Optical Properties of Black Carbon Aggregates: Parallel Experimental and Numerical Study

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EMPIR BLACK CARBON

Introduction

- Light absorbing carbon (LAC), also called black & brown carbon, is pointed out to be the second most contributor to global warming after CO_2.
- Measurements of optical properties of LAC are required to analyze the radiative impacts of this aerosol species. Primary particles up to 30nm in diameter are produced in flames and aggregate to form fractal structures.
- These aggregates can be purely black carbon and black carbon coated with organic (brown) carbon depending upon the burning conditions.
- Simultaneous numerical and experimental studies are important to improve our understanding of optical and microphysical properties of BC aggregates, which is the key aspect of this study.
- We have carried out experimental and numerical studies (based on scattering theory) in parallel to calculate and examine the optical properties of pure BC aggregates

Mini - Cast Experiment

- For laboratory study, a diffusion flame-based BC generator (mini-CAST 5203C, Jing Ltd, Switzerland) is used at different operating points (Table 1) to generate BC aggregates.

Table 1. Operating conditions of the mini-CAST burner

Condition	Fuel (C_3H_8) flow rate (L min $^{-1}$)	Air flow rate (L min $^{-1}$)	Nitrogen (N_2) flow rate (L min $^{-1}$)	Flame Equivalence Ratio(ϕ)	D_p(nm)
OP100a	0.185	2.950	0.000	1.50	152.06
OP100b	0.180	3.400	0.180	1.26	153.06
OP100c	0.180	3.700	0.260	1.16	166.04
OP100d	0.140	3.300	0.330	1.01	157.72
OP100e	0.084	2.720	0.000	0.74	158.42

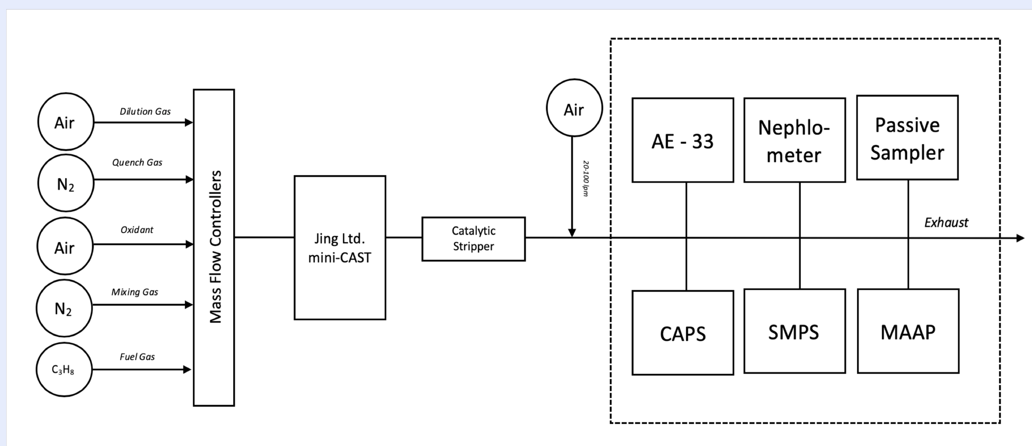


Figure 1. Descriptive sketch of the experimental setup

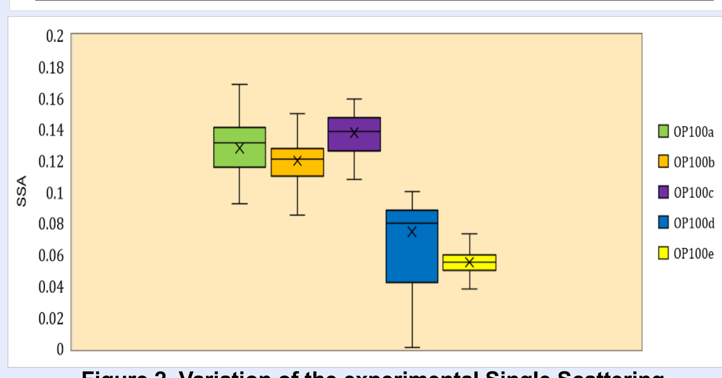
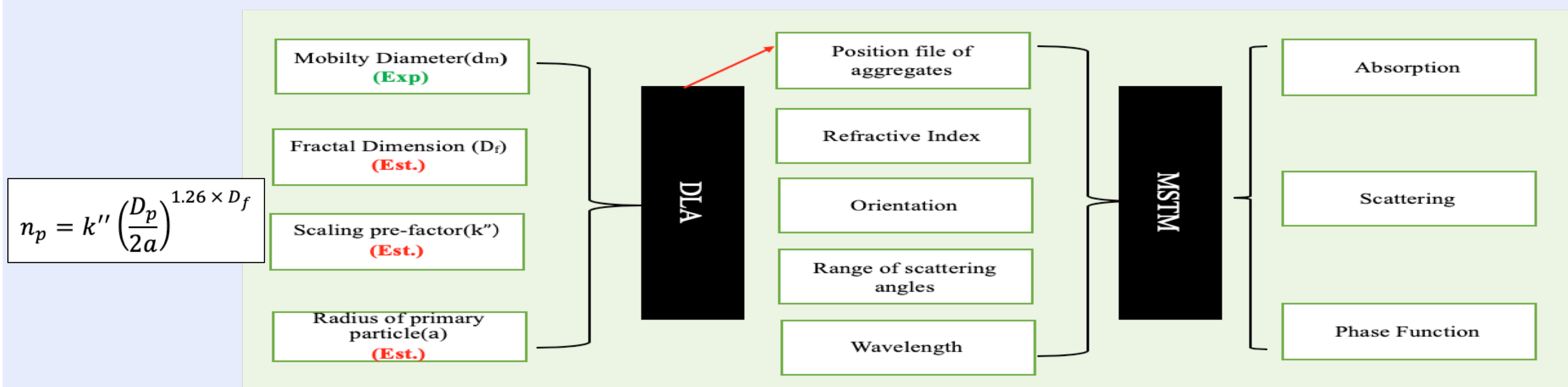


Figure 2. Variation of the experimental Single Scattering Albedo(SSA) for operating conditions mentioned in Table 1

Methodology for Numerical Computations



- DLA [1] is a tunable Diffusion Limited Aggregation software used to simulate BC aggregates .
- "Exp" are the inputs parameters taken from the results of mini-CAST experiments and "Est" are the estimated parameters.
- Fractal Dimension (*Df*) is estimated between *1.6-2.5*, depending upon the burning conditions (ϕ). Radii of primary particles (a) are varied from *10-30nm* [2] *and k"=1.81.*
- Multi Sphere T Matrix Code (MSTM) [3] applies the multiple sphere T matrix method for calculating light scattering properties of a group of spheres.

Single Sphere vs Aggregate Approach

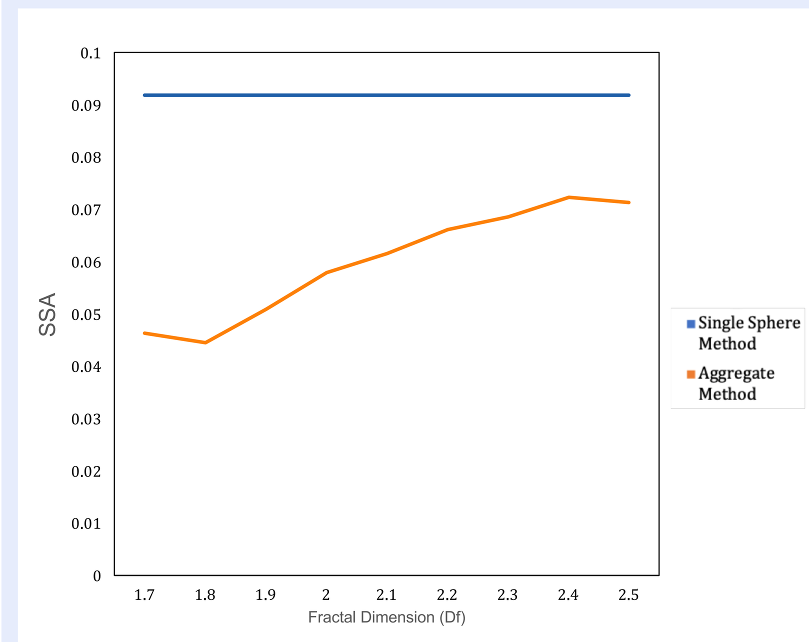


Figure 3. Discrepancy between SSA values using volume equivalent single sphere approach vs aggregate morphology approach

- For OP100a (D_p=152nm), the optical properties were modelled with two different morphological assumptions: single sphere and aggregate.
- For single the sphere approach (n_p=1, a=55nm, RI=1.7+0.7i), the SSA is equal to *0.09.* In this case, D_f is a constant equal to 3.
- For aggregate approach (*n_p=50, a=15nm, RI=1.7+0.7i) and* varying fractal dimensions (D_f) from *1.7-2.5*, we obtain SSA in the range *0.04-0.07.*
- The sphere approximation is mostly used in large scale modelling. It's not able to consider the factor of fractal dimension, which causes a discrepancy (*figure 3*) that decreases when D_f*~3* (more compact aggregate*).*

Result 1 : Dependence of Optical Properties on Morphology

For D_f *= 2.3-2.5,* an increasing trend for MAC is seen as primary particle size (a) increases.

When *a>25nm*, the MAC shows a steep increase due more compact structure of the aggregates at this point

There is a general increase in MSC as primary particle size (a) increases.

There is a increase in SSA as the *Df* increases, also seen in experimental results (*figure 2*)

For asymmetry parameter, a dependence on primary particle size is seen.

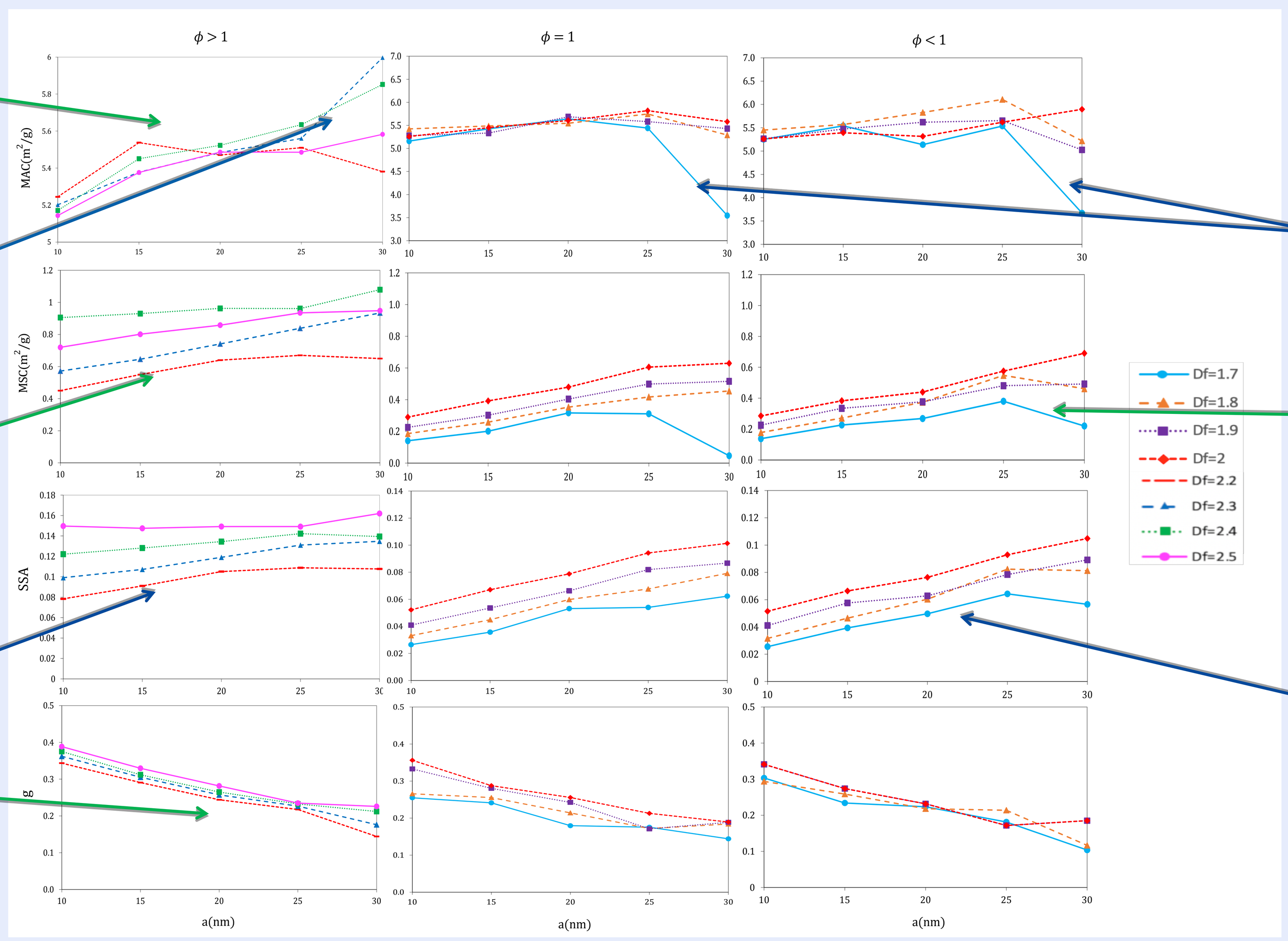


Figure 4. Modelled mass absorption coefficients (MAC), mass scattering coefficients (MSC), single scattering albedos (SSA) and asymmetry parameters (g) of BC aggregates for three experimental cases (flame equivalence ratios(ϕ>1, ϕ=1 and ϕ<1)) are plotted. In all cases, the dependence of each optical parameter with primary particle size (a) and fractal dimension (Df) is studied. The refractive index is taken as *1.7+0.7*i [4]. For MAC and MSC calculation, the density of BC is assumed as *1.8g/m³.*

For *Df<2.3, when a=20nm* the MAC decreases.

This may be because in this case, for the same equivalent mobility diameter there is less number of primary particles of larger radius. With such less numbers of primary particle*(19-8*), the influence of morphology (lacy chain like structure) is more pronounced.

When *Df<1.8*, at *a<25nm* there is decrease in MSC due to very less number of primary particles

It is observed that SSA is almost proportional to the increase in primary particle size.

Result 2 : Dependence of Optical Properties on Complex Refractive Index

Table 2. Operating points chosen for the study of refractive index

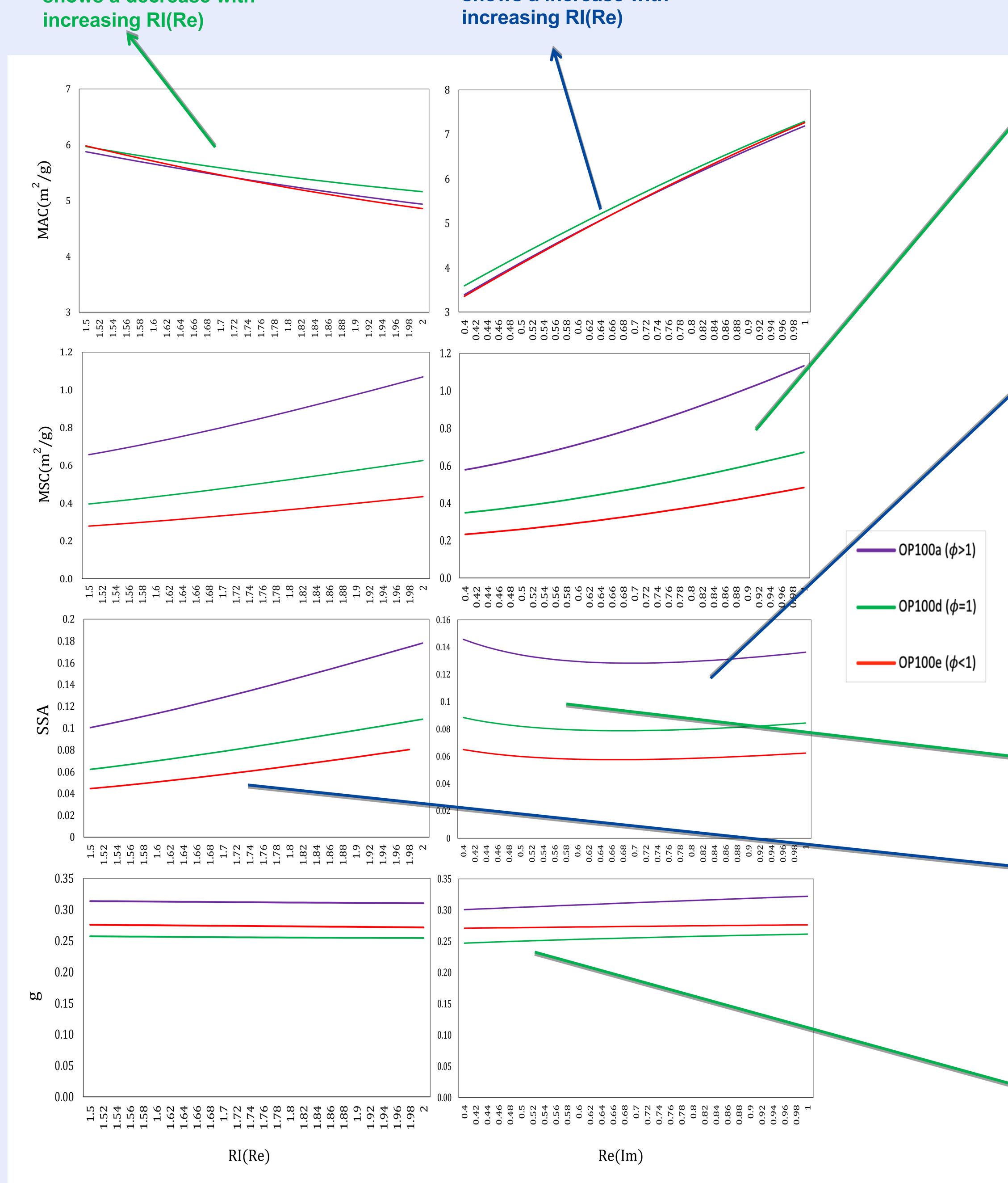
Operating Point	D_f	a(nm)	n_p	Flame Equivalence Ratio(ϕ)	D_p(nm)
OP100a	2.4	15	135	1.5	152
OP100d	2.0	20	32	1	157
Op100e	1.9	15	54	0.74	158

Table 3. Uncertainty values for modelled optical properties

Operating Point	MAC	MSC	SSA	g
OP100a	±0.091	±0.018	±0.004	±0.009
OP100d	±0.096	±0.015	±0.003	±0.008
Op100e	±0.092	±0.0112	±0.001	±0.007

For *RI(Re)=1.5-2*, the *MAC is between 4.8 and 6*, and shows a decrease with increasing RI(Re)

For *RI(Im)=0.4-1*, the *MAC is between 3.3 and 7.3,* and shows a increase with increasing RI(Re)



For MSC, both cases(*Re and Im*) show increasing trend as the value of RI is increased. Values of *MSC is between 0.2 and 1.13*

For *RI(Im)=0.4-1*, the *SSA is between 0.6 and 0.15*

The SSA increases with increase in Ø , this is because the aggregate gets more compact in shape.

For *RI(Re)=1.5-2*, the *SSA is between 0.045 and 0.17*, and shows a increase with increasing RI(Re)

The asymmetry parameter lies between *0.24 and 0.322* showing less dependence on RI.

Figure 5. Modelled mass absorption coefficients (MAC), mass scattering coefficients (MSC), single scattering albedos (SSA) and asymmetry parameters (g) of BC aggregates for three experimental cases(ϕ>1, ϕ=1 and ϕ<1) are plotted. In all cases, the dependence of each optical parameter with real and imaginary part of refractive index is studied.

Result 3 : Illustrative Study of the Morphology of BC Aggregates

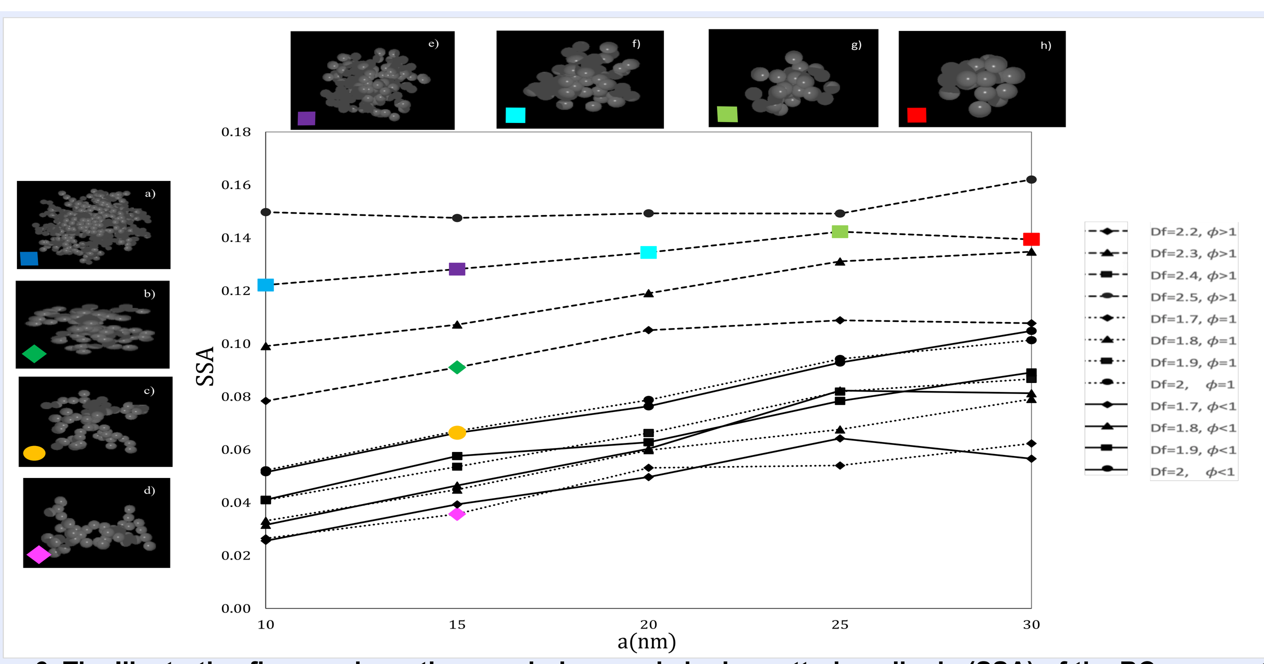


Figure 6. The Illustrative figures show the morphology and single scattering albedo (SSA) of the BC aggregates in different cases of fractal dimension (D_f) and flame equivalence ratios (ϕ): a)*Df=1.7, a=15nm; b) Df=1.9, a=15nm; c) Df=2.2, a=15nm; d)Df=2.4, a=10nm; e) Df=2.4, a=15nm; f) Df=2.4, a=20nm; g) Df=2.4, a=25nm; h) Df=2.4, a=30nm*

Summary/Outlook

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- Optical properties of BC aggregates produced by mini-CAST 5203C with mobility diameter (D_p) between *152nm* to *166nm* were measured and later simulated using Multi Sphere T Matrix Code (MSTM).
- The dependence of optical properties on morphology (*figure 4 & 6*) and complex refractive index (*figure 5*) were studied.
- The experimentally measured optical properties can be retraced by modelling to a narrow range of points depending upon primary particle size(*a*) and fractal dimension (D_f).

Outlook

- Conduct laboratory studies to further narrow down the range of fractal dimension (D_f) and primary particle size (*a*).
- Study the BC aggregates with thin organic coatings.

Acknowledgement

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