Laboratory experiments on cluster/aerosol formation by colliding ablation plumes

Y Hirooka1,4, K A Tanaka2, H Sato2, K Ishihara2 and A Sunahara3

1National Institute for Fusion Science, 322-6 Oroshi, Toki, Gifu 509-5292, Japan
2Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan
3Institute for Laser Technology, 2-6 Yamadaoka, Suita, Osaka 565-0871, Japan

E-mail: hirooka.yoshihiko@nifs.ac.jp

Abstract. First-of-a-kind experiments on cluster/aerosol formation by colliding ablation plumes have been conducted, radiating Al, Cu and C with 3ω-YAG laser at power densities between 2~30 J/cm²/pulse. Visible spectroscopy indicates that the excitation light intensities of Cu and Al plumes are not necessarily be doubled in collision, but can rather be weakened due to atomic and molecular reactions. For colliding C plumes, Swan band radiation has been observed, indicative of C2 and/or C2+ formation, and ion mass spectrometry has identified Cn+ clusters, including C+, C2+, C3+, C4+ and C5+. From ICCD camera observations, C plumes generated at power densities above ~15 J/cm²/pulse tend to split into two components with respective velocities, only the slow component of which appears to be interactive to form clusters. Nano structures like CNT have been identified in deposits from colliding C plumes.

1. Introduction

It is predicted that, along with DT-pellet implosions in inertial fusion power reactors, target chamber walls will repeatedly be exposed to intense pulses of 14MeV neutrons, X-rays, high-energy unburned fuel particles and pellet debris such as hydrocarbon fragments. As a result, wall materials will be subjected to thermo-physical erosion due to evaporation, sputtering and ablation, etc.

Eroded materials will either be re-condensed elsewhere on the wall after travelling across the chamber or collide with each other perhaps in the centre or the symmetric axis region to form clusters which can grow into aerosol to float until it is pumped out. These processes directly affect the wall lifetime as well as reactor performance in terms of pulse repetition rate because laser can be reflected, scattered or refracted by floating aerosol, which hinders subsequent implosions. Despite its critical importance, this chamber clearing issue has not clearly been addressed in the ICF research community. The present work is intended to investigate fundamental aspects of materials ablation behavior with the particular emphasis on cluster/aerosol formation by colliding plumes.

2. Experimental

A new setup shown in figure 1 has been put together for the Laboratory Experiments on cluster/Aerosol Formation by Colliding Ablation Plumes (to be referred to as LEAF-CAP in the remainder of the paper). In this setup, a 3ω-converted YAG laser beam (355nm, 320mJ/pulse, 6ns, 10Hz) is split into two equal-power beams, each line-focused into ~0.1cm x ~1cm to

Figure 1 The LEAF-CAP setup.
radiate two targets at room temperature in vacuum (~10^{-5} Torr), i.e. a double target setup. These targets are in the form of rectangular disk (~5cm x ~1cm x ~0.5cm) but arc-shaped on the laser-facing side, and are positioned in such a way that two ablation plumes collide with each other in the centre-of-arc region. Used as target materials in the LEAF-CAP experiments are Al, Cu and C (isotropic graphite).

3. Results and discussion

3.1. CCD camera observations

Shown in figure 2 are some of the typical CCD camera images of laser ablation plumes of Al: (a) and (b), Cu: (c) and (d), and C: (e) and (f), all observed at 2.2J/cm²/pulse in the single and double target setups, respectively, in the LEAF-CAP experiments. Notice that Al and Cu plume emission tails seen in the single target setup appear to be cut in the middle, as shown in figures (b) and (d), when two self-plumes collide with each other in the double target setup. Interestingly, although the plasma density tends to be doubled in the centre-of-arc region [1], when plumes collide with each other, the excitation light intensity is not necessarily doubled but can be weakened in some cases, contrary to expectation, presumably due to complicated atomic and molecular reactions. In contrast, C plumes rather strongly radiate in the centre-of-arc region when they collide with each other, as can be seen in figure (f), indicative of an exothermic reaction although they are barely visible individually.

![Figure 2 Ablation plumes of Al: (a, b), Cu: (c, d) and C: (e, f) in the single and double target setups, where the distance from the target surface and plume colliding center is about 1.5cm.](image)

3.2. Visible spectroscopy

Visible spectroscopy measurements have provided data, essentially consistent with these CCD camera observations. In the case of Al, the intensity of Al-I approximately is doubled at 2.2J/cm²/pulse (not shown here), whereas in the case of Cu, as shown in figure 3(a), the intensity of Cu-I at the same power density becomes smaller in the double target setup. In the case of C, the Swan band structure is clearly identified in spectra, indicating the formation of C₂ and/or C₂⁺ when plumes collide with each other. Presumably related to C_n and/or C_n⁺ cluster formation to be described in 3.3, however, the line radiation marked with ? at around 580nm is yet to be identified in these spectra.

![Figure 3 Visible spectroscopy of (a) Cu plumes at 2.2J/cm²/pulse and (b) C plumes at 6.5 J/cm²/pulse in the single and double target setups.](image)
3.3. Ion mass spectrometry
As shown in figure 1, a quadrupole mass analyzer is employed, but used without hot filament in order that ions in colliding ablation plumes can directly be identified. A comparison between the mass spectra taken for Al and C plumes, all generated at 2.2J/cm²/pulse, in the single and double target setups are shown in figure 4. The formation of Al⁺ and Al²⁺ is exhibited when Al plumes collide with each other although no sign of cluster formation is seen. In the case of C plume, C₁⁺, C₂⁺, C₃⁺, C₄⁺ and C₅⁺ are clearly identified in mass spectra, which is believed to be due to their strong interaction potential, i.e. C-C bonding strength. Interestingly, Cₙ-cluster ion formation has been observed mainly at low laser power densities where the ion energy is also relatively low. Details will be described next.

![Figure 4](image)

**Figure 4** Ion mass spectra of (a): Al and (b): C plumes in the single and double target setups.

3.4. Plasma parameter and deposition rate measurements
For plasma parameter measurements, a linearly moveable Langmuir probe is inserted into the centre-of-arc region. Also, a quartz crystal film thickness monitor is positioned at about 2.5cm from the centre-of-arc, replacing the quadrupole mass analyzer. Data have been taken from C plumes generated at laser power densities between 2 and 13J/cm²/pulse. As shown in figure 5 (a), the plasma density appears to increase linearly on the order of 10¹⁰ 1/cc, finally reaching ~3 x 10¹¹ 1/cc, as the laser power density increases. Along with the increase in plasma density, the deposition rate also increases due to the relation: Γ = (n₀ + nᵢ)vₑ, where Γ is the deposition rate, n₀ is the neutral density, nᵢ is the ion density, and vₑ is assumed to be the velocity of C₁₀ or C₁⁺ instead of Cₙ₀ or Cₙ⁺ for simplicity. Meanwhile, the electron temperature increases from 0.5 to 3eV, as shown in figure 5 (b). Under adiabatic expansion conditions, the electron temperature at the point-of-origin for plume generation can be as high as ~15eV [1], where nᵢ << n₀ readily holds [2], so that the deposition rate may be given by: Γ ≅ nᵢvₑ. As plotted in figure 5 (b), the ion temperature has been found to vary from 0.1 to 0.3eV, based on the relation: kTᵢ = mᵢvₑ², where mᵢ is assumed to be the mass of C₁₀ or C₁⁺ for simplicity.

![Figure 5](image)

**Figure 5** Plasma parameters and deposition rates measured for C plumes at 3-13J/cm²/pulse.
3.5. ICCD camera observations

Shown in figure 6 are a series of the ICCD camera images taken for C plumes generated at 29J/cm$^2$/pulse in the double target setup. Notice that there are two distinctive components with their respective velocities, as seen in figure 6 (a). The kinetic energy estimated from these ICCD image data for the fast component is $E_f \approx 10\text{eV}$ and that for the slow component is $E_s \approx 1\text{eV}$. One possible mechanism to explain this phenomenon is as follows [3]. Up on materials ablation, electrons tend to be emitted first, which will then drag ions out of the energy deposited volume. Multiply charged ions, if they are formed, will be attracted to these electrons more strongly than singly charged ions, leading to the separation of plumes. Here, one might conjecture that the formation of multiply charged ions would require power densities above a certain threshold. The threshold energy density in the present work seems to be around 15J/cm$^2$/pulse, which is relatively in agreement with earlier studies [4].

Interestingly and perhaps most importantly from the ICF reactor operation point of view, the fast components don’t seem to collide but to miss each other, as can be seen in figure 6 (b), whereas the slow components apparently collide to interact, slowing down even further with strong radiation, as shown in figures 6 (c, d, e, and f). After the slow down, the directional kinetic energy of the combined plumes falls down on the order of 0.1eV, related to the deposition rate data analysis described in 3.4. Corroborating these findings, the ion-ion collision mean free paths for the fast and slow components are of the order of 10cm and 0.1cm, respectively, both estimated from $\lambda_{ii}[\text{cm}]=10^{12}T[\text{eV}]/n[1/\text{cc}]$ [5].

![Figure 6](image)

Figure 6 ICCD images of C plumes at 29J/cm$^2$/pulse in the double target setup.

3.6. Ablation deposits analysis

Carbon deposits generated after repeated colliding plume experiments have been analyzed with TEM. As shown in figure 7, data indicate the presence of nano structures like CNT (for carbon nano tube). From elemental analysis, other than carbon, no impurities have been detected in these deposits. The formation of these nano structures apparently favour low power density ablation conditions although details are still unclear at this point.

![Figure 7](image)

Figure 7 CNT-like nano structures.

4. Conclusion

To address the ICF chamber clearing issue, the first-of-a-kind demonstration has been performed, using a laboratory-scale experimental setup in which two laser ablation plumes collide with each other. Chosen as the target materials are Al, Cu and C. Data indicate that under identical conditions only colliding C plumes exhibit the formation of clusters. Also, cluster formation has been found to favour relatively low kinetic energy plumes. From these findings, one predicts that materials re-condensation as well as cluster/aerosol formation will most likely occur in ICF target chambers with carbon armors.

References

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