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Gas－Tungsten－Electrode (Report 5)†

－ Effect of Combined Additives of Rare Earth Metal Oxides on Arc Characteristics of Tungsten Electrode －

Fukuhisa MATSUDA*, Masao USHIO** and Alber A. SADEK***

Abstract

A comparative study has been made on the arc characteristics of gas-tungsten-arc (GTA) cathodes, consisting of tungsten mixed with a small quantity of two or three combined additives of Y-, Ce- and La-oxides respectively. Arc starting characteristics, arc pressure, electrode consumption, change in electrode tip shape due to long time operation and stability of rare earth metal oxides during arcing are compared. The present results have indicated that the arc characteristics and electrode durability depends on the stability and the behavior of rare earth metal oxides during arcing.

Electrodes activated with three combined additives of rare earth metal oxides showed good arc characteristics and high endurance to long time operation related to the changes in the grain boundary morphology which retardes the migration of La-oxide and Ce-oxide.

KEY WORDS: (Tungsten) (Oxide Tungsten) (GTA Welding) (Nonconsumable Electrode) (Refractory Metals) (Electrode Temperature)

1. Introduction

A comparison study had been made on the arc characteristics of tungsten electrodes activated with single additives of rare earth metal oxides[1]. From that study, the W-La_2O_3 electrode showed a superior arc characteristics among the investigated electrodes followed by W-Y_2O_3, W-CeO_2 and W-ThO_2 electrodes in that order.

Also, the observation of rare earth metal oxides behavior during arcing[2] and for long time operation pointed out the following:

(1) Generally, the rare earth metal oxides react with tungsten forming tungstate and/or oxy-tungstate. These tungstate or oxy-tungstate melt and migrate from the lower temperature zone to the higher temperature zone. The differences in their migration rates was attributed to their melting points and the grain boundary morphology.

(2) Ce-oxide has the most higher migration rate among those oxides which has lower melting point (even for the oxide it self or for the tungstate). Then, it is easy to migrate and make a continuous feeding of oxides to the electrode tip. But, in that case the consumption rate of oxide is much higher.

(3) According to the higher melting point of Y-oxide and the shape of tungsten grain boundary in that case, which exhibits granular shape, the migration rate along the grain boundary takes more time to rich the electrode tip or surface and evaporate comparing with the other oxides. However, the rim was formed at the tip during long time operation and then the arc characteristics became worse.

(4) The tungsten electrodes activated with La-oxide showed acceptable compromization between the stability behavior and arc characteristics among the investigated electrodes. The stability of La-oxide was related to the higher melting point of the oxide as well as for its oxy-tungstate. Also, there is a reasonable equilibrium between the migration and vaporization rates.

It is apparent that, every oxide has its characteristics and behavior and all of these electrodes fall to achieve good durability or endurance for long time operation. Then, this study was carried out to study the effect of combined additives of rare earth metal oxides on the arc characteristics of tungsten electrodes as well as the durability during long time operation.

2. Experimental Procedure

The investigated electrodes in this work were produced by the conventional powder metallurgy process with 2.4 mm diameter, centerless ground rods, with the chemical composition shown in Table 1. The power system for arc characteristics measurement was a conventional constant current type, with a negative direct current in the electrode and a copper water cooled anode. A constant arc length of 3 mm and torch angle of 90 degree to the anode were used throughout the series of the tests. Arcing with constant arc current (180 amp) at time

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Table 1 Produced electrodes and their oxides content.

	La_2O_3	CeO_2	Y_2O_3
Type A	1.0	1.0	---
Type B	1.5	0.5	---
Type C	0.5	1.5	---
Type D	1.8	0.2	---
Type E	1.5	1.0	---
Type F	1.0	1.5	---
Type G	0.5	2.0	---
Type H	2.0	0.5	---
Type K	---	1.0	1.0
Type L	---	0.5	1.5
Type M	---	1.5	0.5
Type N	1.0	---	1.0
Type O	1.5	---	0.5
Type P	1.8	---	0.2
Type Q	0.5	---	1.5
Type R	0.4	0.4	1.2
Type S	0.4	1.2	0.4
Type T	0.6	0.8	0.6

intervals until 10 hr in pure argon had been done to observe the metallurgical changes, the behavior of oxides and the electrode durability.

Specimens were prepared for metallographic examination by means of hard grinding through No. 1500 emery papers and polishing with fine alumina. Polished specimens were electrolytically etched in a solution consisting of Na-OH-10% normal for macrostructure observations. During the electrolytic etching, the current voltage aids was set at 30V for 50 sec. It for delineating the distribution of oxides after arcing. To observe the microstructure and the recrystalized grains after arcing Murakami's reagent was used. Etching reagent was applied by swabbing for 5–10 sec.

3. Results and Discussion

It will not be appropriate to report, in this short discussion the details of all experimental results. We shall restrict ourselves to a brief summary of the most important facts and prameters.

3.1 Arc characteristics

3.1.1 Arc starting characteristics

The arc starting characteristics were evaluated by the repetitive arc start test in pure argon. **Figure 1** shows the distributions of the time between onset of HF power and following-up of 20 A arc. In this case the HF power was applied about 2 sec and the iteration interval was adjusted at 20 sec. The best electrode was considered that one which shows the largest number of arc starts at shorter time.

From Fig. 1 it is obvious that both of Type R and Type T have superior arc starting characteristics, followed by Type B, Type P, Type A and Type D in that order.

3.1.2 Arc voltage-current characteristics

The voltage-current characteristics for GTA with pure tungsten, W-La_2O_3 and tungsten electrodes activated with combined oxide additives have been investigated and shown in **Figure 2.**

The differences among arc voltage-current characteristics of various tungsten oxide electrodes are essentially negligible, and were represented in Fig. 2 as a band, comparing with pure tungsten electrodes which shows a comparatively large deformation due to melting. The observed total arc voltage-current changes are believed to be due to the changes in the cathode potential drop which depends on the tip geometry changes when all other factores are fixed (such as current, anode condition, etc.).

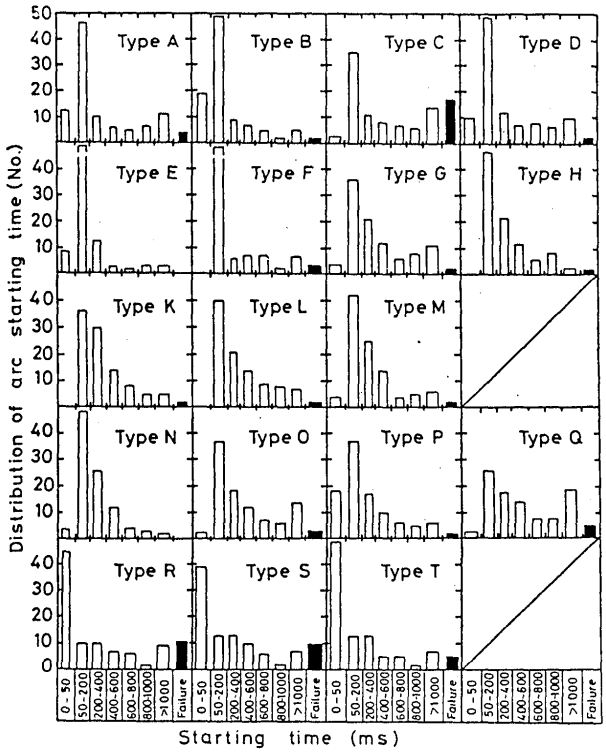

Fig. 1 Distribution of arc starting time. Arc starting time means the time between onset of HF power and following of arc.

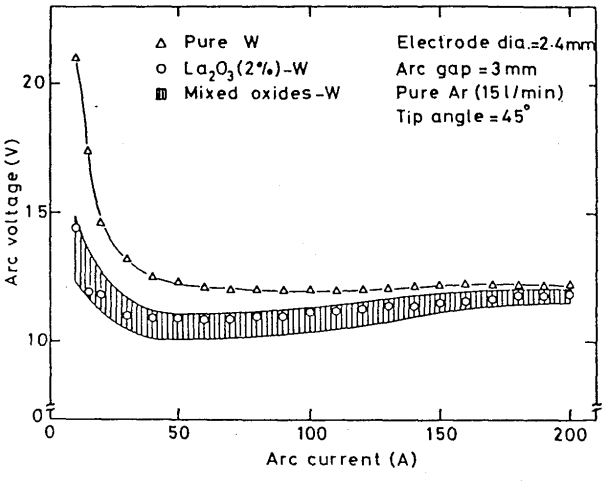

Fig. 2 Arc voltage-current relationship.

3.1.3 Arc pressure

The arc pressure distribution at the water cooled copper anode for several electrode materials as measured by a semiconductor transducer is shown in **Figure 3.** The arc pressure as a function of arc current is illustrated in **Figure 4.** The arc pressure is originated mainly by the induced flow of plasma gas due to the expantion of the current path near the cathode. Therefore, any deformation due to melting must change the current distribution near cathode and the impedance for induced flow of plasma gas, consequently leading to a change in the arc pressure. Accordingly, the tungsten electrodes activated with combined oxide additives, which have little deformation at the tip, showed high arc pressure.

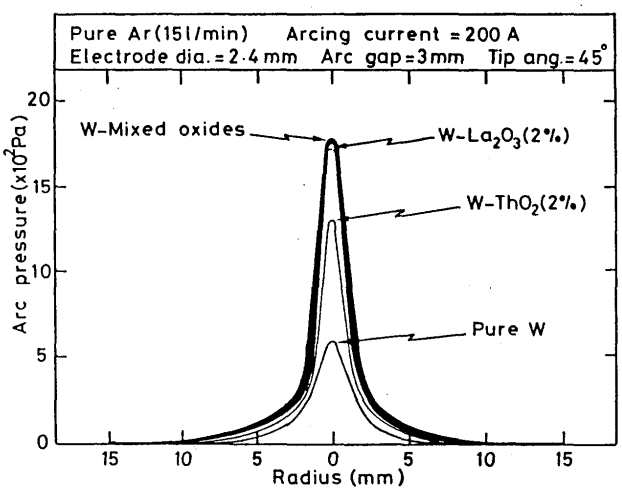

Fig. 3 Arc pressure distribution.

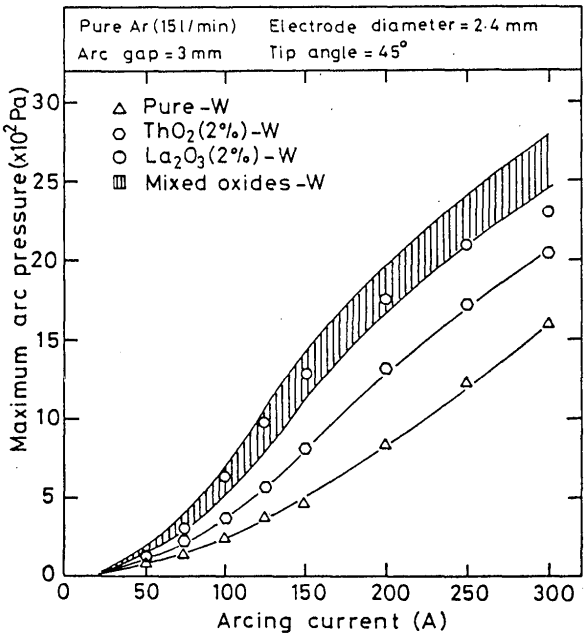

Fig. 4 Effect of current on maximum arc pressure.

3.2 Electrode durability

Electrode durability was evaluated during long time operation until 10 hr in pure argon at 180 A.

Two criteria for durability evaluation were adopted in this work, these are:

(1) The stability of tip shape, for partial and complete melting or any geometry changes.

(2) The formation of rim.

The collected data for long time operation and the appearance of the electrode tip for all investigated electrodes are shown in **Figures 5, 6, 7, 8, 9** and for W-La_2O_3 electrode as a refrence in **Figure 10.** Based on such data, the following statements become obvious.

(1) The electrode activated with the addition of La-oxide and Ce-oxide, in total amount of 2% or 2.5% showed good durability until 4 hrs as represented by Type A, Type H, Type D and Type G, in that order. The other electrodes were stoped until 3 hrs arcing, and all of them were melted in case of 10 hrs arcing. It may be benefit to point out that the addition of both La- and Ce-oxides goes under critical chemical composition controlling the beavior and stability of these oxides during arcing, and hence the durability of GTA electrodes. Then among

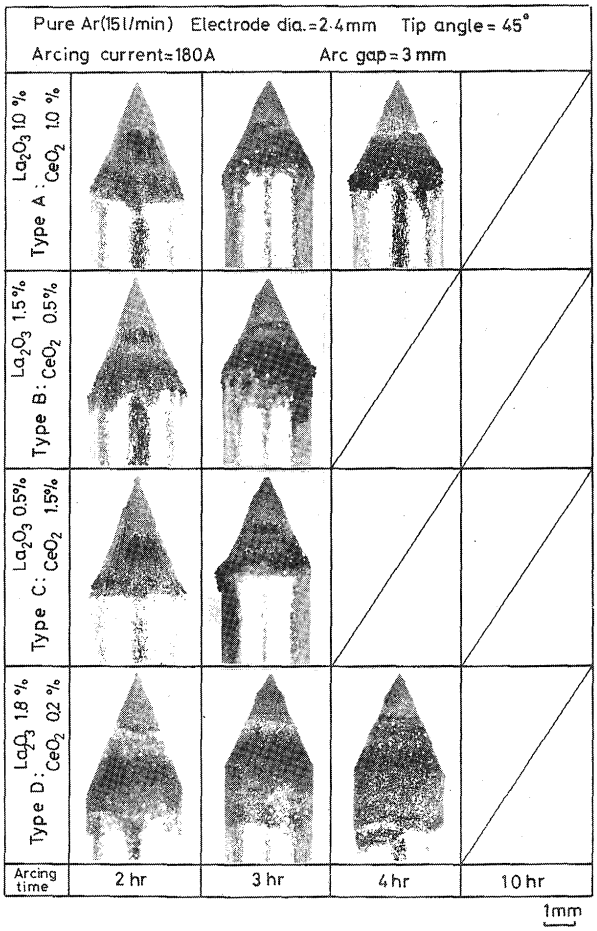

Fig. 5 Change in electrode tip shape due to long time operation (La_2O_3 + CeO_2 = 2%)

these investigated electrodes the best mixing ratio between La-, Ce-oxides is 1:1 as shown in Figs. 5&6.

(2) The electrodes activated with the additon of La-oxide and Y-oxide in total amount of 2% also showed good durability until 4 hrs and acceptable durability until 10 hrs. as represented by Type Q, which have the mixing ratio of 1:3 for La-oxide to Y-oxide respectively, as shown in Fig. 7.

(3) The electrodes activated with the addition of Ce-oxide and Y-oxide in total amount of 2% showed acceptable durability until 4 hrs as represented by Type L and Type M as shown in Fig. 8. All of these kind of electrodes suffer from rim formation during arcing at 10 hrs. or at 3 hrs. like in case of Type K. Here also, the best durability was achieved by the mixing ratio of 1:3 for Ce-oxide and Y-oxide respectively.

It is inferred from this observation that the durability of GTA electrodes activated with combined additives of rare earth metal oxides controlled by the mixing ratio of those oxides.

(4) With this in mind, a detailed investigation and analysis of the next group of electrodes have been made and a summary of the results are contained in Fig. 9.

The electrodes activated with the addition of La-oxide,

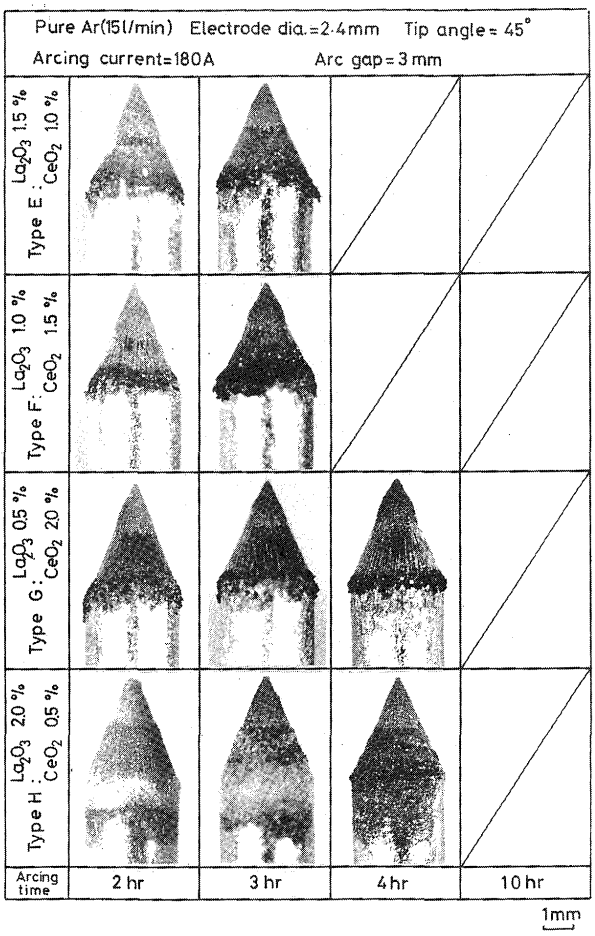

Fig. 6 Change in electrode tip shape due to long time operation ($La_2O_3 + CeO_2 = 2.5\%$)

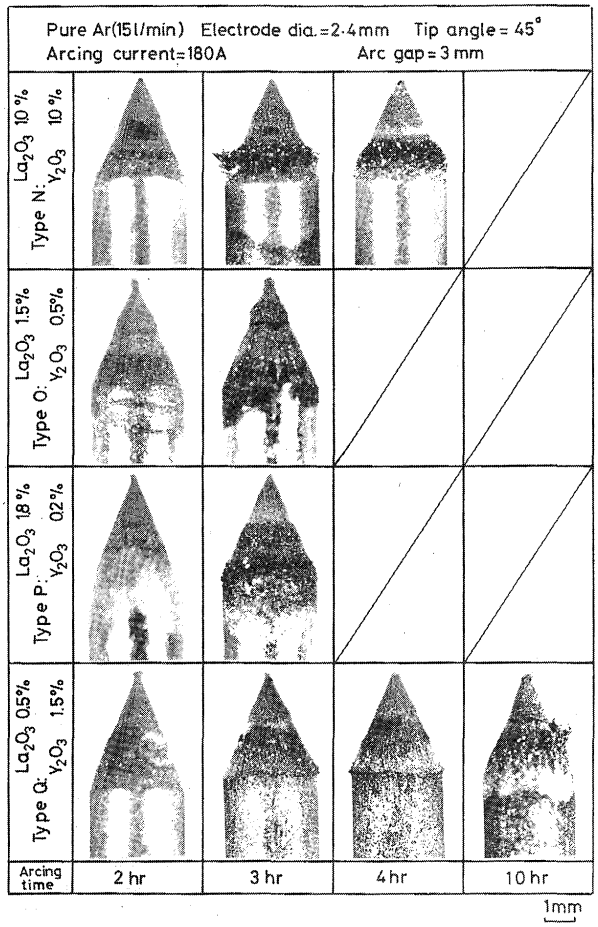

Fig. 7 Change in electrode tip shape due to long time operation ($La_2O_3 + Y_2O_3 = 2\%$)

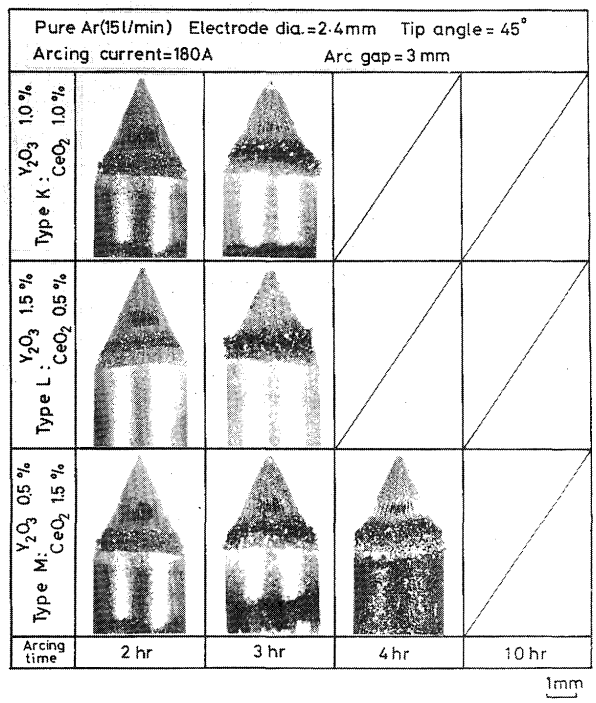

Fig. 8 Change in electrode tip shape due to long time operation ($Y_2O_3 + CeO_2 = 2\%$)

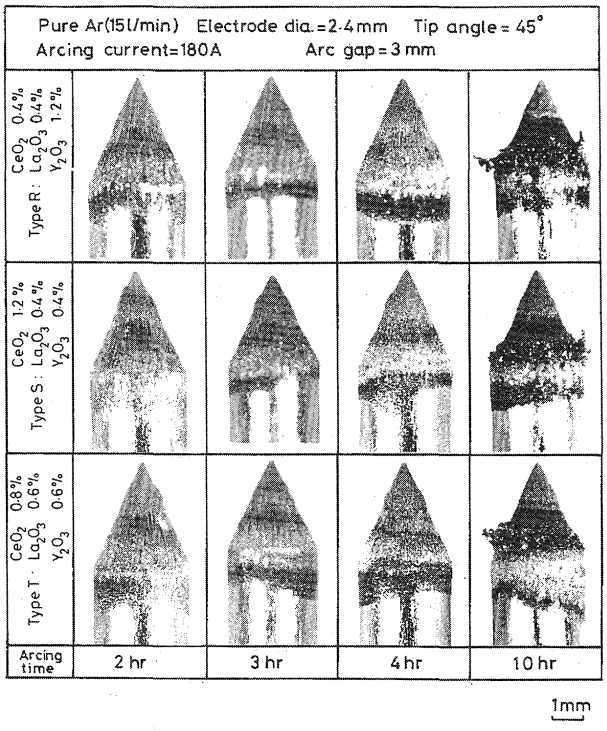

Fig. 9 Change in electrode tip shape due to long time operation ($La_2O_3 + CeO_2 + Y_2O_3 = 2\%$)

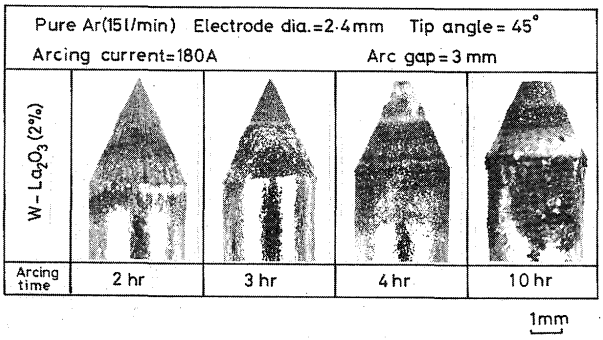

Fig. 10 Change in electrode tip shape due to long time operation of W-La_2O_3 (2%) electrode.

Ce-oxide and Y-oxide in total amount of 2%, showed good and superior durability until 10 hrs. comparing with either the previous electrodes or W-La_2O_3 electrodes, which suffer from sever melting during arcing until 10 hrs (Fig. 10). All of these electrodes types have a very small rim at the tip comparing with the other electrodes. However the tip was kept sharp.

3.3 Metallographic examination

In order to clarify the differences between all of these mixing ratios of oxides and their effects on arc characteristics, it is necessary to make a study on the comparable metallurgical characteristics and the behavior of these oxides during arcing. Here the microstructures of the electrodes which showed good durability as mentioned before in the previous section will be observed. Also, the microstructures in case of tungsten electrodes activated with two combined addititives of rare earth metal oxides after 30 and 60 mins will be discussed to observe clearly the behavior of rare earth metal oxides, because after long time operation the amount of residual oxides became very small and it will not be clear to observe the oxide behavior in that case. But in case of tungsten electrodes activated with three combined additives of rare earth metal oxides the microstructures will be discussed. Finally, all the results of these metallurgical studies will be concluded.

3.3.1 Tungsten electrodes activated with two combined additives of rare earth metal oxides

Figures 11, 12 and **13** shows the typical microstructures throughout the electrode tip of Type A, Type L and Type Q electrodes after arcing in pure argon for 30 and 60 mins at 180 A. From these results and the results of oxide behavior study[2], which were summarized before in the introduction section, the following statements become obvious:

(1) In case of mixed La-Ce-oxide additives (type A) the behavior of both oxides were represented. This can be understood if the two denoted zones at 2&4 mm in Fig. 11 investigated precisely. The SEM and EDX spot analysis for the oxide particles which shown in **Figure 14** indicated that:

(a) Generally the La-oxide and Ce-oxide reacts with tungsten forming tungstate and oxy-tungstate (as in case of separate addition). But in this case the new combined types of tungstate or oxy-tungstate may have a lower melting point than La-tungstate and higher melting point than Ce-tungstate (depending on the chemical composition). Then these new compounds can achieve a considerable balance between vaporization and migration rates as well be shown in the following statements.

(b) At 2 mm (B zone) the concentration of La-oxide differe from the particles near the surface to the particles just below the surface. In other words at this zone the La-oxide evaporate from the surface, as in case of W-La_2O_3 electrodes which showed complet denoted zone at this area, (particle (a)) but as shown here the denoted zone is not so biger due to the supplemental effect of new oxide particles (particles (b) and the C zone).

(c) At 4 mm (D zone) the concentration of La-oxide became much higher than the concentration of Ce-oxide, that the same behavior of Ce-oxide which had been obvious remarks it can be concluded that the mixing process is not so benefit due to the double loss of both oxides.

(2) In both cases of mixed Ce-Y-oxides (Type L) and La-Y-oxides (Type Q), The phenomena which was observed in case of W-Y_2O_3 electrode occurred[2], That the tungsten grains recrystallized and showed granular grains. This grain boundary morphology retarted the migration

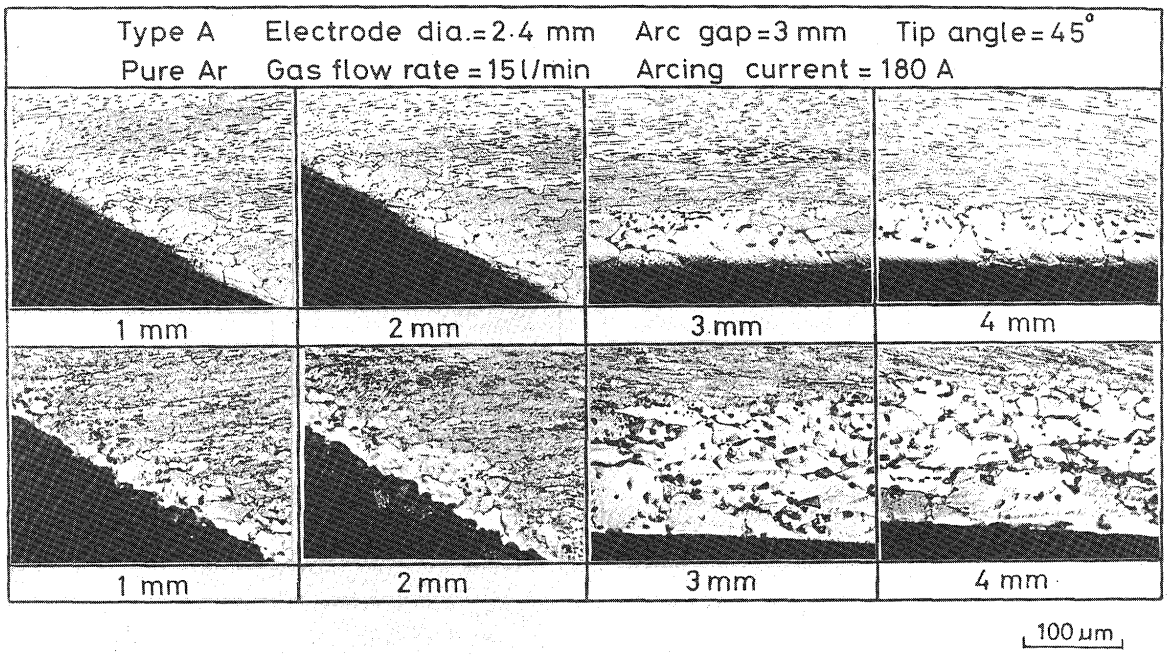

Fig. 11 Typical microstructure of Type A electrode at different distances from the electrode tip. Upper part after 30 min, lower part after 60 min.

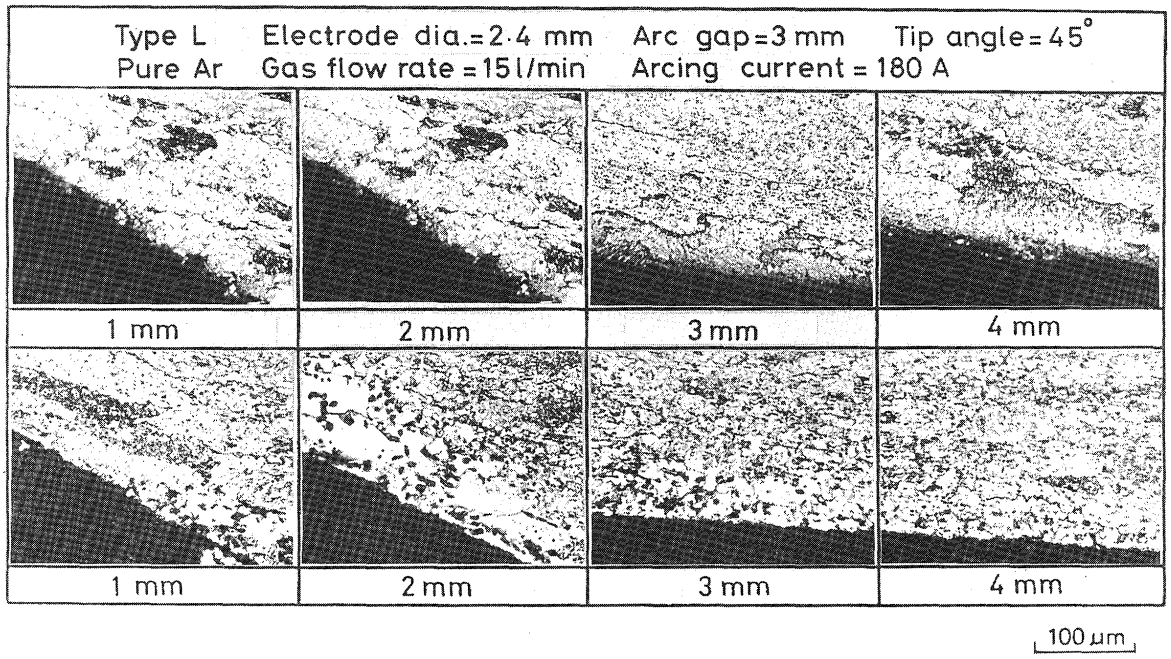

Fig. 12 Typical microstructure of Type L electrode at different distances from the electrode tip. Upper part after 30 min, lower part after 60 min.

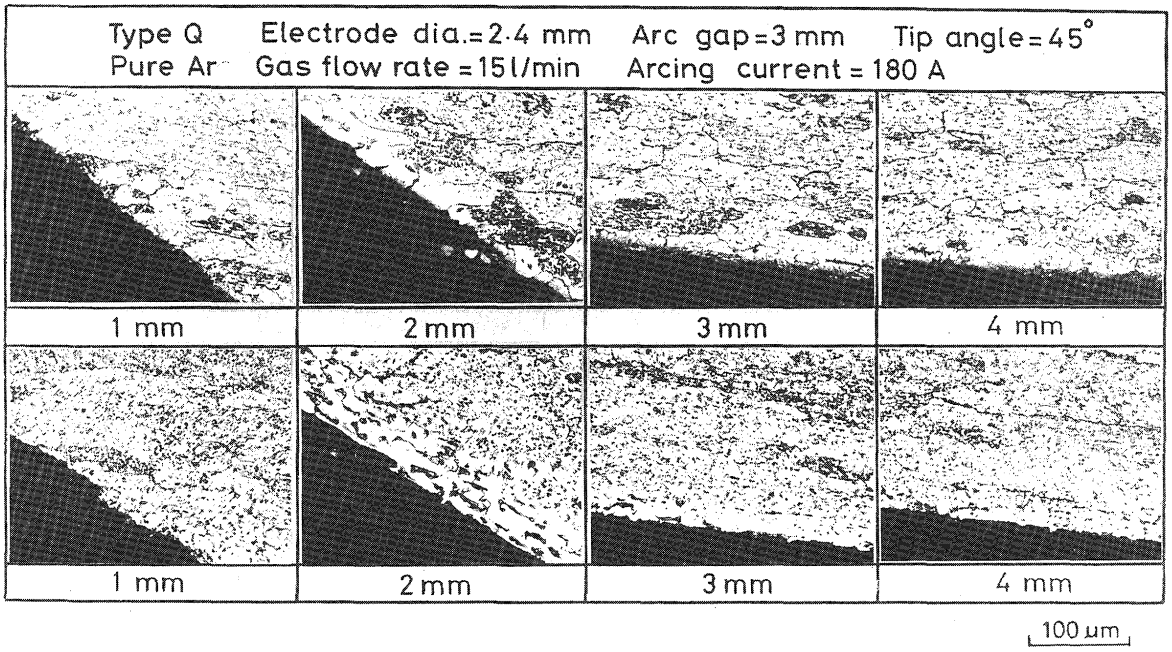

Fig. 13 Typical microstructure of Type Q electrode at different distances from the electrode tip. Upper part after 30 min, lower part after 60 min.

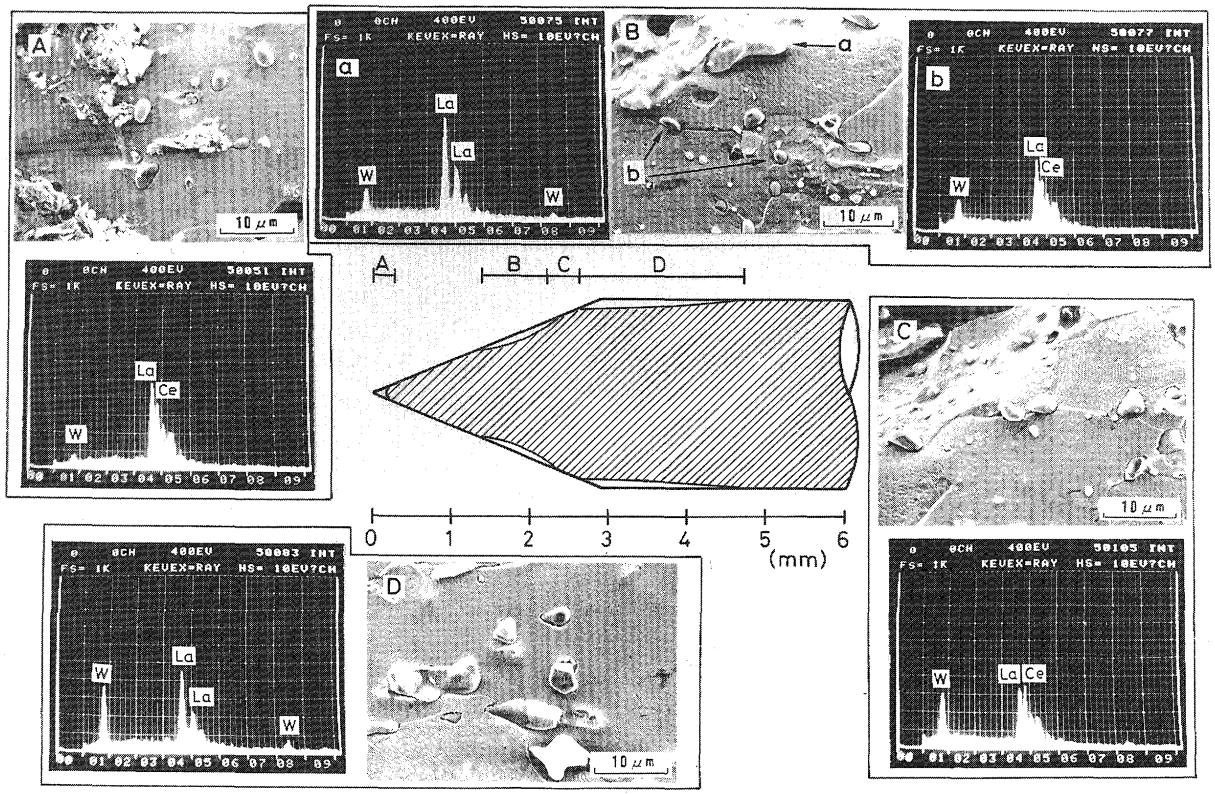

Fig. 14 The distribution of La_2O_3 and CeO_2 throughout the electrode tip for Type A electrode after 60 min arcing at 180 A in pure Ar, by using SEM and EDX spot analysis.

process of both La-oxide and Ce-oxide as well as the vaporization of these oxides from the electrode surface, as shown with the comparison between the microstructures of these electrodes. Also, due to the high stability of Y-oxide, which related to its higher melting point and lower migration rate, it can replace the vaporized oxides of La-Ce-oxides.

These two main results gives much more evidence on the different behaviors of the rare earth metal oxides during arcing and the critical mixing ratio which was discussed previously. Also, these results reflects the importance and the predicted benefits which can be gained when these three types of rare earth metal oxides were mixed.

3.3.2 Tungsten electrodes activated with three combined additives of rare earth metal oxides

According to the highest stability of these types of electrodes until 10 hrs arcing, the macrostructures of these electrodes comparing with that of W-La_2O_3 electrode shown in **Figure 15** will be discussed.

From Fig. 15 it is obvious that Type R has high concentration of oxide remained after arcing (gray area) followed by Type S and Type T in that order. While the W-La_2O_3 electrode showed high consumption rate of La-oxide. That results achieved the previous prediction about the critical mixing ratio between the rare earth metal oxides as $La_2O_3 : CeO_2 : Y_2O_3 = 1:1:3$ respectively.

The most favorable mechanism for the stabilization of rare earth metal oxides inside the electrode was thought as follows; the presence of Y-oxide encourage the recrystalization of tungsten grains and exhibits granular shape instead of its longitudinal shape. Then the migration of both La-oxide and Ce-oxide will be retarted, leading to high stability. Moreover, that mixing ratio (1:1:3) achieved good compromization between the arc characteristics, high durability, stability of oxides and acceptable resistance to the rim formation.

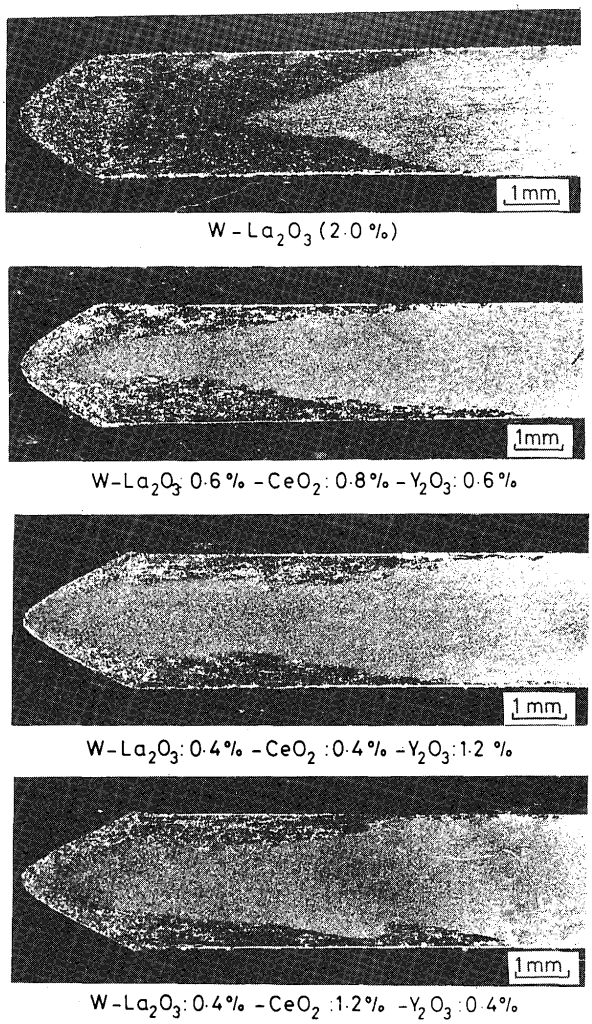

Fig. 15 Typical macrostructure of tungsten electrode activated with combined additives of rare earth metal oxides after 10 hr arcing at 180 A in pure Ar.

4. Conclusions

This study has been carried out to investigate the effects of two and three combined additives of small amounts of rare earth metal oxides on the arc characteristics and the electrode durability for long time arcing of GTA electrodes. Results were compared with those of electrodes activated with single addition of rare earth metal oxides.

The main conclusions drawn from the experimental data are:

(1) The electrodes activated with three combined additions of rare earth metal oxides specially Type R reflected higher endurance to long time operation than the other electrodes. This higher endurance was correlated with the changes in the grain boundary morphology due to the presence of Y-oxide and the migration process retardation of La-oxide and Ce-oxide.

(2) All of the investigated electrodes represented superiority on arc characteristics, than that of the electrodes activated with single addition of rare earth metal oxides.

(3) The mixing ratio of $La_2O_3 : CeO_2 : Y_2O_3$ as 1:1:3 respectively achieved the best compromization between the arc characteristics, high endurance to long time operation, good stability of oxides and acceptable resistance to the rim formation.

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