Pergamon

PII: S0021-8502(96)00220-0

J. Aerosol Sci., Vol. 27, Suppl. 1, pp. S295–S296, 1996

0021-8502/96 $15.00 + 0.00

Raman Spectroscopy as an On-line Measurement Technique in a Laser-CVD Reactor During Production of Si_3N_4

I.L. Tuinman, J. Veenstra, J.C.M. Marijnissen, B. Scarlett and J. Schoonman

Delft University of Technology, Department of Chemical Engineering and Materials Science, Julianalaan 136, 2628 BL, Delft, The Netherlands

Keywords: Laser CVD, Raman spectroscopy, silicon nitride

Because of its good high-temperature properties, silicon nitride (Si_3N_4) has good prospects as a structural ceramic material. Its low reactivity, however, demands a very small particle size (in the nanometer range) to facilitate processing, i.e. sintering, of the precursor powder. The production of this nanosized powder in a laser induced flame directly from the gas phase has been investigated with PCS and with Raman spectroscopy. The energy to induce the reaction is supplied by a CO_2-laser and this energy is transferred to the reactants silane (SiH_4) and ammonia (NH_3) by IR-absorption of the 10.6 μm wavelength radiation.

The aim of the project was to investigate all steps of the laser-CVP process. The current project studies the mixing of the reactants and the nucleation with Raman spectroscopy and CFD simulations. The results of both studies were in good agreement with each other, both showing hardly any mixing at room temperature and very fast but incomplete mixing of the reactants in heat-up zone right below the reaction zone in the lower part of the laser beam.

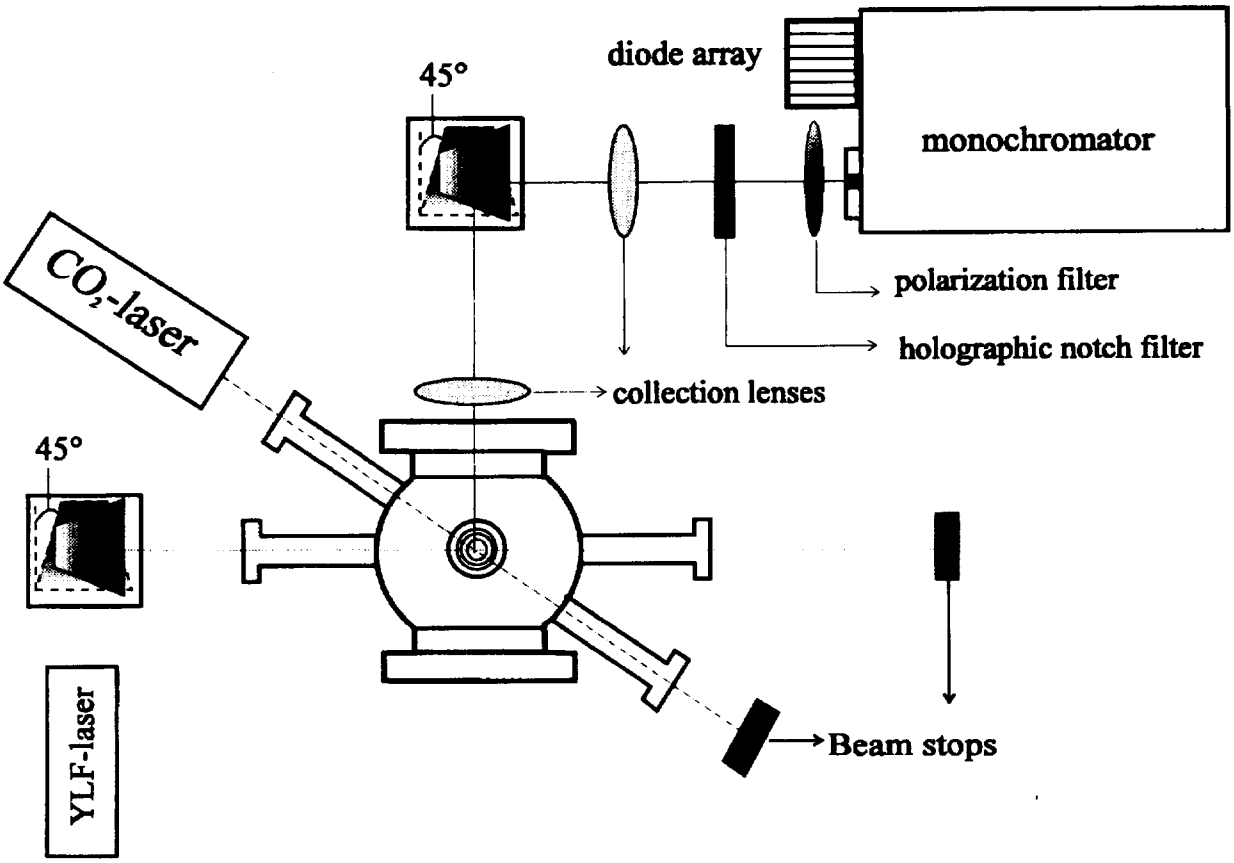

Figure 1 Top view of the laser reactor and Raman spectroscopy set-up

Figure 1 shows the reactor with Raman spectroscopy set-up. To eliminate the background signal of the flame, a gated detector system is used. The light source is a frequency-doubled, pulsed Nd-YLF laser (523 nm), the detector is an intensified diode array and for stray light rejection a holographic notch filter and single-stage monochromator are used. Both a backscattering and a right-angle configuration were applied. The backscattering spectra had a better signal to noise ratio and alignment was easier than for the right angle configuration. The right angle spectra, however, had a better spatial resolution.

The Raman show that the reaction is completed in a 0.25 mm belt (fig.2a). Besides silane, nitrogen and ammonia these Raman spectra also showed peaks belonging to NH_2, SiH_2 and other decomposition products of silane (fig. 2b).

The Mie scattering of the formed particles caused overirradiation of the notch filter. As a result the filter emitted a strong fluorescence-like signal that obscured the weak Raman signals and made measurements higher in the flame impossible.

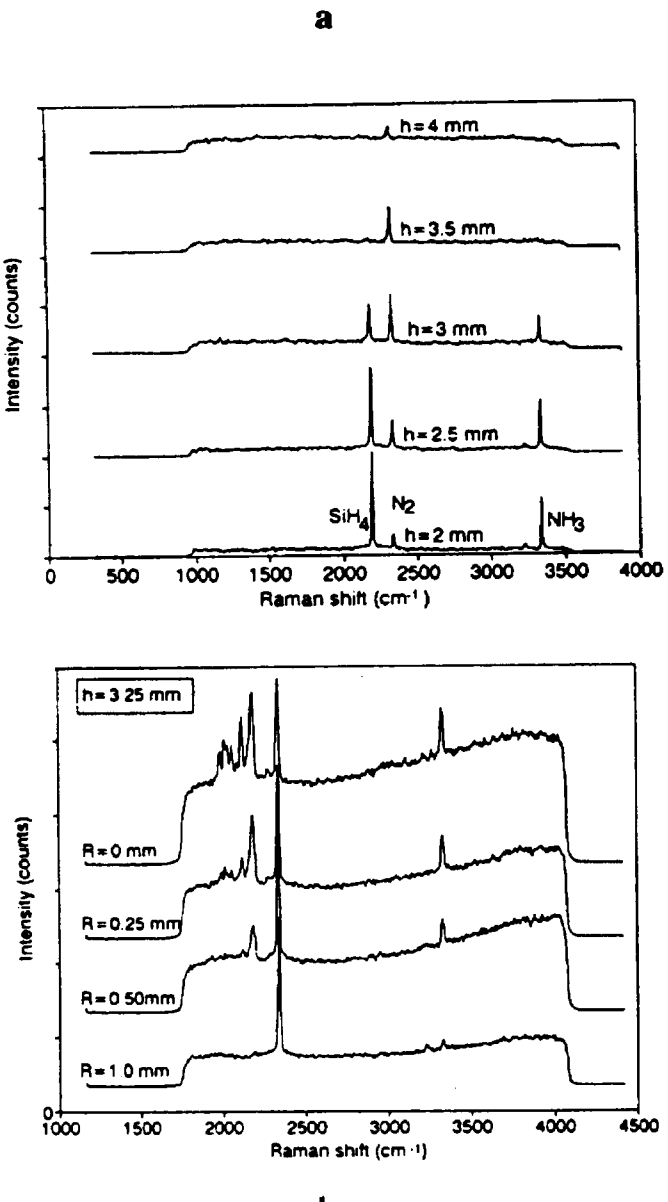

Figure 2 Raman spectra of the silicon nitride flame (a) as a function of height above the nozzles and (b) as a function of radius in the reaction zone. The radius R = 0 in the center.