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THE ELECTRON NUMBER DENSITY OF ARGON-OXYGEN ATMOSPHERIC PRESSURE PLASMA

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ABSTRACT

The studied plasma source was a wall stabilized direct current arc burning in argon at atmospheric pressure, originally designed for elemental spectrochemical analysis. The influence of oxygen addition (1 - 20 %) on the electron density of mixed gas plasma was investigated for two initial plasma compositions, i.e. argon gas with water aerosol and argon with water aerosol containing 0.5 % KCl. The electron number densities were calculated from the measured *Stark* profiles of H β spectral line. The results have shown that the addition of oxygen has weak influence on electron number density in the studied central plasma zones. Similar trends were observed regardless of the presence or absence of easily ionized element (potassium).

INTRODUCTION

A direct current (DC) argon arc is an atmospheric-pressure plasma source that has several important operational characteristics in common with inductively coupled plasma (ICP): argon as the main gas, similar values of temperature and electron number density, and analyte introduction in the form of aerosol. The detection power of this device is comparable with ICP devices, especially for analysis of heavy metals in aqueous solutions. The addition of easily ionisable elements or molecular gases to argon plasma is a common spectrochemical practice, applied to improve excitation conditions of the source, or to minimize matrix effects [1]. Our previous studies of argon-nitrogen plasma have shown that the addition of molecular gas may improve the analytical performance of DC arc source [2,3]. The aim of the present research is to test the applicability of another molecular gas, oxygen, for the same purpose. This paper describes the effects of various proportions of added oxygen on electron number density (n_e) in argon DC arc burning in atmospheric pressure. As an air constituent, oxygen diffuses into the open-

air plasma, thus, our results may be of interest for other atmospheric plasma sources as well.

EXPERIMENTAL

The excitation source under study is an arc specifically designed for spectrochemical analysis. A detailed description of U-shaped direct current (DC) argon stabilized arc was given in our previous papers [2,3]. The oxygen was added to two initial plasma gas compositions, argon gas with water aerosol, and argon gas with water aerosol containing 0.5% KCl. The addition of oxygen gas was carried out by increasing the flow rate of the O₂ and decreasing that of Ar, so that the total flow rate of the mixed gas was kept constant at 2.7 dm³min⁻¹, which is the optimal flow for plasma operating in pure argon. An appropriate amount of oxygen, in the range from 1 to 20 %, was gradually added. The effect of the addition of oxygen on electron number density at the arc axis was investigated for three arc currents 5, 7 and 9 A. Also, radially resolved electron density profiles were obtained for the arc current of 7A, a current that is commonly used in spectrochemical applications of this source.

Our prime goal was to measure changes of the electron number density (n_e) caused by the addition of oxygen to the argon plasma. Among the various methods available for this purpose, measurement of the half widths of the *Stark* broadened *Balmer*- H β spectral line (486.13 nm) was chosen for two reasons. Firstly, the method is independent on local thermodynamic equilibrium assumption (LTE). Secondly, hydrogen exhibits a linear *Stark* effect that is readily measured even at densities lower than 10²¹/m³. Another accurate method for determination of n_e (that is also independent on LTE assumption), the absolute measurement of the continuum emission, was a rather complicated task for mixed gas plasma. For evaluation of n_e a program proposed by *Zikic et al* [4] was used. The program enables evaluation of electron number densities by comparing the experimentally obtained emission profiles of H β with the theoretically calculated ones.

RESULTS AND DISCUSSION

For our working conditions (atmospheric pressure, not too high values of electron number densities and temperature), the most important cause of hydrogen spectral lines broadening was *Stark* broadening (full width at half maximum, FWHM between 5·10⁻² and 3·10⁻¹ nm), while instrumental and *Doppler* widths were much smaller (FWHM < 10⁻² nm). The accuracy of the electron density determination was estimated to be 5–7%. Addition of oxygen to the argon plasma with an aqueous aerosol (with or without KCl)

caused negligible changes in the electron number density at the arc axis, for all investigated arc currents (Figure 1).

Moving away from the arc axis, small changes in radial distribution of electron number density could be observed (Figure 2). The largest change (increase) of n_e was observed in the zone 0.5 mm to 1 mm from the arc axis. At greater distances from the arc axis, n_e is somewhat higher than in the plasma without oxygen, but does not depend on the concentration of added oxygen. In the presence of potassium, density of electrons in the plasma core was smaller

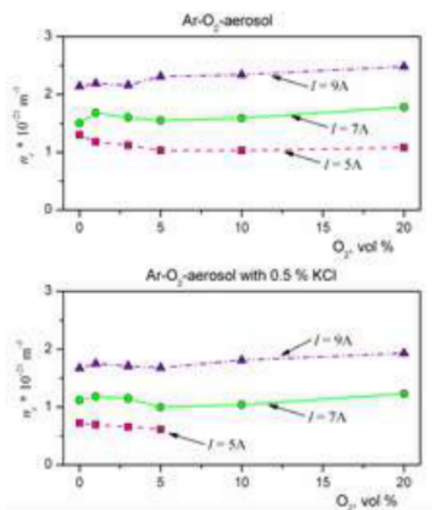


Figure 1. The influence of oxygen gas addition on electron number densities at the arc axis, for different arc currents.

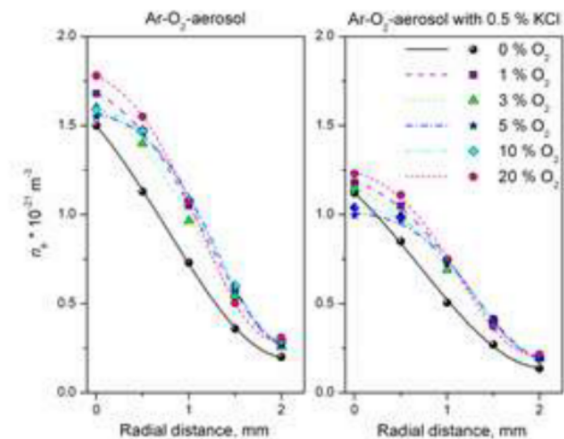


Figure 2. The influence of oxygen gas addition on radial distribution of electron number density in arc plasma operating at 7 A.

compared to the plasma without KCl, however, the added oxygen had a same effect on n_e . DC argon arc plasma is characterized by a large electron number density and temperature gradients in radial direction. The addition of oxygen may influence the energy transport through the discharge causing the changes of the fundamental plasma properties (n_e , electron and gas temperature), the bulk plasma characteristics (thermodynamic functions and transport coefficients, diffusion coefficient, viscosity, thermal and electrical conductivity) and radiation properties of the plasma. In the present case, these changes are found to have minor influence on plasma parameters in the arc core.

Dissociation of oxygen contributes to a local increase in thermal conductivity of plasma through the heat transferred as chemical enthalpy of molecules. However, the largest effect is expected in plasma zones where the temperature corresponds to the dissociation temperature of molecular oxygen, which is around 3500 K, and for the studied argon arc, this zone is at the plasma periphery. Consequently, the presence of oxygen will reduce the temperature gradient in the peripheral plasma zones which will indirectly induce small changes of the plasma parameters in the arc core. At the same time, due to high temperature, and relatively low dissociation energy of O₂ (5.2 eV), a dissociation degree of oxygen in the arc core is probably high. If that is the case, mixtures of oxygen and argon would behave as a mixture of two atomic gases, which could explain the small effect of oxygen on the electron number density of the arc core.

CONCLUSION

Influence of different oxygen contents (1 % to 20 %) on the electron number density in atmospheric pressure argon DC arc has been studied. The results have shown that the changes in oxygen content slightly influence the changes in electron number density through changes in plasma temperature and thermal conductivity. The presence of easily ionized element lowers electron number densities in the central plasma zones of argon plasma, but the effect of oxygen on n_e is the same as for mixed gas plasma without potassium.

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