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The removal of co-deposited hydrocarbon films from plasma facing components using high-power pulsed flashlamp irradiation

K.J. Gibson [a,*], G.F. Counsell [b], C. Curran [c], M.J. Forrest [b], M.J. Kay [a], K.G. Watkins [c]

[a] *Department of Physics, University of Manchester Institute of Science and Technology (UMIST), Manchester, M60 1QD, UK*
[b] *EURATOM/UKAEA Fusion Association, Culham Science Centre, Oxon OX13 3DB, UK*
[c] *Department of Engineering, University of Liverpool, Liverpool, L69 3GH, UK*

Abstract

The use of carbon-based materials for first wall components in tokamaks results in the formation of hydrocarbon deposits on divertor components that could lead to a high level of tritium retention in future fusion devices. Experiments at UMIST have demonstrated that photonic cleaning using high power Xenon flashlamp sources is an efficient method for removing such films and represents a good candidate technology for international thermonuclear experimental reactor (ITER) operations. Studies have shown that effective film removal occurs at a fluence threshold of between 1.9 and 2.5 J/cm^2. The by-products of the cleaning process, both particulates and gases, have been characterised using particle sizing spectrometry and quadrupole mass spectrometry respectively. It is found that hydrogen, methane, acetylene, ethylene, ethane and carbon dioxide are the principal gaseous products produced during the cleaning process, which also produces a significant fraction of particulates in the size range 2–20 μm.

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1. Introduction

The use of carbon-based materials for first wall components in the divertor region of tokamaks results in the formation of hydrocarbon film deposits on plasma facing components and sub-divertor assemblies. The form of film that can result from such ‘co-deposition’ can vary greatly from hard films with H/C ratios of 0.4 to polymer-like films with H/C ratios approaching 0.8 [1]. As well as leading to enhanced levels of debris at the edge plasma regions (and hence influencing plasma performance [2]), such films have been shown to produce high rates of in-vessel tritium retention [3] which would be problematic in future devices. Calculations for ITER predict an accumulation of 10–20 g of tritium per 1000 s pulse [4,5], whilst the in-vessel inventory for ITER is approximately 1 kg. It is clear that the unwanted deposition of hydrocarbon films in tokamaks is a key area of study for future fusion devices and a

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[*] Corresponding author. Fax: +44 161 200 3941.
E-mail address: k.gibson@umist.ac.uk (K.J. Gibson).

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crucial factor in determining their efficient operation. In particular, the development of techniques for the removal of such films is a critical issue [5].

Photonic cleaning has been demonstrated to provide an efficient method for removing hydrocarbon films [6] and represents a good candidate de-tritiation technology for ITER [7]. Although much of the work has been based on laser irradiation, there has been increasing interest in utilising high-power xenon flashlamps for photo-cleaning since such technology could lead to systems that are more easily scaled to high average powers. In this paper we report on studies of flashlamp removal of hydrocarbon films from carbon fibre composite (CFC) substrates in inert atmosphere and in vacuum. In particular, we focus on the threshold fluences for effective film removal and the characterisation of the cleaning by-products using mass spectrometry and particle sizing spectrometry.

2. Experimental set-up

To study the effectiveness of flashlamp cleaning, tile samples coated with hydrocarbon films were irradiated in a test chamber offering optical, probe, and vacuum diagnostic access. The stainless steel vacuum vessel used is approximately 650 mm long by 275 mm diameter, taking the form of a 6-way cruciform. This chamber can be evacuated to base pressures of typically around 5×10^{-7} mbar but also allows experiments to be performed at atmospheric pressure in an inert gas environment. The tile samples used for this work were 50 mm × 50mm square machined from carbon fibre composite. To produce a deposited film, the tiles were subjected to a plasma-assisted chemical vapour deposition process (PaCVD) using an acetylene discharge. The resulting hydrocarbon coatings were between 11 and 14 μm thick. Fig. 1 shows a scanning electron microscopy (SEM) image of the coating, revealing a structure quite similar to deposits found on tiles in tokamaks (for e.g. [8]).

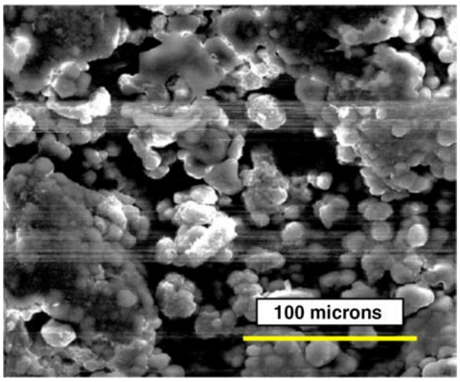

Fig. 1. Scanning electron microscopy (SEM) image of the synthesised hydrocarbon coatings used for these studies.

The photo-cleaning source is based around a Xenon flashtube having an active length of about 50 mm and a 9 mm bore: the flashtube envelope and the surrounding cooling jacket are made from fused quartz silica providing high optical transmission in the UV and visible. The tube was mounted at one focus of an elliptical aluminium reflector with the sample tile mounted at the other focus: the whole assembly is then mounted within the test chamber. The flash-lamp power supply was capable of providing fixed pulse widths of approximately 100 μs with a repetition rate of up to 10 Hz at a maximum electrical energy of 100 J per pulse.

Gaseous by-products during flashlamp operation under vacuum were monitored using a quadrupole mass spectrometer capable of acquiring full mass scans (1–50 amu) at a maximum rate of just under 2 Hz. In addition, the instrument can be used to follow time histories of individual species with a maximum time resolution of 0.3 ms: in practice the overall response time is longer than this and is determined by the gas throughput to the system.

Particulates that are produced during flashlamp cleaning are detected using a Grimm Aerosol Technologies Model 1.108 aerosol particle sizing spectrometer. Although this device is not compatible with vacuum operation, it provides detailed information on particle emission during photo-cleaning in inert gas environments. For these studies, particles were drawn via a volume-controlled pump into a radial sampling head located adjacent to the irradiated tile. The sampled particulates enter a laser scattering chamber where they are counted in one of 15 separate particle size channels ranging from less than 0.3 μm up to 20 μm.

3. Results

We first consider the results of mass spectrometry of the gaseous by-products produced during flashlamp cleaning. Fig. 2(a) shows a mass spectrum over the range 0–45 amu for a single 100 J pulse of the flashlamp: three curves are shown corresponding to the background, irradiation of the hydrocarbon coating and irradiation of the underlying substrate.

The most significant mass peaks obtained during irradiation of the coating are mass 2 (hydrogen), mass 16 (CH_4 and cracking products at masses 12–15 produced within the spectrometer), mass 26 (C_2H_2 and cracking products), mass 28 (either N_2 or C_2H_4), mass 30 (C_2H_6) and finally mass 44 (which we ascribe to CO_2). Thus it would appear that the major gaseous hydrocarbons produced during the cleaning process are methane, acetylene, ethylene and ethane. However,

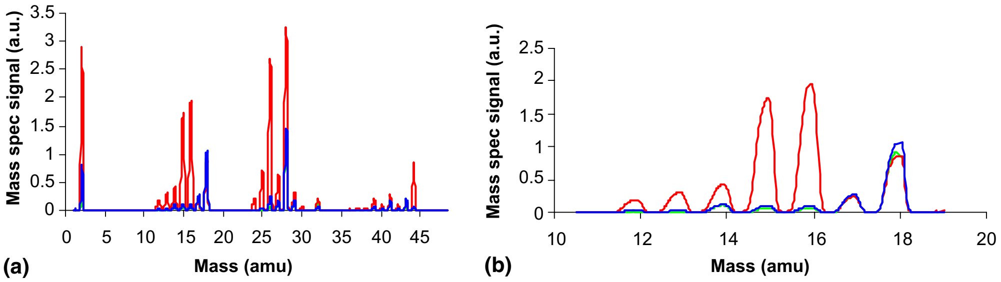

Fig. 2. (a) Mass spectrum covering the range 0–50 amu. The three traces correspond to background (green), flashlamp irradiation of coating (red) and flashlamp irradiation of the substrate (blue). (b) Detail of the same spectrum for the mass range 10–20 amu. For interpretation of the references in color in the figure legend, the reader is referred to the web version of this article.

it should be noted that the mass spectrometer only detects products having low sticking coefficients since the sampling orifice of the mass spectrometer is some distance from the tile sample. Higher sticking coefficient products, such as reactive species (e.g. C, CH, C_2H etc.) or higher mass alkanes will be absorbed onto the wall before entering the spectrometer. The absence of such products from the mass spectrometer trace does not necessarily infer that they are not produced in the photo-cleaning process.

Fig. 2(b) shows the detail over the range 10–20m/z for the spectrum in Fig. 2(a). We note that the mass 18 peak (H_2O), and the associated cracking peak at mass 17, is largely the same for all cases, suggesting that photo-cleaning in this regime does not lead to the production of water. This is a significant result for the removal of tritiated films in next step devices where the production of T_2O would be undesirable. This finding is in agreement with Shu et al. [6] who also report no increase in the partial pressure of water in experiments where an excimer laser was used to irradiate co-deposits on carbon tiles from JT-60U. The most striking feature of Fig. 2(b) is the very large methane (mass 16 and cracking products down to mass 12) signal during coating removal, compared with both the background case and the irradiation of the substrate. The difference between the irradiation of the hydrocarbon film and the uncoated substrate suggests that the methane signal can be used as a signature of coating removal, as well as having the potential to be used to determine when removal is completed for future full scale cleaning experiments.

The correlation between coating removal and the methane mass spectrometer signal can be used to infer the scaling of cleaning efficiency as a function of flashlamp fluence. Fig. 3 shows the methane signal produced for single shot flashlamp energies ranging from 20 to 100 J (for each measurement the flashlamp was repositioned onto a new area of coated tile). It can be seen that there is apparently a threshold for effective cleaning, with the results indicating a marked increase in the production of methane at fluences between 1.9 and 2.5 J/cm^2. At the highest fluences the rate of removal is such that the coating is completely removed (as determined subsequently using SEM) within between 5 and 10 flashlamp pulses.

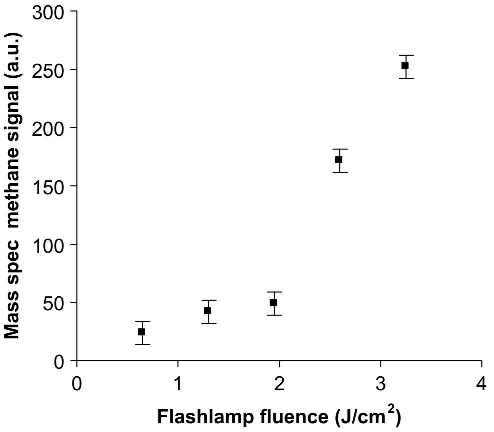

Fig. 3. Methane (mass 16) mass spectrometer signal level as a function of the flashlamp energy fluence, indicating a threshold for coating removal of between 1.9 and 2.5 J/cm^2.

As well as producing gaseous products, it is known that photo-cleaning of films can produce copious quantities of particulates [9]. To characterise the particulates produced by flashlamp cleaning, a series of experiments were performed in Argon at atmospheric pressure so that the aerosol spectrometer described previously could be used to determine the size distribution of the particles. Fig. 4(a) shows an example of a spectrum obtained (with background for comparison) for a single flashlamp pulse at an energy of 100 J.

It is apparent that irradiation of the coating produces a significant increase (up to 100 times) over the background in the population of sub-micron particles as well as generating a population of larger (>5 μm) particles. The origin of this strongly bi-modal distribution is not

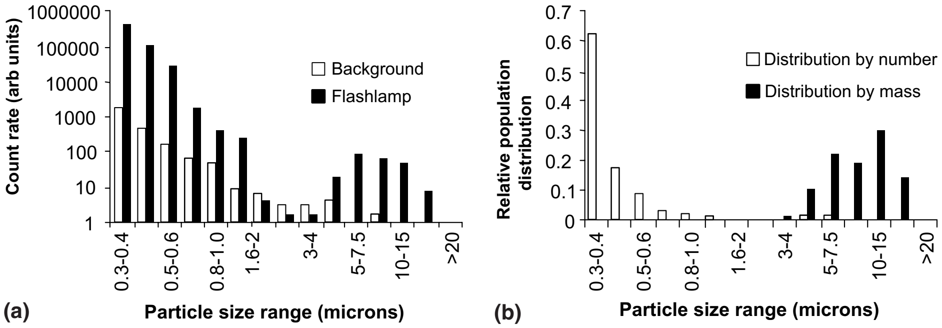

Fig. 4. (a) Particle size distribution for particulates produced during a single 100 J flashlamp pulse and (b) normalised distributions by particle number and by mass content (assuming uniform particulate density) for the same results.

clear at this stage and will require further work. A possible explanation is that the distribution originates in the microstructure of the coating which is found to take the form of large globular clusters made up of assemblages of smaller particles in the size range 5–10 µm (see Fig. 1). It might be that the larger particles that are observed in the spectrum originate from the break up of these clusters, producing particles of the order of a few microns. The production of smaller particles may be the result of fracturing these clusters or may be characteristic of a separate component of particles originating from the CFC substrate.

The presence of such a large population of sub-micron particles will have an important influence on the design of removal and filtration systems that will be required for any future in-vessel flashlamp cleaning. However, it is important not to overstate the significance of this class of particulates. The results presented in Fig. 4(a) represent the distribution of *number of particulates* of different sizes, and not the distribution of *mass* across the particulates. To illustrate this, Fig. 4(b) compares both classes of distribution, each normalised to unity, again for the case of a single 100 J flashlamp pulse. In producing this figure, the data for particle size distribution is simply taken from Fig. 4(a), whilst the relative mass distribution has been calculated on the basis of a uniform density for all particle sizes (i.e. assuming all particles produced during removal originate from the same material). When the relative mass of particles of different sizes is taken into account, then from the point of view of mass-removal during photo-cleaning the most significant particulate size range is between 5 and 20 µm, with the mass removed associated with particles having sub-micron size being less than 1%. Although further work on the composition of both size groups is needed, it is clear that even if the density of the smaller group of particles was several times that of the larger particles, the bulk of the mass of the removed particulates lies in the range above about 4 µm.

Finally it is possible to follow the time history of the particle distribution during flashlamp cleaning. For these studies, coated tiles are irradiated with 100 J pulses at a repetition rate of 8 Hz. Fig. 5 shows this data presented in the form of the time histories of the total number of particles in each of the two size groups shown in Fig. 4(a). After the initial burst of particles produced when the flashlamp is turned on, both the 'large' particles (>4 µm) and the sub-micron particles decrease with approximately the same characteristic time (10 s). Following full cleaning of the tile, we observe that the 'large' particles remains close to the background level whilst the number of sub-micron particles begins to rise to high levels again. Although further investigation is needed, we speculate that this may be due to some surface ablation of the exposed CFC substrate.

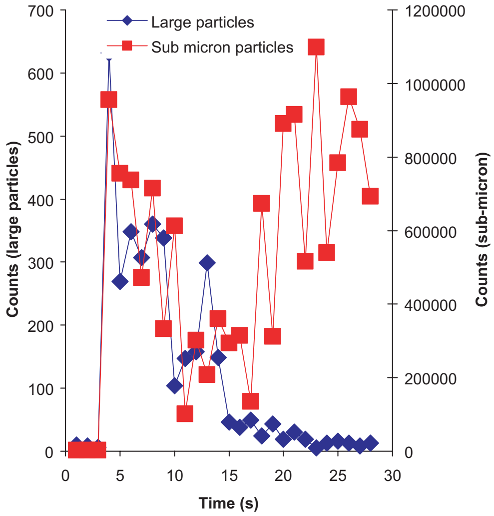

Fig. 5. Time histories of the total number of particles in the two particle size groups during flashlamp cleaning with 100 J pulses at 8 Hz repetition rate.

4. Summary

The use of flashlamps for photonic cleaning of hydrocarbon films from CFC substrates has been demonstrated in a series of laboratory experiments. Mass spectrometry has demonstrated that hydrogen, methane, acetylene, ethylene, ethane and carbon dioxide are the principal gaseous by-products produced. Of these species, methane provides a means of assessing the rate of coating removal and on the basis of the scaling of the methane signal with flashlamp energy, the fluence threshold for cleaning with single pulses has been found to be between 1.9 and 2.5 J/cm^2.

Particle size spectrometry has shown that there are a significant number of particulates produced during photo-cleaning. The distribution in particle sizes is found to have the form of a bi-modal distribution with a small fraction of particles having sizes greater than 5 μm, and a large number of sub-micron particles forming a second distinct population. When evaluated in terms of mass removal from the coatings, the bulk of the particulate masses are in the larger of the two size groups. Time resolved measurements of particulates indicate that the evolution of the larger particle size group (4–20 μm) is well correlated with the overall rate of coating removal.

Future work will focus on more detailed studies of the removal process, particularly to gain an understanding of how the properties of the flashlamp source are related to the film removal rate and the relative proportions of gaseous and particulate cleaning by-products. In addition, a series of experiments are proposed for the demonstration of in-situ coating removal on JET using an upgraded flashlamp source.

Acknowledgments

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