Silica formation in flames with siloxane admixture: effects of admixture concentration, flame temperature and equivalence ratio

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Introduction

Biogases can play an important role in a transition away from fossil fuels, but often contain impurities such as siloxanes. SiO_2 molecules generated in the combustion of the siloxanes coalesce together into particles which subsequently form even larger aggregates and deposit on internal parts of combustion equipment. We used angle-dependent light scattering as a quicker and less invasive alternative to ex-situ methods such as TEM to investigate aggregate growth in premixed CH_4/hexamethyldisoloxane (L2)/air flames at various siloxane concentrations, flame temperatures and fuel equivalence ratios.

## Experimental setup / procedure

Measurement environment
- Burner-stabilized premixed (methane/air) flames.
- L2 added through bubbler system.
- Temperature controlled through exit velocity gas mixture.
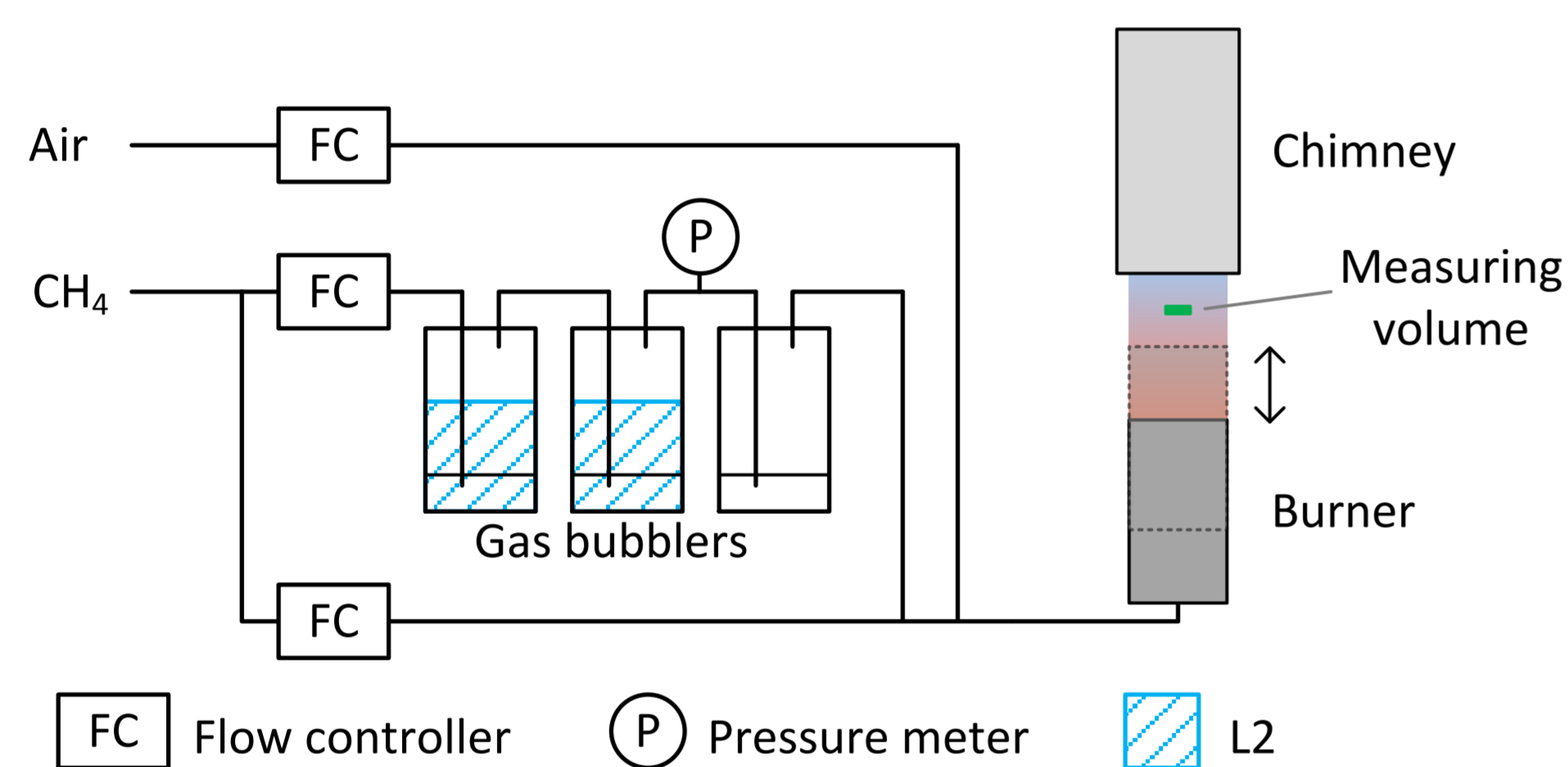
Measurement / detection
- 532 nm cw laser beam directed through the flames.
- Detection of scattered light at four angles simultaneously.
- Calibrated using Rayleigh scattering from SF_6.
- Guinier analysis tot determine aggregate size.
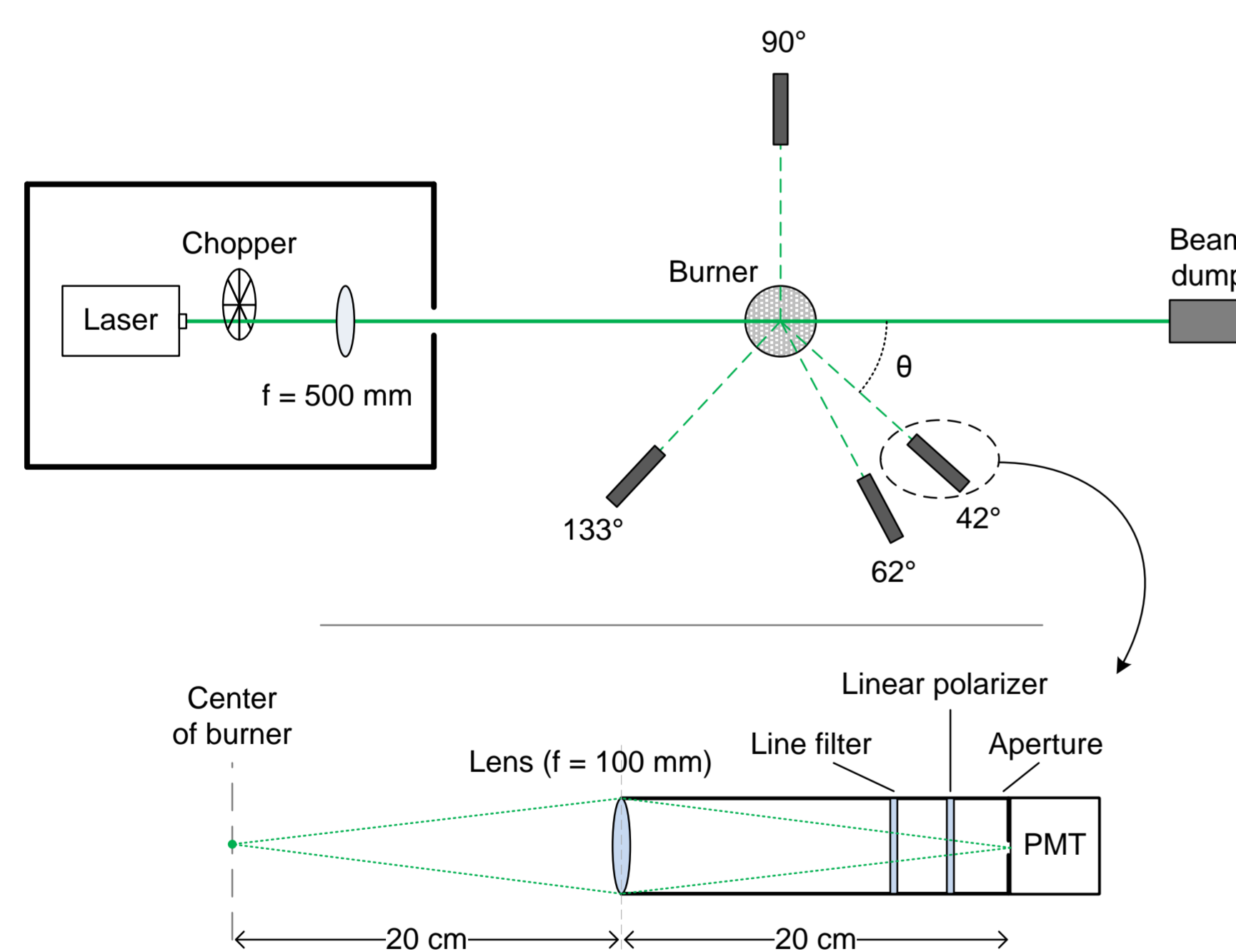
For small aggregates the angle dependence of the scattered signal I can be approximated by:

$$\frac{I(0)}{I(q)} \approx 1 + \frac{1}{3}q^2 R_g^2, \qquad q = 2k \sin\left(\frac{\theta}{2}\right)$$

where k is the magnitude of the wave vector, θ is the scattering angle, and R_g is the mass-averaged root-mean-square radius (radius of gyration) of an aggregate.

Results

- Measured R_g in the range of 10 to 120 nm.
- Sublinear dependence of R_g on residence time.

Admixture concentration dependence of growth
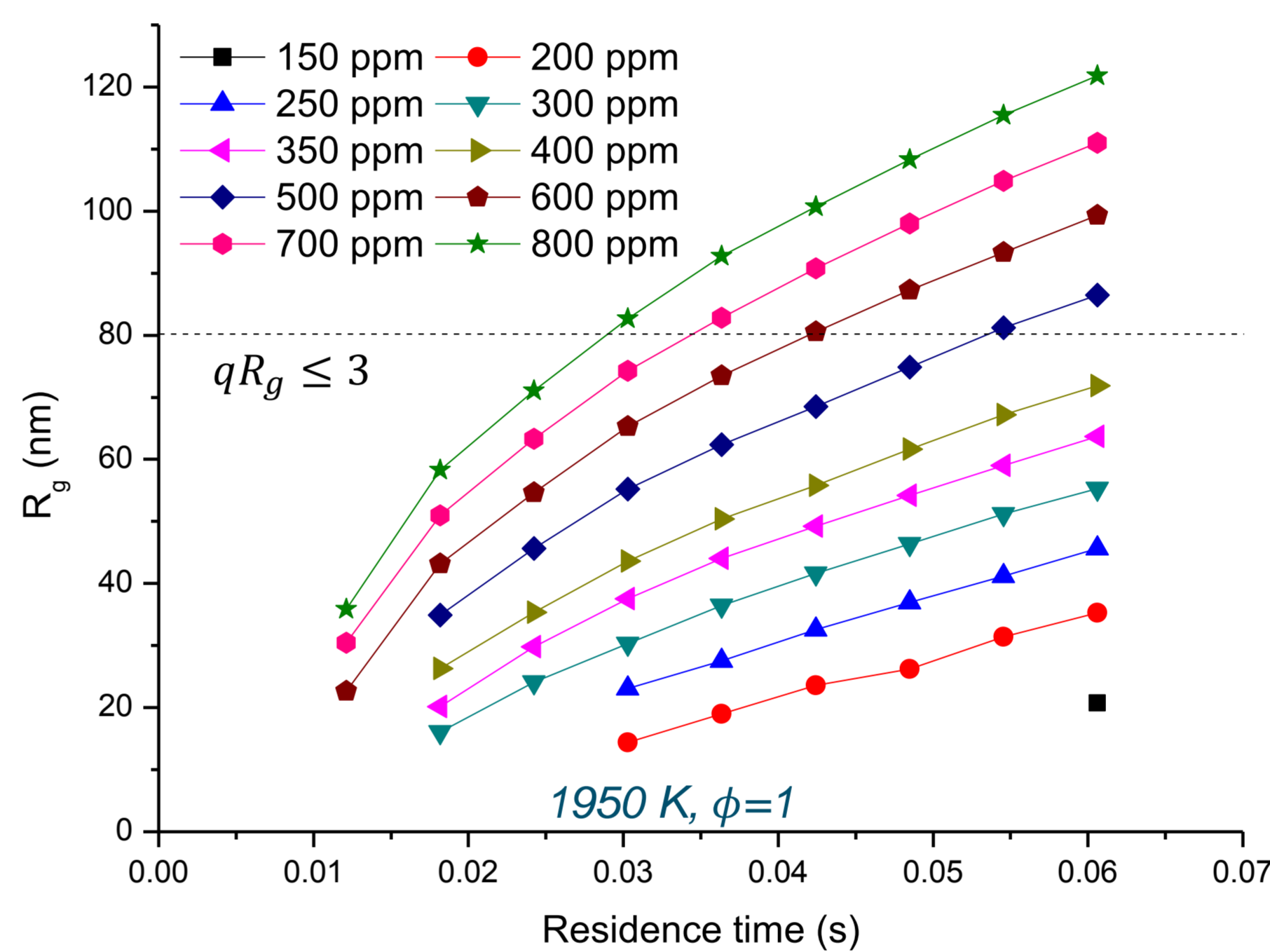
Sublinear dependence of R_g on doping concentration.

Temperature dependence of growth
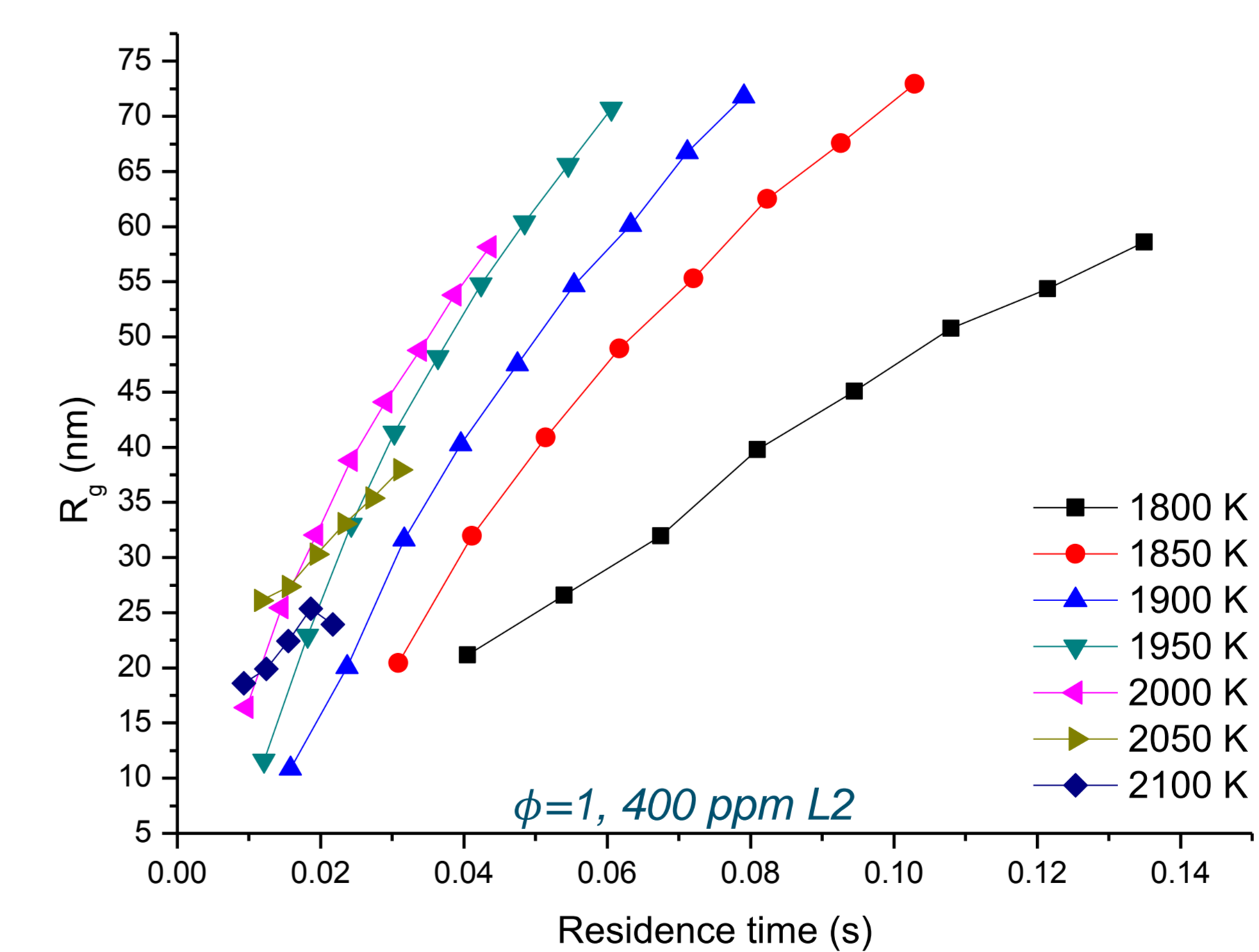
Non-monotonic dependence of R_g on flame temperature: R_g increases with flame temperature up to around 2000 K, but decreases thereafter.

Fuel equivalence ratio dependence of growth
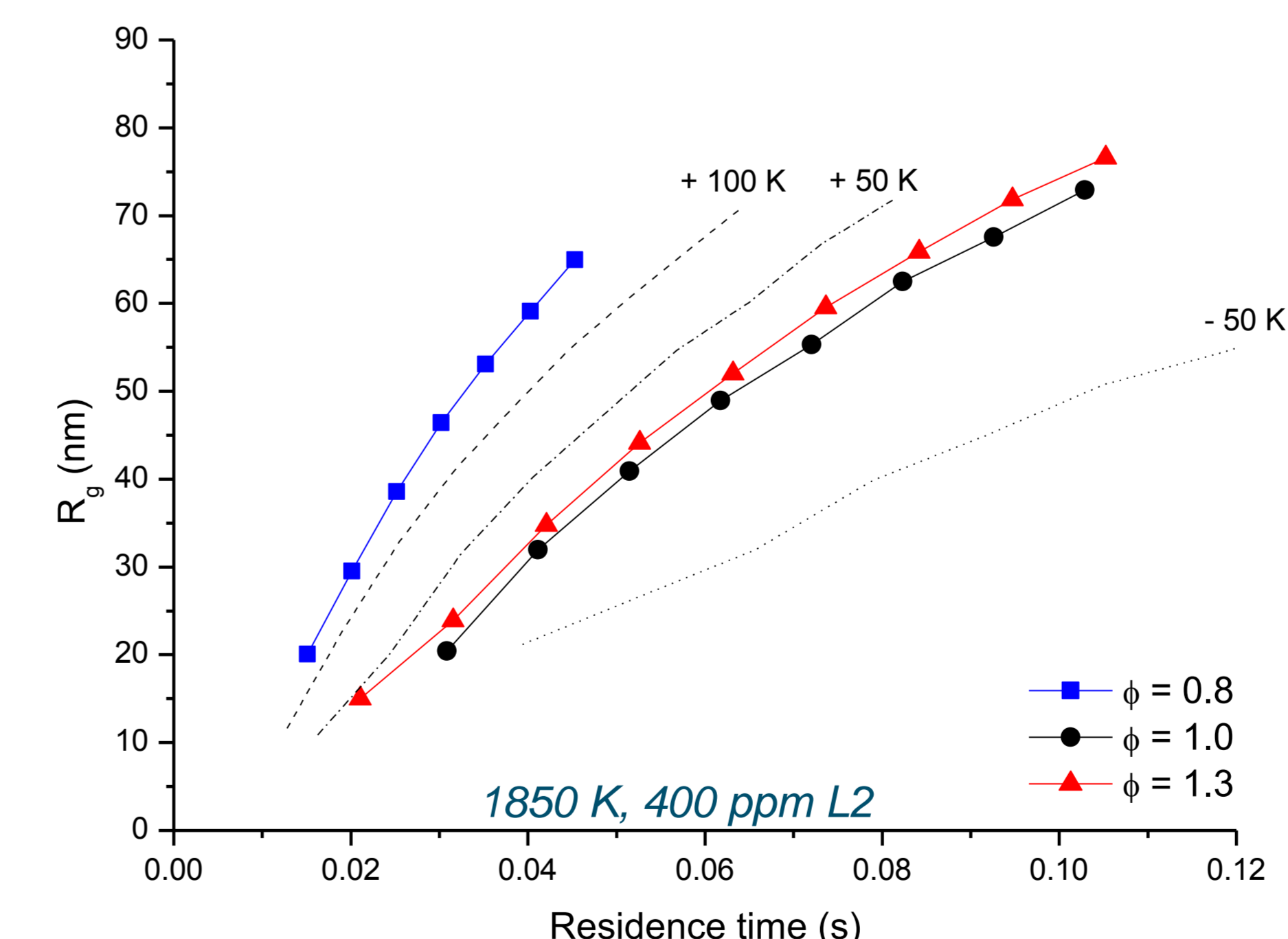
A lean flame environment (ϕ =0.8) appears to foster aggregate growth compared to rich (ϕ =1.3) and stoichiometric flames in which growth is very similar.

Discussion

- When fixing the initial conditions at the residence time corresponding to the first measurement point, a simple model describing particle evolution as a result of collisional growth and sintering predicts the functional dependence of the growth of particle radii well.
- The relatively limited growth at the highest used flame temperatures is likely because the melting point of SiO_2 (about 2000 K) is exceeded, which quite possibly affects the primary particle size and fractal dimension.