Kinetics of an argon inductively coupled plasma

KINETICS OF AN ARGON INDUCTIVELY COUPLED PLASMA

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ABSTRACT

We studied the kinetics of an argon inductively coupled plasma (ICP) by measuring the temperatures of electrons and heavy particles and the drift velocity. The experimental technique was based on analysis of small disturbances, induced by temporal changes in the power supply or the central gas flow. By applying a simple model for the energy balance we found that the most important terms for the energy loss of the plasma are the axial convective energy transport and the radial heat conduction.

INTRODUCTION

In the last decade much attention is paid to the kinetics and the elementary balances in an argon ICP, especially to improve the quality of the plasma as a spectrochemical source. At the level of elementary balances it has been found that the ICP is in a state which is close to LTE and that it is possible to characterize the plasma by giving the electron density and temperature, n_e and T_e /1/.

However, this method for characterizing the plasma is not sufficient to study the transport phenomena associated with the energy balance. We only can understand this balance if we have additional information to estimate the role of heat conduction towards the surroundings, radiation losses and the energy flow caused by convective transport of heated heavy particles.

Although many parameters necessary for this estimation have been measured before, only one parameter still is rather uncertain, the drift velocity of the plasma. In this paper we present two methods of measuring the velocity and the temperatures of the plasma by means of small temporal disturbances.

THEORETICAL

The energy balance used is a simplified model without difference in drift velocity between the electrons and the heavy particles. Also the excitation energy of excited states and ions is neglected. In this case the energy balance reads:

$$\frac{5}{2}\vec{\nabla}\cdot(\vec{w}p) - \vec{\nabla}\cdot(\lambda_e\vec{\nabla}T_e + \lambda_h\vec{\nabla}T_h) = \sigma E^2 - Q_R, \qquad (1)$$

in which $\vec{w}$ is the drift velocity of the plasma, p the pressure, λ_e and λ_h the thermal conductivities of respectively the electrons and the heavy particles, σE^2 the energy input per unit of volume and Q_R the energy loss per unit of volume caused by escape of radiation. After integrating eq. 1 over the volume V in which the energy is coupled into the plasma we find:

$$\int_A (\tfrac{5}{2}p\vec{w} - \lambda_e\vec{\nabla}T_e - \lambda_h\vec{\nabla}T_h)\cdot\vec{n}\,dA + \int_V Q_{CR}\,dV = P_{EM}, \qquad (2)$$

in which A is the surface of the integration volume, $\vec{n}$ the normal vector to the surface and P_{EM} the power input into the plasma. The integration volume we take is a cylinder with a radius of 6 mm starting just below the coil and ending at 5 mm above the load coil. This is shown in fig. 1.

T_e and T_h are obtained from the intensity jumps at the switching of the generator. The analysis of the power interruption measurements is based on the assumption that the Saha balance is totally dominant for highly excited states of argon and hydrogen. The intensity jump due to the cooling of the electrons to the heavy particle temperature is given by /2,3,4/:

$$\ln\left[\frac{I^*}{I}\right] = \frac{3}{2}\ln(\gamma) + \frac{\gamma - 1}{kT_e}E_p, \qquad (3)$$

in which I and I* are the intensities before and directly after the switch off of the generator, γ is the ratio of T_e and T_h and E_p is the ionization energy of the excited state p.

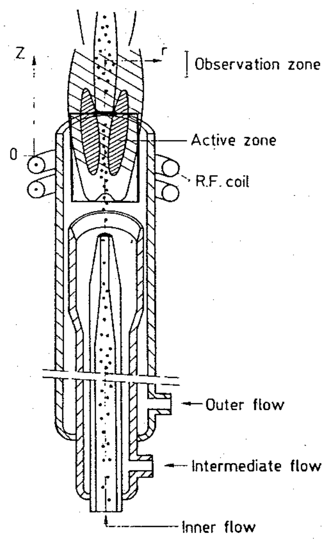

Fig. 1. Plasma and integration volume

EXPERIMENTAL

For the experiments a 100 MHz Philips RF–generator with a possibility of interrupting the power (typically 20–100 μs), was used. The RF–coil consists of two windings with a diameter of 35 mm and a height of 15 mm. The plasma torch consists of three concentric quartz tubes and is a standard torch developed by Philips /5/. The standard argon flow conditions are 12 l/min outer flow, 0 l/min intermediate flow and 0.7 l/min inner flow. Further a piezo electrical controlled valve with an opening and closing time of 2 ms for injecting small amounts of gas into the central flow has been installed. The power input is 0.8 kW. The plasma radiation is focused on the entrance slit of a B&M BM100 monochromator with a grating of 1200 g/mm. The wavelength selected light is led to an EMI 9698 QB photomultiplier, operating at a voltage of 1400 V. After being amplified by a Phillips variable gain amplifier model 777, the anode signal is discriminated at height by a Phillips discriminator model 715. The pulses from the discriminator can be counted as a function of the time by an EG&G Ortec ACE–MCS Multi Channel Scaler. By using this MCS we can observe about 8 ms continuously with an accuracy of 2 μs. The time dependent measurements are controlled by an AT computer.

RESULTS AND DISCUSSION

In fig. 2 the relative jumps of various line intensities of argon are plotted as a function of the ionization energy, E_p. These measurements have been carried out at a height of 5 mm above the load coil (ALC), 6 mm from the center. The plasma was operated under standard conditions without water injection. By assuming Saha equilibrium for all states and applying eq. 3 we find $\gamma = 1.31$, $T_e = 0.72$ eV and $T_h = 0.55$ eV. The method is in agreement with those obtained by other techniques /6,7/.
For measuring the drift velocity of the plasma we used two methods. The first method is based on measuring the velocity of disturbances /3/ of the plasma, created in the expansion zone of the plasma during the interruption of the generator, while the second method is based on measuring the drift velocity of small gas pulses of H_2 or Ne, which are put into the

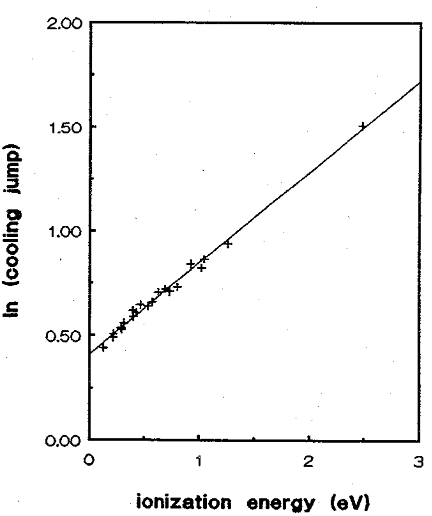

Fig. 2. Relative cooling jump of argon levels as a function of the ionization energy at 5 mm ALC and r = 6 mm. The least squares fit gives $T_e = 0.72$ eV and $T_h = 0.55$ eV.

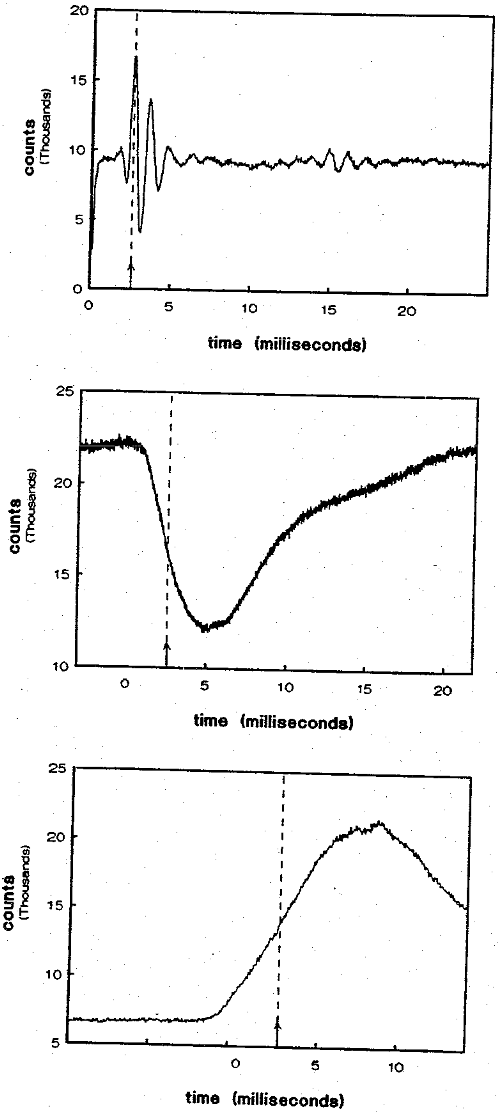

Fig. 3. a. Typical delayed responses of Ar(6d) caused by power interruption, b. Response of Ar(6d) after injection of a Ne gas pulse, c. Response of Hα after injection of a H_2 gas pulse. The arrows indicate the time of arrival

central flow. Fig. 3a shows the delayed response of an argon line intensity to the interruption of the generator power. Changes in intensity occur in typically 0.5 ms. The addition of a small gas pulse gives much slower reactions due to (a) the delay caused by transport of the gas pulse through the central channel and (b) the diffusion of the gas in the plasma. This is shown in fig. 3b and 3c for Ne and H_2 respectively. Because the Ne line intensities are too small we measured the reaction of the Ar(6d) line (549.59 nm) to the disturbance. The hydrogen was detected by observing the H_α line.
The velocity can now be found by measuring the time of arrival of the disturbances. The results are shown in fig. 4. We defined the time of arrival for the power interruption method as the time at which the highest maximum arrived and for the gas pulse methods the time at which the disturbance was at half of its maximal height. The 'delayed power interruption' method gives the best temporal resolution and provides a velocity of the plasma of 12.1 ± 0.1 m/s. The 'gas pulse' method is less accurate and we also suspect that the plasma is disturbed quite substantially. The velocities obtained with H_2 and Ne as pulse gas are respectively 10 ± 1 m/s and 16 ± 1 m/s.

The next part is the estimation of the terms of eq. 2. by inserting the measured parameters and using the following assumptions:

1. the axial drift velocity at the entrance of the integration volume equals the mean axial velocity of the cold gas at the beginning of the torch, which is in our case 0.85 m/s,
2. the radial drift velocity is that small (compared to the axial component) that it can be neglected in the convective term,
3. the thermal conductivity of the electrons is calculated using the Frost rules,
4. the thermal conductivity of the heavy particles is calculated using the Chapman–Enskog theory,
5. axial thermal conductivity will be neglected because the axial temperature gradient is much smaller than the radial temperature gradient,
6. T_e and T_h drop to a low temperature approaching the wall; we use $T_e = T_h = 2500$ K at r = 8 mm,
7. radiation losses are estimated to be 80 W, which is 10 % of the total measured power input of 800 W /8/,
8. the pressure equals 10^5 Pa.

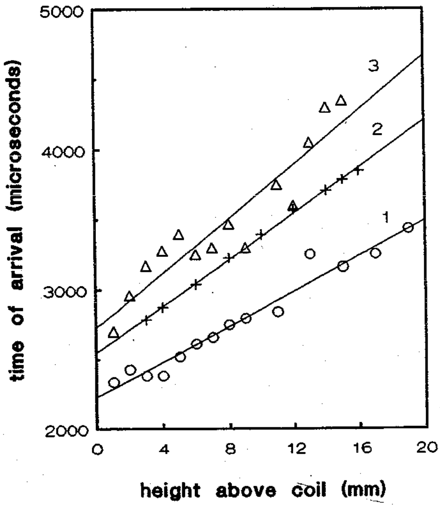

Fig. 4. Determination of the velocity by the determination of the arrival of (1) Ne pulses, (2) delayed responses to power interruption and (3) H_2 pulses. By using least squares fits we find for (1) w = 15±1 m/s, (2) 12±0.1 m/s and (3) 10±1 m/s.

By implementing all parameters we find:

$$\int_A \frac{5}{2} p \vec{w} \cdot \vec{n} \, dA \simeq 2.5 \cdot 10^5 \cdot (12-0.85) \cdot 1.1 \cdot 10^{-4} = \quad 318 \text{ W},$$

$$\int_A -\lambda_e \nabla T_e \cdot \vec{n} \, dA \simeq 0.04 \cdot \frac{6000}{2 \cdot 10^{-3}} \cdot 7.5 \cdot 10^{-4} = \quad 90 \text{ W},$$

$$\int_A -\lambda_h \nabla T_h \cdot \vec{n} \, dA \simeq 0.2 \cdot \frac{4000}{2 \cdot 10^{-3}} \cdot 7.5 \cdot 10^{-4} = \quad 300 \text{ W},$$

$$\int_V Q_{CR} \, dV \simeq \quad 80 \text{ W},$$

so the power input is:

$$\int \sigma E^2 \, dV \simeq \quad 788 \text{ W}.$$

This result agrees surprisingly well with the experimental value, although we must realize that we probably overestimated the thermal conduction in radial direction because the temperature is smaller in the lower part of the integration volume, while we underestimated (by neglecting) radial convective energy transport and axial thermal conduction.

As a final conclusion we want to state that we measured the drift velocity of the plasma by inducing small disturbances in the plasma and that applying these results gives valuable information about the energy balance

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