In-situ Laser Diagnostics of Gas-Phase Kinetics during Carbon Nanotube Synthesis from Hydrocarbon Pyrolysis

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Abstract: In-situ laser and optical diagnostic techniques provide spatially and temporally resolved measurements of gas-phase species concentrations, temperature gradients, and nanoparticle sizes within the FCCVD reactor. This study investigates the measurement of spatially-resolved gas temperature within a carbon nanotube synthesis reactor involving plasma-generated iron catalyst nanoparticles, using Coherent Anti-Stokes Raman Spectroscopy (CARS).

1. Introduction

Carbon nanotubes (CNTs) exhibit exceptional mechanical, electrical, and thermal properties, making them valuable across diverse applications [1]. This study employs nanosecond CARS combined with LIF/TALIF for the determination of gas phase kinetics at play during the synthesis of CNT from hydrocarbon pyrolysis, with iron and sulphur-containing gases leveraged as catalyst and growth promoter, respectively. These advanced diagnostics will enhance understanding of CNT nucleation and growth dynamics, optimizing synthesis conditions for scalable, high-quality CNT production. We are particularly interested in understanding why only about 0.5% of iron nanoparticle catalysts are effectively utilized as supports for carbon nanotube (CNT) growth [2].

2. Methods

The N_2 degenerate CARS setup in an optically accessible reactor is pictured in the top part of Figure 1. It comprises four main components: (i) an injection-seeded pump/probe laser (Q-switched Nd:YAG, 532 nm, 8 ns pulse duration), (ii) a broadband Stokes beam around 607 nm generated by an OPO laser (RADIANT QX4130), (iii) beam management optics, and (iv) a detection system combining a grating spectrometer and an ICCD camera. The pump/probe and Stokes beams, with orthogonal polarizations, are combined using a dichroic mirror and directed into the reactor, where the four-wave mixing CARS signal is generated. The probed volume size can be adjusted using either collinear or BoxCARS phase-matching geometries.

3. Results and Discussion

The bottom part of Figure 1 illustrates a sample CARS spectrum (without injection-seeding) recorded in room temperature N_2 at 600 Torr. By utilizing the CARSFIT simulation tool, N_2 rotational temperatures (which are proxy for the gas temperature) can be inferred from the experimentally measured spectra. The full conference contribution will include CARS-based gas temperature measurements at different spatial locations in the reactive zone of the reactor as well as intermediate species number density measurements and visualization through laser-induced fluorescence.

4. Conclusion

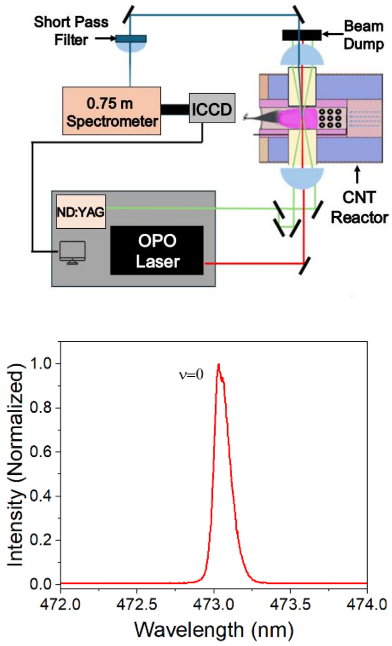

Fig. 1. Proposed CARS Setup [Top]. CARS spectrum measured in N_2 at 600 Torr and room temperature [Bottom].

We have developed a CARS setup suitable for measurements of N_2 rotational temperatures. We expect to leverage this setup to resolve gas temperature gradients within the reaction zone of the CNT synthesis reactor. These measurements, when combined with the visualization of C_2, CN and H_2 species would allow to gain insights necessary for the optimization of reactor conditions to improve CNT yield and quality.

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References

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