Experimental quantification of the transient heat flux transferred to the electrodes in a carbon nanotubes synthesis reactor

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Abstract. The aim of this work was to determine the energy exchange between the plasma and the electrodes in a carbon nanotubes synthesis reactor by applying an inverse method. Argon was used as plasma gas and the cylindrical electrodes were made of graphite. A space-marching method was used to recover the transient temperature profile in the electrodes and then to quantify the applied heat flux at their surfaces. Due to the high emissivity of graphite, lateral radiation of the electrodes was taken into account in the inverse model. The estimated heat flux deduced from the inverse method was validated by measuring the arc radiation losses obtained with radiant flux sensors. This approach provided power balance in the system. More than 60% of the power injected was transferred to the electrodes.

1. Introduction
Thermal plasmas created by an electrical arc are present in numerous industrial applications [1-3] and in laboratory experimental setups [4-10]. For many decades, experimental studies have been run to understand the phenomena occurring in arc plasma in order to optimize industrial processes such as cutting and welding systems, to enhance the quality of devices like circuit breakers and the resistance of different materials submitted to extreme conditions (aircraft under lightning, high-energy reactors, carbon nanotubes synthesis, etc. [e.g. 4, 11, 12]) and to validate numerical models. One point of interest concerns investigating heat transfer between the plasma and the electrodes. Carbon nanotubes synthesis by electrical arc is relatively recent; it is cheaper and more reliable than chemical approaches [12, 13]. However, the complexity due to numerous combinations of conditions and parameters [4, 11] implies that synthesis is still very difficult to control. Carbon nanotubes production is mainly influenced by anode erosion and by the plasma temperature gradients. An understanding of arc-electrode energy transfer provides a better interpretation of the results [4, 14]. This work deals with the experimental quantification of the heat flux deposited by thermal plasma on the electrodes in a carbon nanotubes reactor. The heat flux cannot be determined by direct measurement as sensors cannot resist...
the high temperatures involved. Several numerical studies on arc power balance can be found in the literature [15-18], but only few deals with experimental measurements of the high heat flux deposited by the plasma on the electrodes. Direct measurement based on calorimeter devices [19] is one simple way to access the anode heat flux; this can also be used with an inverse method in more complicated systems [20]. In our configuration, due to lateral radiation losses, such approach was not used. Specific sensors like thermocouple-based devices [21, 22], sweeping wire devices, water-cooled probes [23], or calorimeters [24] could be exposed to the plasma flow but not for carbon nanotubes synthesis, it will modify the system configuration. We therefore decided to use an inverse method which consists of reconstituting the deposited heat flux profile by using measurement data taken from a more reachable zone of the electrodes. To this end, thermographic techniques and the data acquired were used in an inverse model for transient heat flux reconstitution. Similar investigations have involved a JET tokamak to control the flux deposited on tokamak’s elements exposed to the plasma [25]. In that study, there was no need to consider material losses by radiation. Nevertheless, in our configuration, due to the nature of the electrodes and their high emissivity, the radiation on their lateral surfaces is taken into account in the model. In parallel with temperature history acquisition, the radiation power of the arc is recorded to be compared with the estimated heat flux. In the next sections, the experimental methods and the associated instrumentation will be presented before analyzing the results.

2. Description of the problem
The problem consists of measuring the transient heat flux deposited on the anode surface (lower electrode position) and on the cathode (upper position) by the thermal plasma. Due to the high flux level, direct measurement with a sensor cannot be performed and an inverse model is proposed (Figure 1).

![Figure 1. Illustration of the problem](image)

For the carbon nanotubes reactor considered, the electrodes were made of graphite cylinders 6mm in diameter and about 7cm in length. The electrodes are assumed to be non-deformable so their lengths remain constant with time. Due to the electrode dimensions, combined with the fact that the thermal conduction inside the graphite electrode is mainly in the axial direction, the system can be assimilated to a one-dimensional problem. The electrode is thin enough to be able to ignore convection exchange with the ambient gas. However, graphite is a very emissive material and the thermal radiation from its surface should be taken into account.

3. The space-marching method
One-dimensional inverse problems have been the subject of many works using different kinds of methods and various approaches [12-19, 24, 31-33]. We used the method described by Raynaud [30] because of its non-iterative approach. It is a variant of the space-marching method established by D’Souza [32] with a Crank-Nicolson implicit scheme. To reconstitute the transient profile of the applied heat flux, the temperature histories in two points of the electrode must be known. Therefore
the method divides the calculation domain into two zones: the direct zone, delimited by the two sensors, and the inverse zone between the first sensor and the active surface. First, the direct zone was dealt with and the temperature map versus time was deduced from the measured temperatures. Therefore, the inverse method was solved by scanning the inverse zone step by step starting from the first sensor location and moving towards the surface as shown in figure 2.

![Figure 2. (a) The direct and the inverse zone. (b) The space marching algorithm](image)

The method proposed is simple and straightforward to implement since the characteristics of the material are known. Both the direct and the inverse zones are solved starting from the energy balance in each cell at each time. The smaller the time step $\Delta t$, the greater the accuracy of the method and the faster the temporal variations that can be reconstituted. However this time resolution is limited by the dimensionless time step $\alpha \frac{\Delta t}{E^2}$ where $\alpha$ is the diffusivity of the material [m².s⁻¹], $\Delta t$ the time step [s] and $E$ [m] the distance from the electrode surface to the first captor position. This expression is indirectly correlated to the Fourier number $\mathcal{M} = \frac{\Delta t}{\Delta x^2}$ ($\Delta x$ the space step size).

So, for a given problem, the space and time resolution should be considered to obtain a compromise between the stability, the precision and possibly the computing time. Numerical tests and parametric studies were investigated in a previous study to find the best combination of parameters for a good experimental result [34]. For the inverse method, the thermophysical properties of the material must also be known. The heat capacity and the thermal conductivity that we used come from the Touloukian, the Janaf and the NIST databases [35-37].

4. Experimental setup
Measurements were made in a fullerene and carbon nanotubes synthesis reactor [4]. The gap between the 6mm-diameter graphite cathode and anode was roughly 5 to 6mm. The plasma medium was composed of argon at atmospheric pressure. Arc voltage and current were measured to calculate the total power injected into the system.
The temperature measurements were made with a FLIR SC6000 infrared camera. In figure 3 (left side) we can observe the experimental setup consisting of the reactor and the camera. The camera lens was located 40cm from the electrode and had space resolution of about 0.2mm for this configuration. The two points of measurement should have temperatures in the range in which the camera operates (600°C-1200°C).
Estimation of the spectral emissivity of the electrodes
To obtain accurate temperatures, the emissivity of the object needs to be known in the interval 3µm-5µm. So, the camera emissivity entry is adjusted to correspond to the temperature given by one thermocouple. This approach allows us to estimate the spectral emissivity of the electrode at 0.85 for the spectral range of the camera. This value is very close to that given by other authors [38, 39].

Radiative power measurement
As convection in the system is assumed to be negligible, the total power injected into the arc should be the sum of the power transferred to the electrodes and the power lost by the arc through radiation. $P_{\text{arc}} = V_{\text{arc}} I_{\text{arc}} = P_{\text{electrodes}} + P_{\text{rad}}$. This expression was used to cross-check the estimated heat flux. We therefore used radiant flux sensors with a large spectral width (0.1µm-12µm). Due to the anisotropy of the arc radiation, the sensors were placed along the azimuthal direction (figure 4). The high emissivity of the graphite in the wavelength range of the captors, implies they receive the flux not only from the arc but also from the electrodes.

![Figure 3](image-url) (a) The IR camera and the reactor (b) Cross-sectional schema of the reactor

![Figure 4](image-url) Assembly of radiant flux sensors
The percentage radiation emitted by the electrodes was obtained by masking them with a refractory material (clay) as shown in Figure 5. The masks were placed far enough away to keep them cool and to avoid fusion. Two other sensors were placed above and below to check that there was no flux produced by any reflection phenomena on the reactor wall. If we note $F$ and $F'$ the flux received by the central captor respectively without and with a mask, the ratio $F/F'$ represents the percentage of arc radiation in the total measured flux. This ratio is equal to 40% and is assumed to be the same for each sensor.

5. Heat flux quantification

In this section we report the results of applying the inverse method to experimental data. The temperatures were picked up 20mm and 25mm from the anode surface, and 10mm and 15mm from the cathode tip (Figure 6). We worked with a DC power supply with current varying from 20A to 50A in 10A steps. The current intensity range was chosen so as to avoid electrode erosion. Indeed this physical mechanism, which could change the electrode length, is not taken into account in our inverse method.

Multiple temperature channels are proposed with the camera. We selected the range [600°C-1200°C] as it simultaneously provides four appropriate measurement points for the cathode and the anode. Two cathode configurations were tested: one where the end of the cathode was flat and the other where cathode was pointed. The pointed cathode tip provided a more stable arc but the results for both setups were practically the same.
Below (Figure 7) we report the results obtained introducing experimental data into the inverse model. Figure 7a gives the temperature histories by thermography. The power transferred by the plasma to the electrodes is plotted in figure 7b. It is deduced from the integration of the calculated heat flux over the effective section of the electrodes. The uncertainty affecting the results is mainly due to the position of the measurement points for temperature assessment (±0.2mm) and to the spectral emissivity of the electrodes (between 0.83 and 0.87). To determine the uncertainty, the method was applied with the two extreme combinations. The ratio between the power deposited in the electrodes and the total power injected was between 60% and 80% (Figure 7c). The incident power to the cathode represented only a small proportion and was only weakly dependent on the power injected. Most of the power deposited was focused on the anode. Figure 7d shows that the variation of the anode power versus the arc current was practically linear, as reported in other experimental works [10, 19, 40].

![Figure 7](image-url)
To check our results, we plotted (figure 8) the total power injected into the system, the total power estimated from the inverse method giving the power transferred to the electrodes and the radiation power. The calculated total power was slightly underestimated. This could be because we chose to ignore convection.

6. Conclusions
Experimental measurements were applied to quantify heat transfer between plasma and electrodes in a carbon nanotubes reactor. A space-marching inverse model was associated to thermographic acquisition to assess the incident heat flux on the surface of the electrodes. In parallel, arc radiation losses were measured to check the results. This allowed us to calculate the power balance in the configuration considered. Good agreement was found between the radiation losses measured directly and those calculated from injected power minus the power transferred to the electrodes. The power transferred to the electrodes was estimated at about 60% of the injected power. This approach can be extended to other problems since the material properties are known. Before being applied as a metrology tool for carbon nanotubes monitoring, the method needs to be improved by taking the erosion into account. The inverse method should be suitable, and the next step of the study will focus on erosion.

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