected by an electrochemical reaction at the weld pool-flux interface

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ABSTRACT. Compositional changes in weld metal from welds made by submerged arc welding of steel using CaF_2-CaO-SiO_2 fluxes are consistent with an electrochemical mechanism in which the filler wire is anodically oxidized to form oxides and fluorides, and metals are cathodically deposited at the weld pool-flux interface. This speculative mechanism, if correct, could make it simpler to predict flux compositions, which will improve the quality of welds and control weld metal microstructures.

Introduction

During the past two decades, the steel industry has experienced tremendous technological progress leading to many alloys possessing excellent mechanical and corrosion-resistant properties. Recent developments in, for example, HSLA steel plate manufacturing technology (Refs. 1, 2) have called for new formulations of welding consumables to produce weld metal deposits with strengths and toughness essentially equivalent to the base metal.

It is well established in submerged arc welding (SAW), as in steel making, that low oxygen levels usually lead to better toughness (Refs. 3–5). There seems to be general agreement that a microstructure normally consisting of acicular ferrite (Fig. 1) (Ref. 6) will yield the best weld metal mechanical properties in terms of strength and toughness, by virtue of its small grain size (typically 1 to 3 μm) and high-angle grain boundaries (Ref. 7).

The microstructural changes taking place in the weld metal on cooling through the critical transformation temperature are, in principle, similar to those occurring during heat treatment of steel. However, because of the large nonisothermal behavior encountered in the welding processes, the circumstances affecting the phase transformations in a weld are significantly different from those occurring in steel production. For instance, the nonmetallic inclusion volume fraction in low-carbon steel welds is much larger than that of normal steel products, due to the very short time available for growth and flotation of such particles (Ref. 8). For a given welding flux, the final weld metal oxygen content has been observed to be directly related to the inclusion population (Ref. 9). On the other hand, oxide inclusions are known to strongly influence the transformation from austenite to ferrite, both by restricting the austenite grain growth (Fig. 2), as well as by becoming nucleation sites for different ferrite morphologies.

High-temperature ferrite products, such as grain boundary ferrite (GF) and polygonal or blocky ferrite (PF) are predominant in welds with high inclusion densities; while welds with low inclusion densities have microstructures consisting primarily of lower temperature transformation products, acicular ferrite and the aligned martensite-austenite-carbide (AC) structure. These microstructures are shown in Fig. 3 (Ref. 10).

KEY WORDS

Submerged Arc Welding
Electrochemical Effects
Weld Pool Chemistry
CaF_2-CaO-SiO_2 Fluxes
Anodic Oxidation
Weld Metal Composition
Composition Changes
Oxidized Welding Wire
Cathodic Deposits
Flux Predictions

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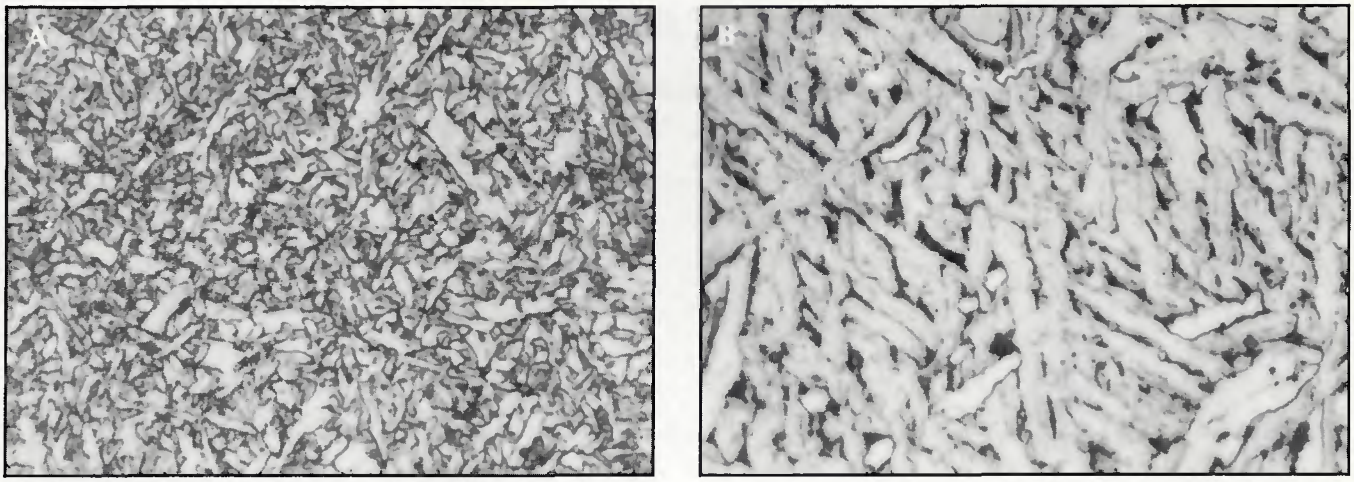

Fig. 1—Acicular ferrite morphology in a low-carbon-steel weld microstructure. A—A low-heat-input weld, 50.3 kJ/in. (1.98 kJ/mm); B—for a high-heat-input weld, 97.4 kJ/in. (3.83 kJ/mm). 25 μm

In view of the significant role of oxygen on the austenite-to-ferrite transformation of low-carbon steel welds, and, consequently, its effect on notch toughness, an understanding of the mechanisms that control the oxygen, in particular, and the chemistry of the weld metal, in general, is important for producing high-quality welds.

Much prior work on weld chemistry has focused primarily on kinetically limited thermodynamically driven reaction mechanisms. This pyrometallurgical approach has proven to be useful in analyzing some data on weld metal chemistry. However, such analyses have not yet led to methods of predicting the chemistry of weld metal. In this paper, we consider the possible importance of electrochemical effects on the chemistry of weld metal produced in submerged arc welding (SAW) with CaF_2-CaO-SiO_2 fluxes.

Thermodynamic models have been proposed to predict the final composition of submerged arc welds (Refs. 11–17). In view of the very high temperatures involved and small molten volumes, some investigators (Ref. 11) assume that equilibrium is attained. Davis and Bailey (Ref. 11) propose that the transfer of elements between the slag and the weld metal depends on the oxide activities in the slag, which are directly connected to the basicity index (BI) of the slags. Limited measurements have indicated that the higher the basicity indexes, the lower the oxygen content of the weld metal. The BI is, however, a poor measure of the chemistry of slags. In addition, it is probable that equilibrium is not attained and that kinetic factors are important. For example, Thier and Dring (Ref. 12) proposed a diffusional model to predict the final content of elements in the weld metal. Thier and Dring concluded that the slag composition where an element was not transferred to the metal, the neutral point, is affected only by the current, but not by the voltage changes and that the neutral point is characteristic for a specific type of flux.

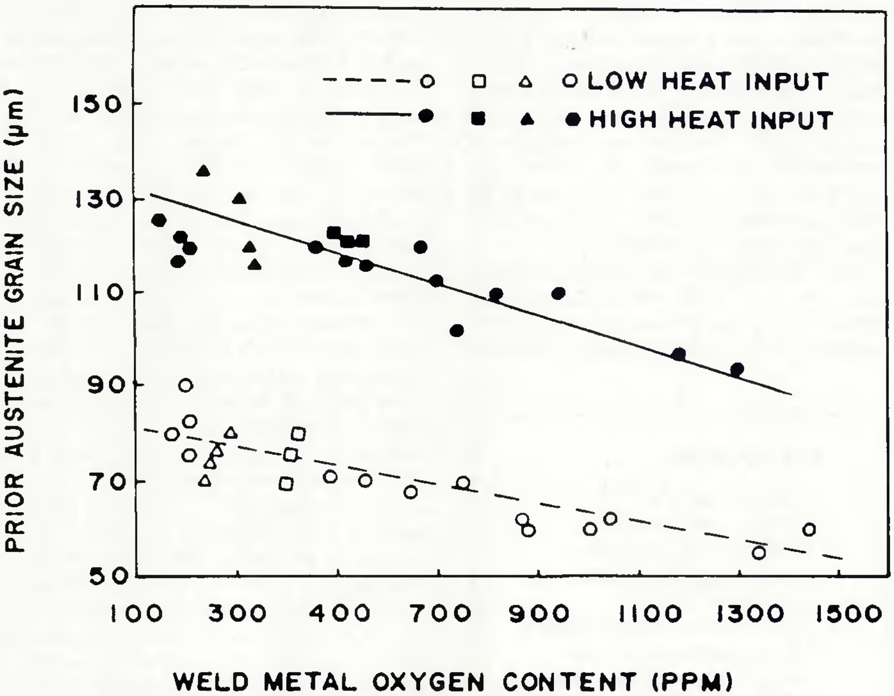

Fig. 2—Prior austenite grain size as a function of oxygen content in the weld metal for two heat inputs (Ref. 6). The "prior austenite" is the parent phase for the lower temperature microstructural products

Ekström and Olson (Ref. 14) reported that the change in Si in weld metal is influenced by the basicity of the slag and that this influence is higher when the basicity index is less than two. Dallam, *et al.* (Ref. 15), found that while the Si level in the weld metal was correlated with the basicity index, the Mn content of the weld depends on the amount of MnO in the slag. Indacochea, *et al.* (Ref. 16), qualitatively showed a similar correlation between flux and weld metal composition.

Despite qualitative agreement among researchers regarding the flux type and direction of elemental transfer, there is no precise determination of a "neutral point," the composition where a given element is not transferred, even though very similar types of fluxes were used in several of these studies. This failing may be attributed to the possibility that the mechanism of elemental transfer is not so simple, to the influence of different welding parameters used, and to differences in the wire and flux compositions.

It is clear that essentially all prior studies have been relatively narrowly defined. A more comprehensive approach is needed in which the diverse kinetic and thermodynamic factors involved in SAW

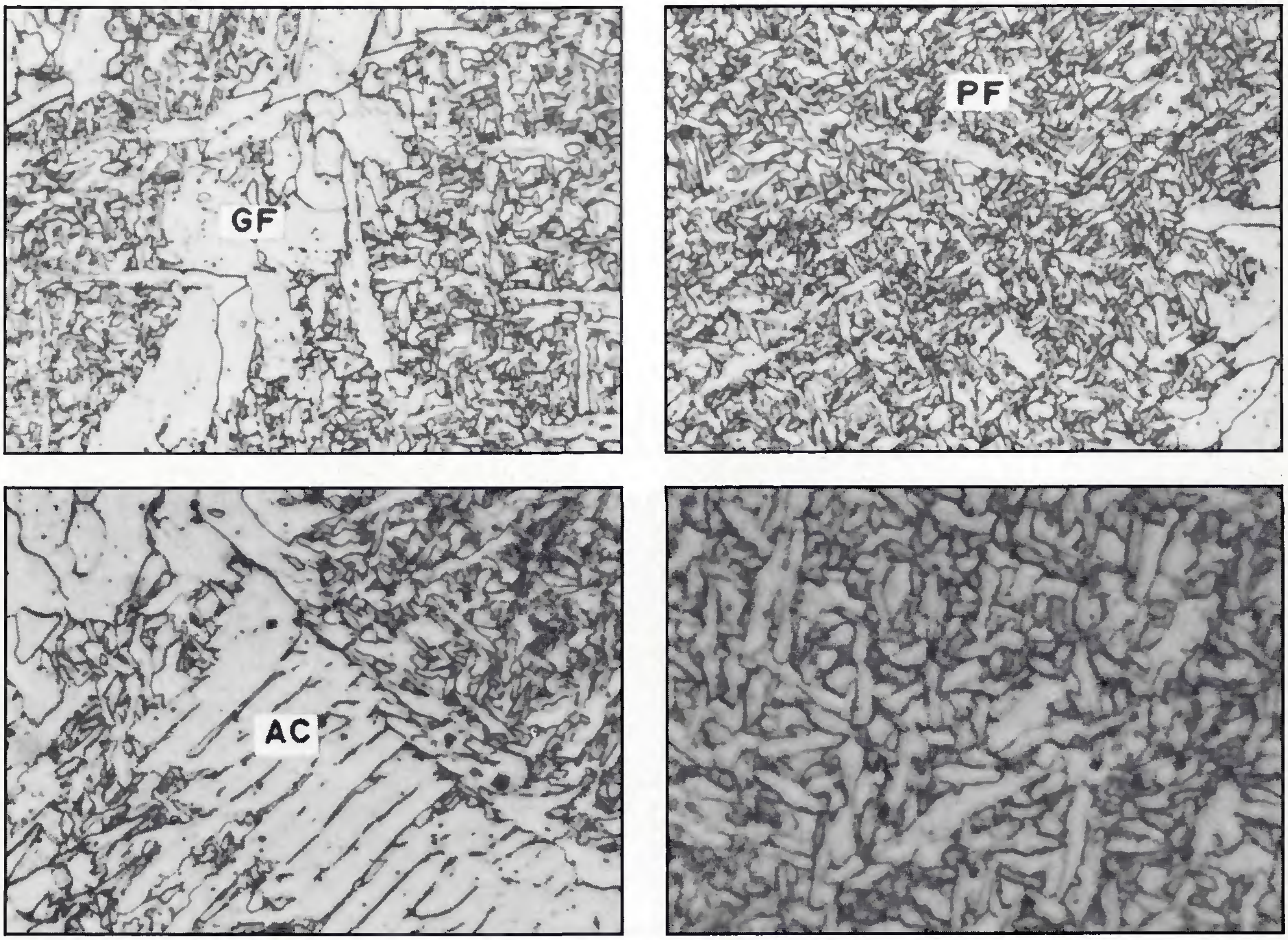

Fig. 3 — Typical ferrite morphologies present in low-carbon-steel weld microstructures. GF — Grain boundary ferrite; PF — polygonal or blocky ferrite; AC — aligned M-A-C (martensite-austenite-carbide); and acicular ferrite in the unlabeled micrograph (Ref. 6)

(Refs. 16, 17) are examined and which also explores the possibility of mechanistic factors that have not been considered. Such an approach is made difficult by the complexity of the processes in SAW. The DC voltages and currents are very high (*e.g.*, 30 V, 400 A) and lead to a system containing the four principal phases, a weld wire, a molten flux, a plasma arc and a weld pool with five interfaces among them. Most of the current is transported by electrons in the plasma from the generally cathodic weld pool to the generally anodic weld wire (or filler wire). The electrons heat the continuously fed wire to melt it and form droplets that fall through the molten flux and the plasma. In addition, the temperatures are high, and there are also large temperature gradients, as well as large temporal changes in temperature as the weld wire position is moved along the weld. An attack on such a complicated problem requires some simplification.

One way to simplify the approach to such a complex system is to isolate the different parts of the problems. For example, in this paper, we chose to examine electrochemical effects, which have not been considered before. All welding fluxes when molten are, at least in part, ionic and the number of coulombs passed per mole of metal is very large. If even a small fraction of the total current (≤ 0.01) is involved in a Faradayic process, electrochemical effects at metal-flux interfaces could be a major factor controlling the chemistry of weld metal (Ref. 18). In addition, because the very hot plasma is essentially electrically neutral and in contact with the relatively volatile fluxes, there have to be at least as many positive ions as there are electrons and other negatively charged particles. Stable negative and positive ions formed from flux evaporates could all participate in Faradayic processes. Thus, there could also be electrochemical reactions at the plasma-metal interfaces (Ref. 18).

In this paper, we examine the possibility that the changes in chemistries of weld metal with changes in the compositions of CaF_2-containing fluxes might be related to an electrochemical mechanism.

Experimental Work

Materials

Low-carbon steel coupons of dimensions 63.5 × 254.0 × 12.7 mm (2.5 × 10 × 0.5 in.) and a 2.38-mm (0.094-in.) diameter filler welding wire (AWS Type ER70S-3) were used to make the weld. The compositions for the base plate and the welding wire are shown in Table 1.

The experimental fluxes were made from reagent-grade SiO_2, CaF_2 and CaO powders. The total of 12 different flux

Table 1—Base Metal and Filler Metal Compositions (wt-%)

Material	C	Mn	P	S	Si	Ti	Al	O ppm
Base metal	0.04	0.37	0.012	0.022	0.03	0.003	0.001	220
Filler metal	0.09	1.18	0.007	0.013	0.58	0.002	<0.008	195

Table 2—Nominal Flux Compositions (wt-%)

Flux Sample	SiO_2	CaF_2	CaO
S11	40	10	50
S12	40	20	40
S13	40	30	30
S14	40	40	20
S15	30	30	40
S16	30	40	30
S17	30	50	20
S18	30	60	10
S19	20	40	40
S20	20	50	30
S21	20	60	20
S22	20	70	10

Table 3—Welding Process Parameters

Voltage	33 V	31 V
Current	600 A	330 A
Travel speed	12.2 ipm	12.2 ipm
Wire speed	75 ipm	40 ipm
Heat input	97.4 kJ/in. 3.83 kJ/mm	50.3 kJ/in. 1.98 kJ/mm

compositions listed in Table 2 were used in this study. All the flux samples were accurately weighed, mixed thoroughly for 2 h, melted in a graphite crucible using a 50-kW induction furnace, and then poured into a vessel containing tap water. This quenching procedure caused the flux to break into small pieces which were dried at 260°C (500°F) for 2 h before being crushed further and sized between −14 and 200 mesh.

In the last step before welding, all fluxes were baked at 700°C (1292°F) for 3 h to burn off the carbon that might have been picked up during melting.

Welding Procedure

The welds were single-pass, laid on top of the test coupons in a horizontal position. The welding was done automatically with a programmable microprocessor controlling the power supply and amount of filler metal deposited. The electrode extension and electrode-to-workpiece distance were maintained in all the welds produced. Two heat inputs were used, and the corresponding welding processing parameters are given in Table 3. The stability of the arc as a function of flux composition and heat input was monitored during welding. It was found that the stability of the arc decreased as the level of silica decreased from 40 to 20%. In addition, the welds produced with the high-heat inputs showed greater arc stabilities than the low-heat input welds.

Chemical Analyses

The analyses for oxygen were performed using a LECO oxygen analyzer. For this, the weld samples were extracted from the middle of the weld metal. For the other chemistries, atomic absorption was used for the filler metal and emission spectrometry was employed for the weld specimens. The compositions of all the weld samples are summarized in Table 4.

Electrochemical Mechanisms

In SAW, the filler wire is generally anodic and the weld pool is cathodic. Current is carried between these metals by the arc plasma and by the molten flux, with the arc plasma carrying most of the current.

At the interface between flux and cathodic weld pool, metals in the flux tend to deposit by reactions such as

$$Ca^{2+}(\text{flux}) + 2e \rightarrow Ca\ (\text{metal})$$
$$Si^{4+}(\text{flux}) + 4e \rightarrow Si\ (\text{metal}) \quad (1)$$

The products, which are formed metastably at the interface, tend to simultaneously diffuse into the metal and back react with the slag components by reactions such as

$$SiO_2 + 2Ca \rightleftharpoons 2CaO + Si \quad (2)$$

Thus, the balance between the different elements that might be deposited is controlled by the relative rates of the electrodeposition process, of the back reactions (or volatilities), and of the diffusive processes that carry the interfacial materials away from the interface. Because of the high volatility of Ca and its relative insolubility in steel, Ca is likely to vaporize through the flux and to partly react according to Equation 2. In addition, Ca is soluble in CaF_2-rich melts.

The most important anodic reaction is

$$nO^{2-} + M\ (\text{metal}) \rightarrow MO_n + 2ne \quad (3)$$

where M is a metal at the weld wire-flux interface and 2n is the valence of the metal in the oxide. Fluoride is probably

Table 4—Chemical Analysis of Weld Specimens

Weld[a] Sample	C	Mn	P	S	Si	Ti	Al	O (ppm)
	Composition (wt-%)							
S11L	0.088	0.53	0.010	0.015	0.54	0.002	<0.008	400
S12L	0.084	0.46	0.012	0.017	0.59	0.002	<0.008	432
S13L	0.076	0.34	0.012	0.020	0.65	0.002	<0.008	405
S14L[b]				No Sample				
S15L	0.106	0.58	0.010	0.013	0.40	0.002	<0.008	240
S16L	0.094	0.45	0.010	0.016	0.51	0.002	<0.008	262
S17L	0.083	0.36	0.010	0.017	0.59	0.002	<0.008	289
S18L	0.094	0.41	0.010	0.016	0.51	0.002	<0.008	250
S19L	0.110	0.69	0.009	0.007	0.35	0.002	<0.008	207
S20L	0.104	0.64	0.009	0.009	0.35	0.002	<0.008	173
S21L	0.101	0.53	0.009	0.014	0.38	0.002	<0.008	204
S22L	0.092	0.52	0.009	0.014	0.54	0.002	<0.008	196
S11H	0.100	0.50	0.011	0.018	0.35	0.002	<0.008	500
S12H	0.092	0.42	0.010	0.018	0.40	0.002	<0.008	554
S13H	0.090	0.40	0.009	0.018	0.43	0.002	<0.008	521
S14H[b]				No Sample				
S15H	0.105	0.53	0.009	0.014	0.30	0.002	<0.008	243
S16H	0.099	0.46	0.010	0.017	0.37	0.002	<0.008	326
S17H	0.100	0.40	0.009	0.015	0.38	0.002	<0.008	336
S18H	0.096	0.42	0.009	0.016	0.36	0.002	<0.008	311
S19H	0.099	0.68	0.009	0.008	0.28	0.002	<0.008	191
S20H	0.106	0.63	0.010	0.008	0.30	0.002	<0.008	154
S21H	0.106	0.50	0.010	0.014	0.33	0.002	<0.008	201
S22H	0.102	0.50	0.009	0.013	0.40	0.002	<0.008	190

(a) L designates low heat input; H designates high heat input.
(b) Excessive weld porosity.

also involved in an anodic reaction, but forms relatively volatile products, which need not be considered here. Because of large overpotentials, all of the metals at the interface, principally Fe, will oxidize. The less noble elements in the alloy, such as Mn and Si, will diffuse toward the interface and react with the FeO by reactions such as

$$FeO + Mn \rightleftharpoons MnO + Fe \quad (4)$$
$$2FeO + Si \rightleftharpoons SiO_2 + 2Fe$$

Also, some oxide will diffuse into the metal and some will dissolve in the flux. The overall effect is to greatly increase the oxygen content of the weld wire tip and to decrease the less noble elements, such as Mn and Si, somewhat. When heated for a sufficiently long time, the tip forms a droplet which will have a high oxide content near the surface and tend to lose some of its less noble elements to the flux. Measurements of such droplets by Lau, *et al.* (Refs. 19, 20), in $CaO-Al_2O_3$ based fluxes have shown their very high oxygen contents.

When these oxide-rich droplets fall into the weld pool, the oxides can react with the more active metals, including those that have been cathodically electrodeposited at the surface of the weld pool. In the measurements of Lau, *et al.* (Refs. 19, 20), a large fraction of the oxide associated with the droplets is removed before being incorporated in the weld, probably by back reactions with the most active elements in the weld, such as Ca or Si (or Al in their measurements).

An understanding of the electrochemistry of the plasma is limited by the sparsity of information on plasma species. Most of the current is carried by electrons. However, because of the constraint of approximate electroneutrality of the dense plasma, the number of positive ions is at least as large as the number of electrons, and these ions can carry a small but significant fraction of the current. Metals such as Fe, Ca and Mn are relatively volatile and are probably ionized to form positive ions. Oxygen can be present as O^+ and possibly as O^-, and the presence of some oxyions ions derived from SiO, SiO_2, FeO or MnO vapor molecules is likely, but there is no information on numbers or distributions of such positively and negatively charged ions. All of these ions can participate in Faradayic processes, which will, to a first approximation, parallel the Faradayic processes involving the slag.

Interpretation of the Experiments

As can be seen in Fig. 4, the silicon contents of the weld metals are generally higher than the nominal values expected from simple mixing of roughly equal amounts of the base metal and filler wire. These silicon contents increase with an increase in the SiO_2 content of the CaF_2-CaO-SiO_2 mixtures, but decrease with an increase in the current density. Since the silica in the fluxes tends to form anionic species (chain silicates or fluorosilicates), it is probable that the kinetics of direct electrochemical deposition of silicon at the cathodic weld pool is not as favorable as that for the deposition of the volatile Ca. Consequently, a small amount of Si would be directly formed electrochemically, but more is likely to form by the evaporation of Ca, which would then reduce SiO_2 in the flux to Si. Later back reactions of the Si with the oxides on weld wire droplets entering the weld pool would reduce the total amount of metallic Si produced by this mechanism. The higher the ratio of CaO to CaF_2 in the melt, the greater the amount of oxide produced at the weld wire tip and the greater the amount of Si which would be reoxidized and redissolved in the flux. Of course, one also expects that the amount of metallic Si produced would be larger the greater the SiO_2 content of the flux. This is consistent with the results in Fig. 4. The decrease of Si content in weld metal with an increase of current could be understood by any one of a number of possible mechanisms, which are too numerous to discuss here. Further work on the effect of current is needed to completely define the mechanism, which probably involves concentration polarization of silica and diffusion-controlled deposition of Si.

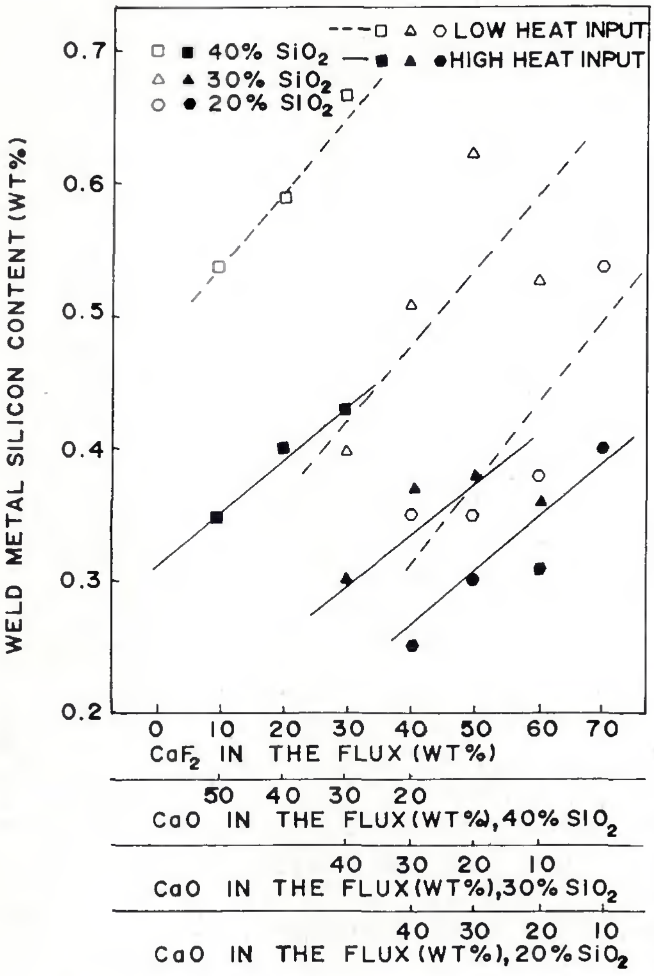

Fig. 4 — Silicon content in the welds produced with the SiO_2-CaO-CaF_2 submerged arc welding fluxes for two heat inputs

As can be seen from Fig. 5, the manganese content of the weld metal is fairly close to that expected for mixtures of weld wire and base plate metal for the lowest contents of SiO_2 in the flux. With an increase in the concentration of SiO_2, one would expect an increased amount of oxide produced at the anode and a larger loss of Mn from the filler wire, which contains the majority of the Mn expected in the weld. This can lead to the observed decrease in Mn with increased SiO_2 in the flux.

The compositional changes of most concern are those related to oxygen. The physical properties and quality of welds are strongly influenced by the amount of oxygen. Figure 6 exhibits the changes in oxygen content for the different flux compositions. It is obvious that the concentration of oxygen in the weld decreases with a decrease in the amount of silica in the flux. For the lowest concentrations of silica, the concentration of oxygen differs little from that of the

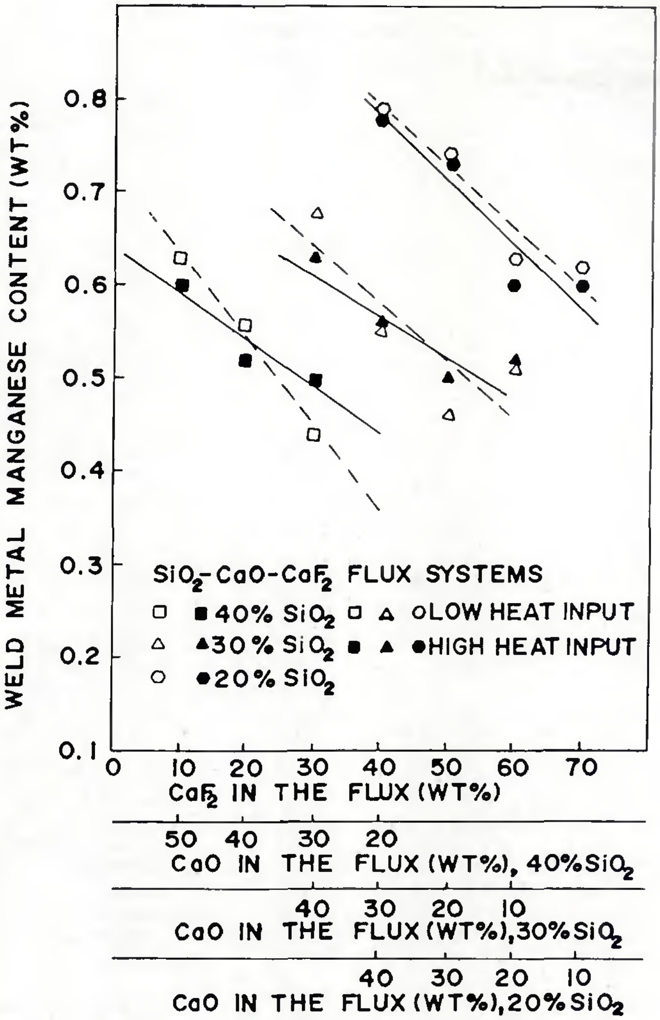

Fig. 5 — Manganese content in the welds produced with the SiO_2-CaO-CaF_2 submerged arc welding fluxes for two heat inputs

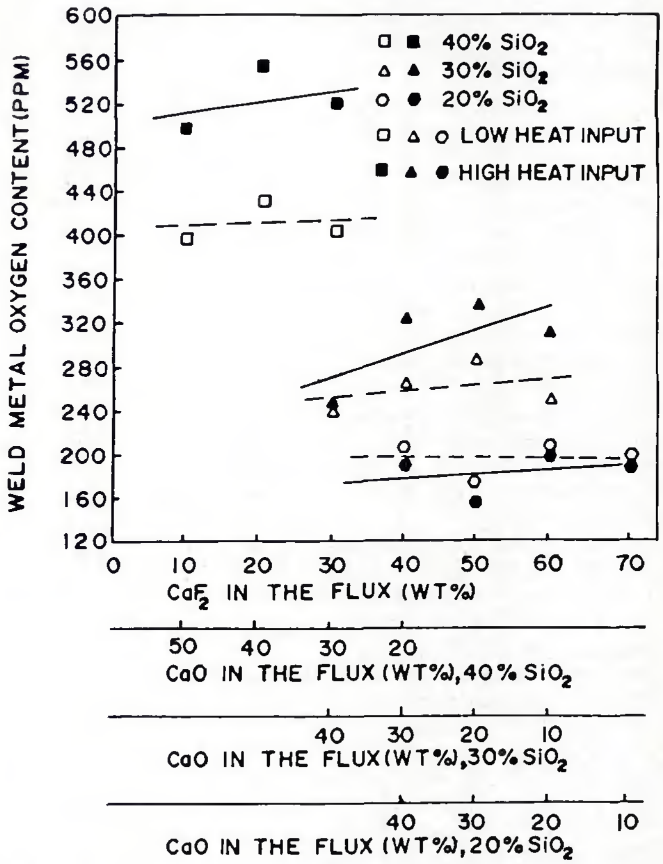

Fig. 6 — Oxygen content in the welds produced with the SiO_2-CaO-CaF_2 submerged arc welding fluxes for two heat inputs

original metals (~200 ppm). This difference is consistent with the electrochemical mechanism we propose. It was noted earlier that flux composition affected the arc stability. An unstable arc could substantially increase the oxygen level in the weld metal; however, our results show that the low silica flux produces the most unstable arc, and yet these welds have the lowest oxygen levels. If anything, a more stable arc with the low silica flux might have led to even less oxygen in the welds.

Discussion

The measurements we present are consistent with the speculation that an electrochemical mechanism is important in altering the chemistry of weld metal in SAW. In prior work (Ref. 18), examination of the effects of this mechanism on weld metal compositions measured by Indacochea, *et al.* (Ref. 16), has indicated that the contents of Mn, Si, O and other elements in welds produced from SiO_2-FeO-MnO fluxes were also consistent with an electrochemical mechanism. However, a thermodynamically driven, kinetically constrained mechanism may also be possible if the slag-metal interface is at very high temperatures (> 2200 K). Heat flow considerations make this high of an average temperature at the flux-metal interface unlikely. A test of the correctness of our speculations will ultimately require much more detailed measurements. One set of such measurements has been made by Lau, *et al.* (Refs. 19, 20), who has shown that metal droplets from the filler wire are very high in oxygen, which is largely present as FeO-, MnO- and SiO_2-containing inclusions in the metal (Ref. 20); these could not have been formed by a thermodynamic mechanism. We are planning measurements of analogous microscopic compositional details to test the validity of our speculations.

The long-term goal of our work is to integrate our findings on this electrochemical mechanism with a comprehensive analysis of the important thermodynamic (Ref. 16) and kinetic (Ref. 17) factors. Although consideration of these factors and of the welding parameters (current, voltage, wire feed rate, welding speed) simultaneously is very difficult and too complex for quantitative predictions, our results to date have led to useful qualitative predictions on flux compositions that should improve the properties of welds. If an electrochemical mechanism can be shown to be important, the deduction of methods for predicting and controlling weld chemistry would be greatly simplified.

Acknowledgments

The authors wish to acknowledge the financial support provided by the Engineering Foundation for the work performed at the University of Illinois at Chicago (Grant R.I.-A-84-4), and the Office of Naval Research for the work performed at Argonne National Laboratory under Navy Order No. N00014-87-F-0064. In addition, the authors would like to thank Inland Steel Co. for the donation of material and the chemical analyses.

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