

Application of TR-LII for the study of carbon vapor condensation at room temperature

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Time-resolved laser-induced incandescence (LII) was applied for the investigation of carbon particle formation at room temperature. A supersaturated atomic carbon vapor was generated by laser photolysis of carbon suboxide. The growth of carbon particles was observed at 0.02 – 1 ms after photolysis. Values of thermal accommodation coefficients of Ar, He, CO and C₃O₂ molecules on the carbon particles surface were determined by the comparison of TR-LII particle sizes with data from Transmission Electron Microscopy.

Introduction

The application of LII to particle-size measurements to various conditions is in the focus of many investigations. Major uncertainties in the interpretation of LII data arise from unknown particle properties like absorption coefficient, density, heat capacity and thermal energy accommodation coefficient of the bath gas molecules on the particle surface. The basic goal of this study is to combine TR-LII technique with sampling measurements to get reliable data about the sizes of carbon particles during carbon vapor condensation at room temperature.

Experimental

Carbon nanoparticle synthesis from laser-induced photo-dissociation of carbon suboxide (C₃O₂) [1] was applied for the study of carbon condensation. A quartz cell (10×10×5 mm³) was evacuated and filled with pure C₃O₂ or C₃O₂ diluted by Ar, He or CO, respectively, at variable mixture ratios and total pressures in the range from 10 mbar to 1 bar. For photodissociation of C₃O₂ radiation from an ArF excimer laser (193 nm) was used. The energy of each laser pulse was measured in front of and behind the cell to evaluate the energy absorbed by C₃O₂ molecules. The resulting C-atom concentration after photo dissociation was 1.15 – 3.4 × 10²² m⁻³.

The equipment for the TR-LII measurements consisted of a pulsed Nd:YAG laser (1064 nm) and two fast photo-multipliers for the detection of particle emission at 694 and 550 nm. The laser beam profile was a disc with a diameter of 1 mm and its energy was measured by a calibrated photo-diode for each individual pulse. The energy density of the Nd:YAG laser pulse was less than 0.4 W/m² to minimize evaporation of particulate material. LII signals were evaluated using a simple model [2]. To get the particle samples, the TEM grids were deposited on the bottom of the quartz cell and the particles were collected from gas phase under natural gravity conditions for following analysis.

Results

Particle sizes as a function of time after photolysis were determined from TR-LII measurements for different vapor concentrations and gas-diluter pressures (Fig.1). Absolute particle sizes, determined with the assumption of a thermal energy accommodation coefficient $\alpha = 1$, yielded maximum particle sizes significantly larger than the primary particle sizes observed by TEM.

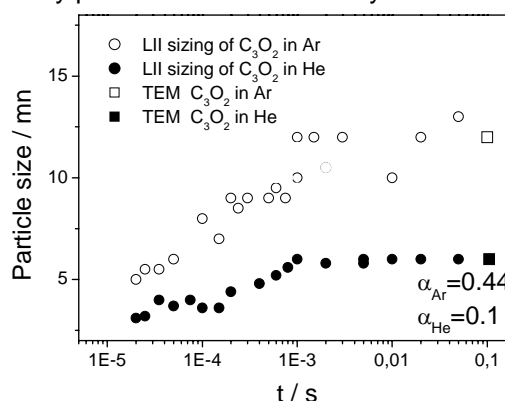


Fig. 1: Carbon-nanoparticle growth in Ar and He. Fit of the TR-LII and TEM particle sizing by thermal energy accommodation coefficients.

The difference between LII and TEM data was attributed to the variations in thermal energy accommodation coefficient α in the different bath gases. Thus, the values of α were obtained for Ar, C₃O₂, He and CO from the comparison of final TR-LII particle size with TEM data.

Gas	Ar	CO	C ₃ O ₂	He
α	0.44	0.44	0.51	0.1

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References

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