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아아크 용접중 아아크 프라스마와 용융 금속간 산소와 질소의 거동에 관한 연구

Transfer of Oxygen and Nitrogen between Arc Plasma and Molten Metal during Arc Welding

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국문요약

직류 아아크 용접시 아아크 프라스마와 용융 금속 사이의 계면에서 일어나는 전기 화학 반응이 용착 금속 내의 산소와 질소의 함량에 미치는 영향이 고찰되었다. 열 화학 반응 뿐만 아니라 전기 화학 반응도 용접부의 산소 및 질소의 함량을 결정하는 중요한 반응 메카니즘이라는 것과 전기 화학 반응의 양을 결정하는 것은 단위 면적당 통과하는 전류의 양을 조절하는 용접 전류와 용접 속도라는 것이 실험적으로 입증되었다. 따라서 이 연구 결과는 아아크 용접시 적당한 용접 조건의 선택 뿐만 아니라 용접 재료의 설계 또는 선택에 중요한 지침을 준다.

1. INTRODUCTION

Relatively small quantities of oxygen and nitrogen contamination during arc welding may affect the cleanliness, toughness and porosity of the resulting weldment. Generally speaking, these elements should be maintained at as low as possible; except in the case of austenitic stainless steels, where the percentage of delta ferrite may be manipulated by changing the nitrogen content of the weld metal. Therefore, the mechanisms which control the oxygen and nitrogen contents during arc welding must be understood in order to control the process and the resultant mechanical properties of the weldment.

A large number of welding processes use the electric arc as the source of heat for fusion, because the heat of the arc may be effectively concentrated and controlled. The welding arc can be defined as a particular group of electrical discharge that is formed and sustained by the de-

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velopment of a gaseous conduction medium (plasma). The main function of the welding arc is to produce a high intensity heat, which is useful for joining metals by fusion, from electrical energy. At the same time it produces a bright light, noise, and, in a special case, bombardment that removes surface films of the base metal.

The potential importance of the arc plasma, with respect to plasma-metal reactions during arc welding, is obvious. The amount of energy supplied by the arc, the distribution of that energy, the characteristics of arc plasma, and the effects of welding process parameters and materials employed on those properties are of fundamental importance in determining the amount and direction of plasma-metal reactions. Chemical changes in the arc plasma, whether resulting from differences in either cover gas, flux or metal vapor composition produce easily measurable changes in weld metal chemistry. Also, changes in arc properties, due to changes in welding process parameters and polarity, produce considerable changes in weld metal chemistry.

There is not a great deal of agreement on what constitutes a welding arc. However, a definition proposed by Jackson[1] seems suitable for the purpose of this investigation : "A welding arc consists of sustained electrical discharge through a high temperature conducting plasma, producing sufficient thermal energy so as to be useful for the joining of metals by fusion." Welding arcs are of high conductivity, carrying a large amount of current at a relatively low voltage.

1.1 Nature of Welding Arc Plasma

All arc discharges in gaseous media produce plasmas which are gaseous clouds composed of free electrons, positive ions, neutral atoms and molecules. This is true for a whole range of arc welding processes. The term "plasma" was originally defined by Langmuir et al[2]. for the conducting regions of gaseous electrical discharges, excluding those areas immediately adjacent to the electrode surfaces. The plasma state is now regarded as "the fourth state of matter" which can be obtained by heating a substance in the gaseous state(the third state of matter). A plasma can be defined as[3] : "a highly ionized gas which contains approximately equal numbers of ions and electrons in sufficient density so that the Debye shielding length is much smaller than the dimensions of the gas." The Debye shielding length is a measure of the distance over which charged particles can exert an influence on each other.

A characteristic feature of a plasma is its macroscopic neutrality, which is maintained because of the mutual compensation of the space charge of the positive ions and electrons. This compensation, however, takes place only in terms of averages, that is, in sufficiently large volumes and for sufficiently long time intervals. Therefore, a plasma is said to be a quasi-neutral medium. Figure 1 shows the schematic representation of the arc plasma. Electrons are emitted from the cathode and flow to the anode, while positively charged ions flow in the reverse direction. Those movements of electrons and ions result in the current flow through an ionized gas.

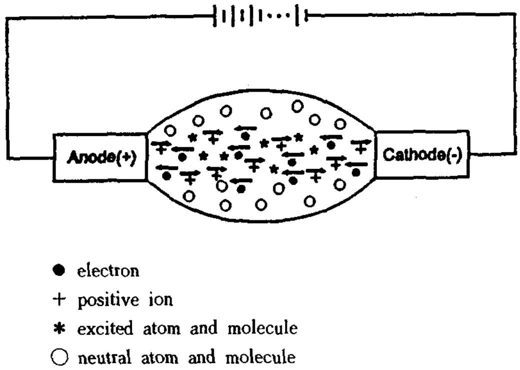

Fig. 1 Schematic Representation of the Conventional Arc Plasma Showing Constituents and Movement of Charged Particles

In arc welding, the plasma is initiated when electrons are accelerated between two electrodes under the application of an electric field. The electrons moving toward the anode collide with and excite the atoms or molecules in the gas. This excitation can cause complete ionization, orbital displacement of an electron, or merely an increase in kinetic energy. The additional electrons produced by ionization are also accelerated toward the anode and these in turn cause further ionization. The species and concentrations of the ionized particles in the plasma control the nature and extent of electrochemical reactions between plasma and molten metal during arc welding.

1.2 Nature of Electrochemical Reactions

The chemistry in steel making tends to be dominated by the thermochemical reactions, because it generally involves large surface areas, low current densities, and alternating current power. However, direct current arc welding processes involve small surface areas, high current densities, and direct current power. Also, the welding current is carried by highly ionized arc plasma and molten slag layer. Therefore, it is to be expected that electrochemical reactions, which result from ionic conduction of the welding current, as well as thermochemical reactions, will exert a significant influence over the final chemistry of the weld metal. The relative influence of these two types of chemical reactions depends on the scale of the process, the power mode, the process parameters, polarity, and the materials employed.

During arc welding the electricity is transferred across the slag/metal and the plasma/metal interfaces. The flow of electricity is due to the movement of charged particles, and the charge transfer across the electrified interfaces implies chemical transformation, i.e., the transformation of substances into other substances. In this study, the electrochemical transfer of oxygen and nitrogen at the plasma/metal interface was investigated. When the interface between the molten metal and the plasma is considered, the electrochemical reactions are relatively simple to understand. This type of interface is present in all kinds of arc welding. At the plasma/metal interface, no anodic electrochemical pickup of anions is expected to occur due to the absence of negatively charged ions in welding arc plasma. Anions can be produced by attachment of electrons to neutral atoms. The probability of attachment increases as the electron energy decreases, because the electron can remain within the sphere of influence of the atom for a longer time. However, the temperature of welding arc plasma is too high to keep the electrons around the atom for a long time. Therefore, the formation of negative ion by electron attachment is difficult in welding arc plasma. Also, at the temperature range of welding arc plasma doubly ionized atoms are less probable than singly ionized atoms due to the high second ionization potential. As a result, the dominant species in welding arc plasma are highly ionized positive ions. Therefore, the possible anodic and cathodic electrochemical reactions for oxygen and nitrogen at the plasma/metal interface are

Anode : $O(metal) \rightarrow O^{+}(plasma) + e^{-}$ (1)

$N(metal) \rightarrow N^{+}(plasma) + e^{-}$ (2)

Cathode : $O^{+}(plasma) + e^{-} \rightarrow O(metal)$ (3)

$N^{+}(plasma) + e^{-} \rightarrow N(metal)$ (4)

The electrochemical reactions at the plasma/metal interface is expected to result in the refining of oxygen and nitrogen at the anode and the pickup at the cathode.

The electrochemical concept was first introduced in welding by Frost et al.[4,5] who have investigated the slag-metal reactions during electroslag welding. They considered the different chemical effects at the anode and cathode buttons. Afterwards, electrochemical reactions have recently been identified to be as important as the thermochemical reactions in direct current submerged arc welding. Initially, Blander and Olson[6] postulated an electrochemical mechanism for the alteration of weld metal composition in direct current submerged arc welding. This postulation has recently been verified experimentally by Kim et al.[7-9] and later by Indacochea et al.[10-12]

The most convincing 'proof' of the electrochemical contributions came from the results of Kim et al.[7-9] They demonstrated the electrochemical effects through chemical analysis of the melted electrode tips, the detached droplets, and the final weld metals for both straight(DCEN) and reverse(DCEP) polarities and concluded that the electrochemical perturbation, followed by partially completed thermochemical back reaction, is also significant in direct current submerged arc welding.

2. EXPERIMENTAL PROCEDURE

The gas tungsten arc welding has been selected as an experimental process, because it has the simplest welding situation among the arc welding processes, and it allows better conditions for the detailed investigation of the electrochemical reactions between the arc plasma and weld metal. This process does not require the consumable electrode and flux, which make such an investigation difficult.

The direct current electrode negative gas tungsten arc welds were made on the low carbon microalloyed steel with constant heat input as a function of welding current and travel speed under the argon gas containing oxygen or nitrogen gas. Also, the stationary gas tungsten arc welds were made on the low carbon microalloyed steel rounds as a function of time and current under the argon gas containing oxygen gas. Figure 2 shows the constructed stationary gas tungsten arc welding machine which has the water cooling system inside the tungsten electrode. Figure 3 shows the initial round sample and welds which were made with straight polarity stationary gas tungsten arc welding. The welding conditions for the conventional and stationary gas tungsten arc welding processes are given in Table 1.

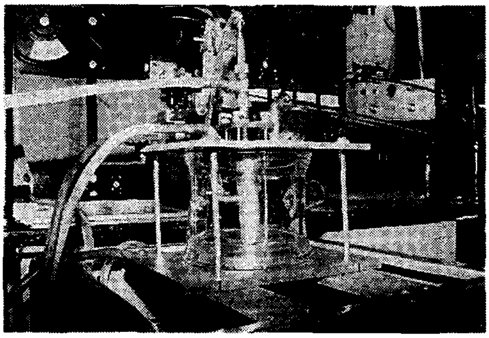

Fig. 2 Photograph of the Apparatus Constructed to Make the Stationary Gas Tungsten Arc Welds; It has the water cooling system at tungsten electrode and copper plate

Fig. 3 Photograph Showing the Initial Sample and the Welds Obtained from Stationary Gas Tungsten Arc Welding

The analyses for oxygen and nitrogen were carried out using LECO Interstitial Analyzer. The weld metal samples were obtained by sectioning out 0.5 to 1.0 gram samples using a vertical saw. The samples were then soaked in hydrochloric acid and cleaned with ultrasonic cleaner for five minutes. After cleaning, the sam-

트륨 주입 온도를 매개변수로 하여 나트륨화재 에어로졸의 aerodynamic diameter와 geometric diameter의 평균크기를 시간에 따라 보여 주고 있다. 나트륨화재 에어로졸의 aerodynamic diameter와 geometric diameter의 평균크기는 용융 나트륨 주입 온도가 높아짐에 따라 감소하는 것으로 나타났다. 이와 같은 경향은 나트륨의 열역학적 특성치들의 온도 의존성에서 기인한 것으로 생각된다. 다른 연구자들에 의하면 용융 나트륨의 밀도, 점도, 표면장력은 온도가 증가하면 감소한다고 보고되어 있다[20]. 그러므로 이와 같은 물성치 변화가 분무노즐에서 용융 나트륨이 분무될 때 같은 압력에서 온도가 높을수록 작은 액적을 생성하게 되어 에어로졸 생성에 직접 영향을 주기 때문인 것으로 생각된다.

Fig. 4는 나트륨화재 에어로졸의 aerodynamic diameter와 geometric diameter의 관계를 보여주고 있다. aerodynamic diameter와 geometric diameter의 관계가 선형적이라는 것은 에어로졸의 겉보기 밀도(apparent density)가 에어로졸 크기의 함수라는 것을 의미한다.

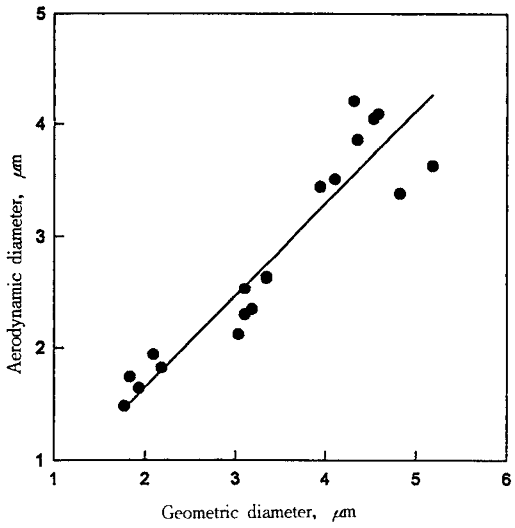

Fig. 4 Aerodynamic diameter vs. Geometric diameter

Fig. 5는 나트륨화재 에어로졸의 aerodynamic diameter와 dynamic shape factor의 관계를 보여 주고 있다. 부유되어 있는 에어로졸 입자의 침강속도(sedimentation velocity)는 에어로졸의 밀도, 크기 등에 영향을 받으며 구형입자(spherical particle)인 경우는 같은 조건에서 크기만의 함수로 특성을 표시할 수 있지만, 비구형 입자(non-spherical particle)인 경우는 dynamic shape factor를 도입하여 설명할 수 있다. dynamic shape factor는 비구형 입자가 어떤 매질속에서 침전 속도, Vt로 움직일 때 받는 drag force와 같은 부피를 갖는 구형입자가 같은 침전속도로 움직일 때 받는 항력(drag force)의 비율로 정의된다[21].

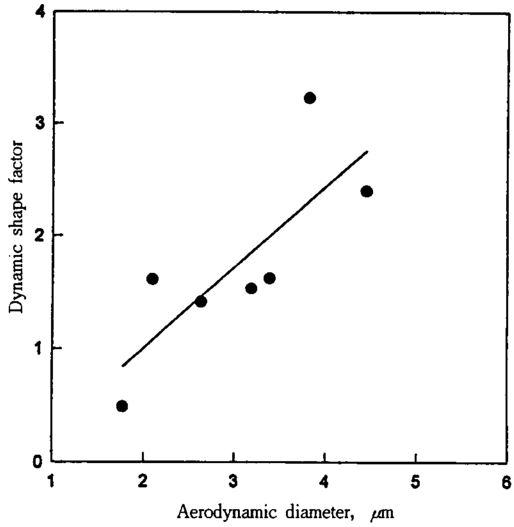

Fig. 5 Dynamic shape factor in terms of aerodynamic diameter

$$K=\frac{\rho D^2_{ge}C(Dge)}{\rho_0 D^2_{ae}C(Dae)} \quad (2)$$

여기서 ρ_0 =unit density, 1g/cm^3

D_{ge} =geometric diameter

D_{ae} =aerodynamic diameter

C(D) =Cunningham slip correction factor

본 연구에서도 식(2)를 이용하여 dynamic shape factor를 구하였다. Fig. 5에서 알 수 있는 것과 같이 나트륨화재 에어로졸의 aerodynamic diameter이 증가할수록 dynamic shape factor의 값도 증가하였다. 이와 같은 연구결과는 M. Barbe-le Borgne를 제외한 다른 연구자들의 연구결과와는 차이가 나는 것으로 본 연구에서 발생한 에어로졸의 크기가 비교적 다른 연구자들의 에어로졸의 크기(2μm이하)보다 크기 때문인 것으로 생각한다. 그러나 M. Barbe-le Borgne는 본 연구와 비슷한 연구 결

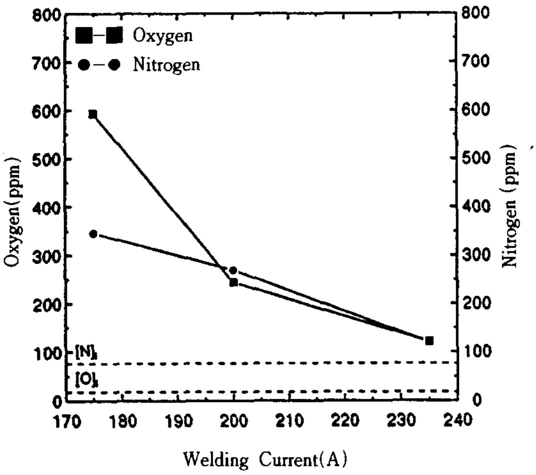

Fig. 5 Variation in the Concentrations of Oxygen and Nitrogen in the Weld for Electrode Negative Polarity Gas Tungsten Arc Welding as a Function of Welding Current

ctrochemical reactions in the arc welding with slag. This behavior is due to the ionization of oxygen atoms into positive ions in welding arc plasma. Oxygen has lower ionization potential than argon and, thus, is ionized preferentially to conduct the welding current.

However, difficulties have arisen in obtaining systematic data in direct current electrode positive polarity, due to melting of the tungsten electrode during welding. When the electrode is positive and the work is negative, electrons are emitted from the work and flow to the electrode where they create intense localized heat. In direct current arc welding, the amount of total energy dissipated near the anode has been estimated to be as high as eighty percent[13,14].

The primary factor in increasing the temperature of the anode is the work function of the atoms on the surface of the anode. As noted previously, the loss of electrons at the cathode is an energy loss, and the cathode is cooled by this thermionic emission. Conversely, the anode is heated as these electrons recombine with the metal lattice, with an increase in temperature corresponding to the decrease in kinetic and binding energy of the electrons. Therefore, the electrode tends to overheat and melt during reverse polarity gas tungsten arc welding, and in addition, the weld metal penetration is not so great since less heat is generated at the work.

To solve this problem, the stationary gas tungsten arc welding system, which has a water cooled tungsten electrode, was constructed and used to make the weld on the low carbon microalloyed steel rounds, as a function of time and current under the argon gas containing oxygen gas. However, the cooling efficiency was not sufficient, and thus, it was difficult to make good welds for reverse polarity.

Even though the data for reverse polarity can not be obtained, the results from the straight

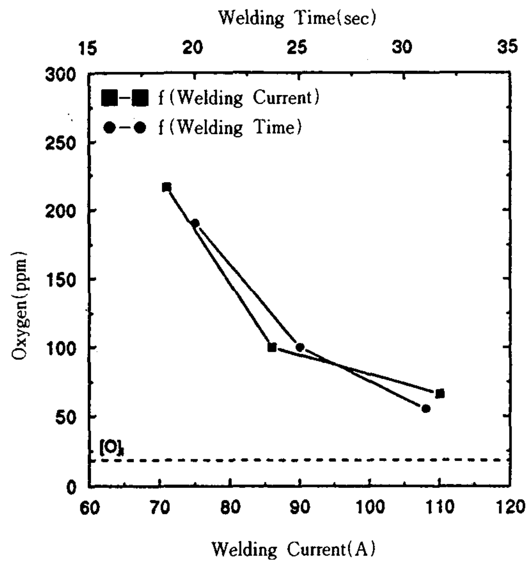

Fig. 6 Variation in the Concentration of Oxygen in the Weld for Electrode Negative Polarity Stationary Gas Tungsten Arc Welding as a Function of Welding Current and Welding Time

polarity stationary gas tungsten arc welding give good evidence for the effect of electrochemical reactions on the transfer of oxygen at the plasma/metal interface. Figure 6 shows the variation of oxygen content as a function of welding current and welding time respectively. As the welding current or welding time increases, weld metal oxygen content decreases, due to an increase in refining reaction of oxygen at the anode.

4. CONCLUSIONS

1) The interface between plasma and molten metal acts as an important site for electrochemical and thermochemical reactions.
2) The transfer of oxygen and nitrogen are controlled by both thermochemical and electrochemical reactions. The thermochemical reaction is the pickup of oxygen and nitrogen from the surrounding atmosphere. The cathodic electrochemical reaction during the reverse polarity gas tungsten arc welding is the refining of oxygen and nitrogen.
3) Both welding current and travel speed are important process parameters controlling the electrochemical transfer of oxygen and nitrogen. The extent of electrochemical reactions increase as the welding current increases and as the travel speed decreases because electrochemical reactions are enhanced by higher total current flow per unit volume of weld metal.
4) The proper selection of welding process parameters can minimize the adverse effects of electrochemical reactions on the chemistry of the weld metal.

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