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Detailed Assessment of Particulate Characteristics from Low-Temperature Combustion (LTC) Engines

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UChicago Argonne LLC

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Overview

Timeline

- Start: Oct 2010
- Finish: September 2011
- 95% completed

Budget

- Funding received from DOE in FY11
  - $300K
- Funding from DOE for FY12
  - $200K

Barriers

- Lack of fundamental knowledge about particulate emissions from advanced engine combustion.
- Lack of actual particulate emissions data for various fuels
- Lack of cost-effective particulate emissions control

Partners

- University of Wisconsin
- Sandia National Laboratory

Relevance and Objectives

- The particle sizes measured by commercial instruments have rarely been verified.
- Soot morphology needs to be evaluated to understand the detailed formation mechanisms of PM emissions in LTC modes.
- The effects of fuel composition are unavailable for PM emissions from LTC modes.

- Assess nano-particles from LTC by comparison of particle sizes measured by scanning mobility particle sizer (SMPS) and transmission electron microscope (TEM) .
- Examine LTC soot morphology and nanostructures to better understand the soot formation mechanism in LTC modes.
- Evaluate the effects of biofuels on particulate emissions from LTC modes.

Background

- It has recently been found that LTC engines produce a larger number of nano-particles pertaining to the nucleation mode, but a less amount of total mass, than does conventional diesels.
  - Emissions regulation for the number of nano-particles (Ø ≥ 23 nm) will be effective soon.
- LTC soot models have been proposed (SAE 2007-01-1945).

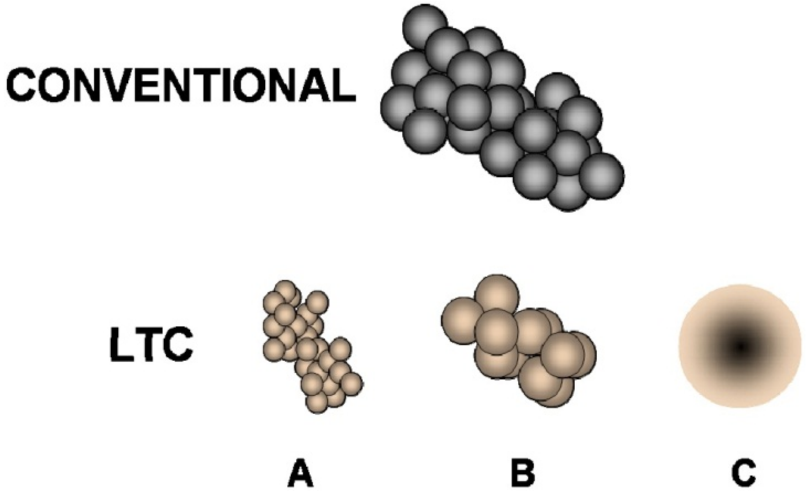
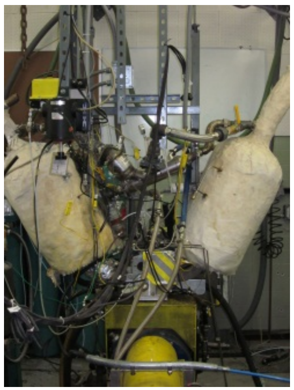
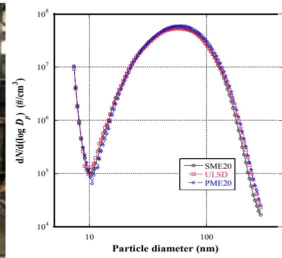


Fig. 1 Proposed LTC soot models:
A: surface growth limited
B: agglomeration limited
C: No agglomeration (coalescence only)

Approach



LTC engine



SMPS measurement
- Particle size distributions

Compare Data

Radius of gyration:

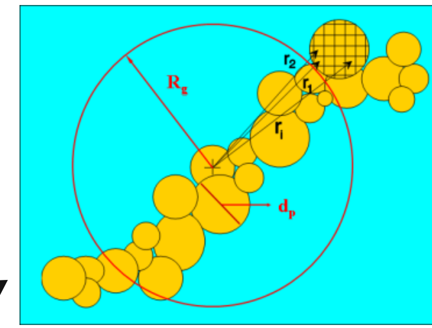
$$R_g = \sqrt{\frac{1}{n}\sum_{i=1}^{n} r_i^2}$$

Number of primary particle:

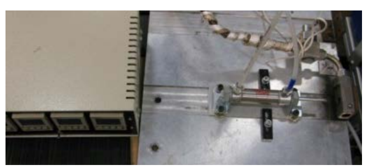
$$n = \left(\frac{A_a}{A_p}\right)^{1.09}$$

Fractal dimension:

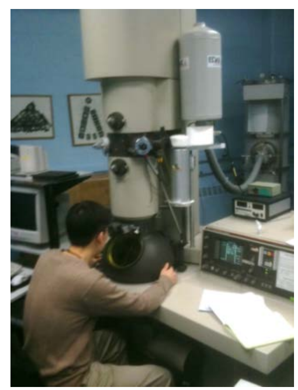
$$\ln(n) = D_f \cdot \ln\left(\frac{R_g}{d_p}\right) + \ln(k_f)$$



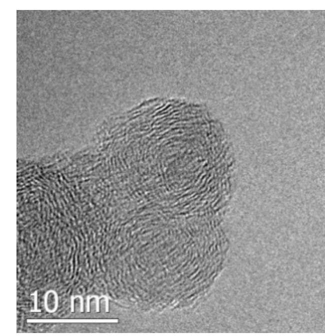
PM morphology



Thermophoretic sampling
- Minimum residence time: 20 ms



TEM analysis



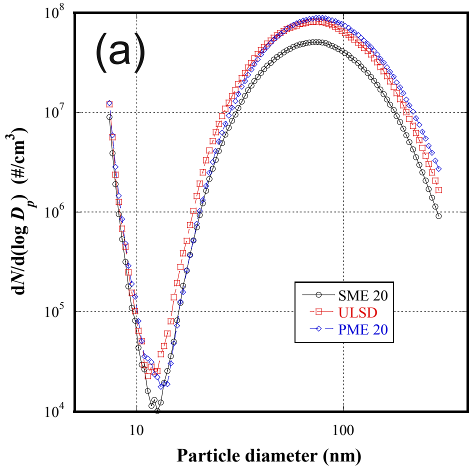
Soot nano-structures

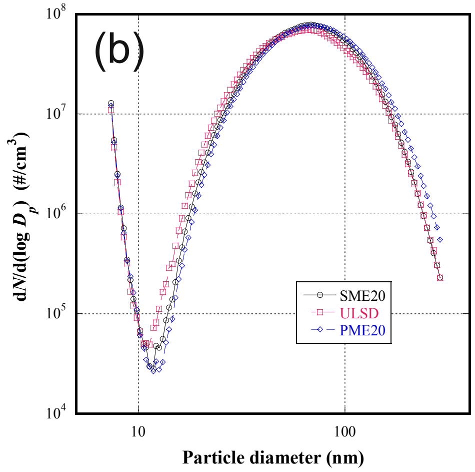
Experimental

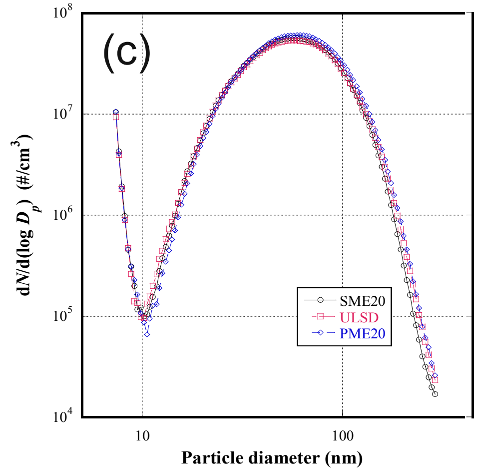
- Engine
  - A single cylinder version of a GM 1.9L 4-cylinder engine modified for low temperature combustion
- Fuel
  - Ultra low sulfur diesel (ULSD)
  - Soy methyl ester (20%)/ULSD mixture (SME20)
  - Palm -oil methyl ester (20%)/ ULSD mixture (PME20)
- Engine operating conditions
  - Speed and torque: 2000 rpm and 5.5 bar IMEP
  - Injection timing: 22, 26 and 30^{o} before top dead center (BTDC)
  - Fuel rail pressure: 860 bar
  - EGR rate: 67%
  - Intake oxygen concentration: 9.5%

Technical Accomplishments

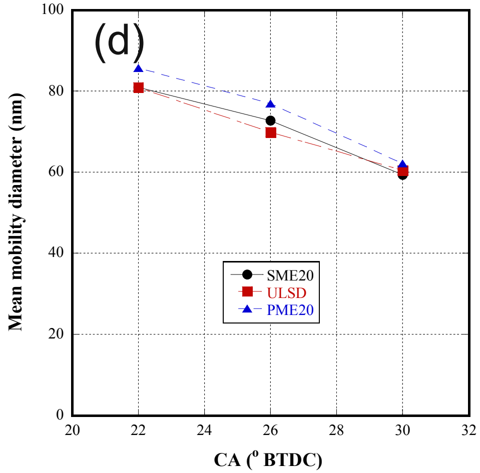
- **Particle size distributions measured by SMPS**





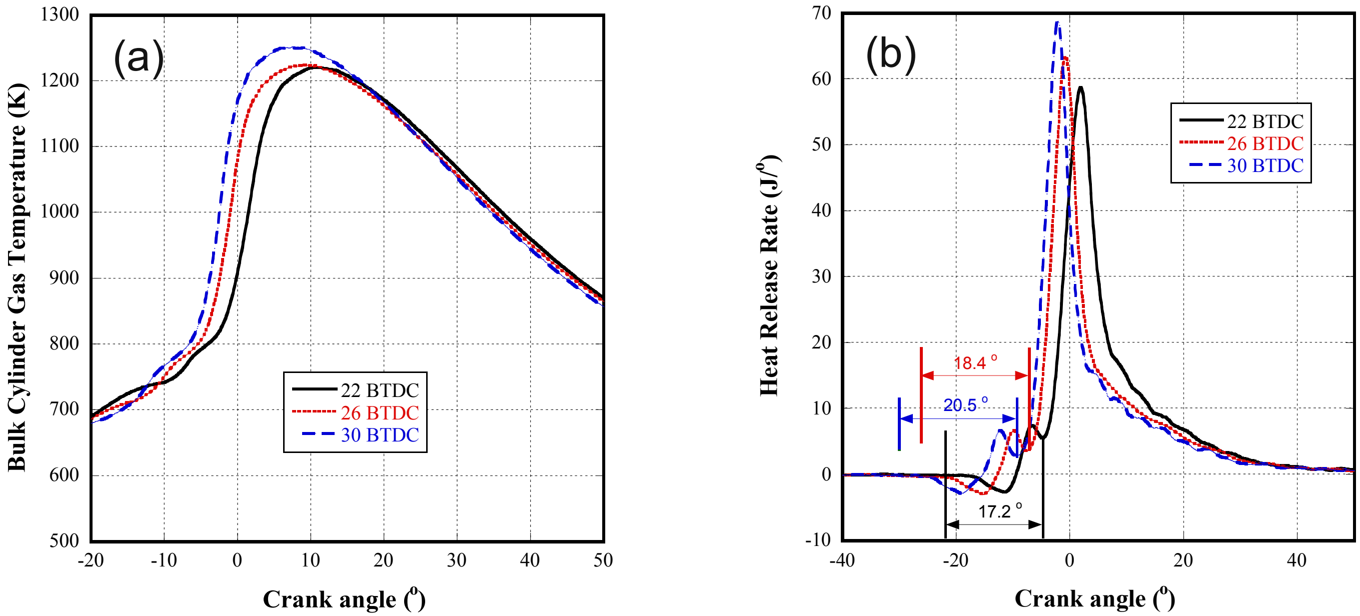


(a) 22°, (b) 26°, (c) 30° BTDC, and (d) mean mobility diameter



- A large population of nano-particles below 10 nm exists at all the conditions.
- With advancement in injection timing, differences in the size distribution are reduced among the different fuels.
- The mean mobility diameter decreases with advancement in injection timing, mainly due to the increased number of small particles in a range of approximately 10 to 20 nm.

Analyses of engine in-cylinder data

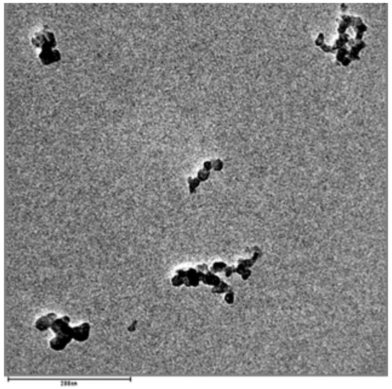


(a) Bulk in-cylinder gas temperatures and (b) calculated heat release rate at different injection timings (SME20)

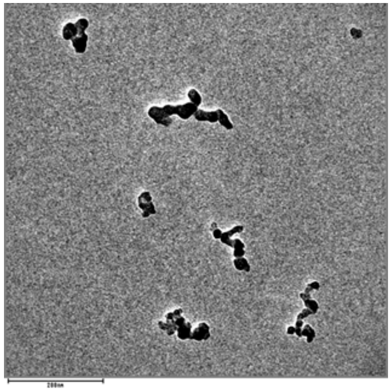
- In-cylinder gas temperatures gradually increased with advanced injection timing.
- Ignition delay appeared to increase with advanced injection timing.

Soot morphology by TEM analysis

(a)

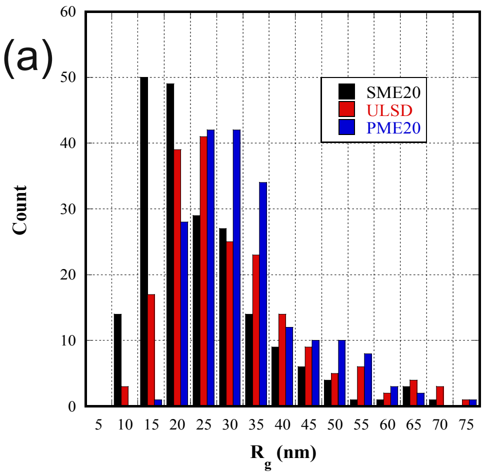


(b)

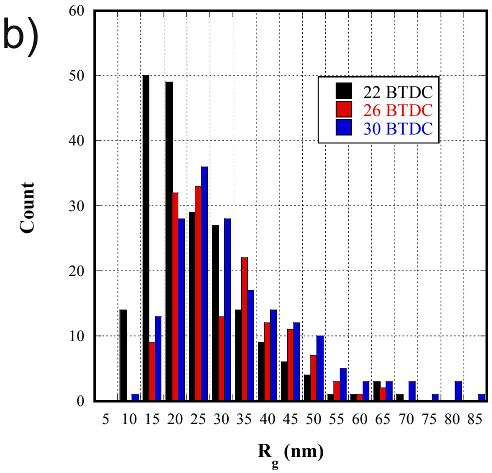


TEM images of soot aggregates for (a) SME20 and (b) ULSD

(a)



(b)



Size distributions of soot aggregates:
(a) radii of gyration for different fuels at 22 º BTDC,
(b) radii of gyration for SME20 at different injection timings,

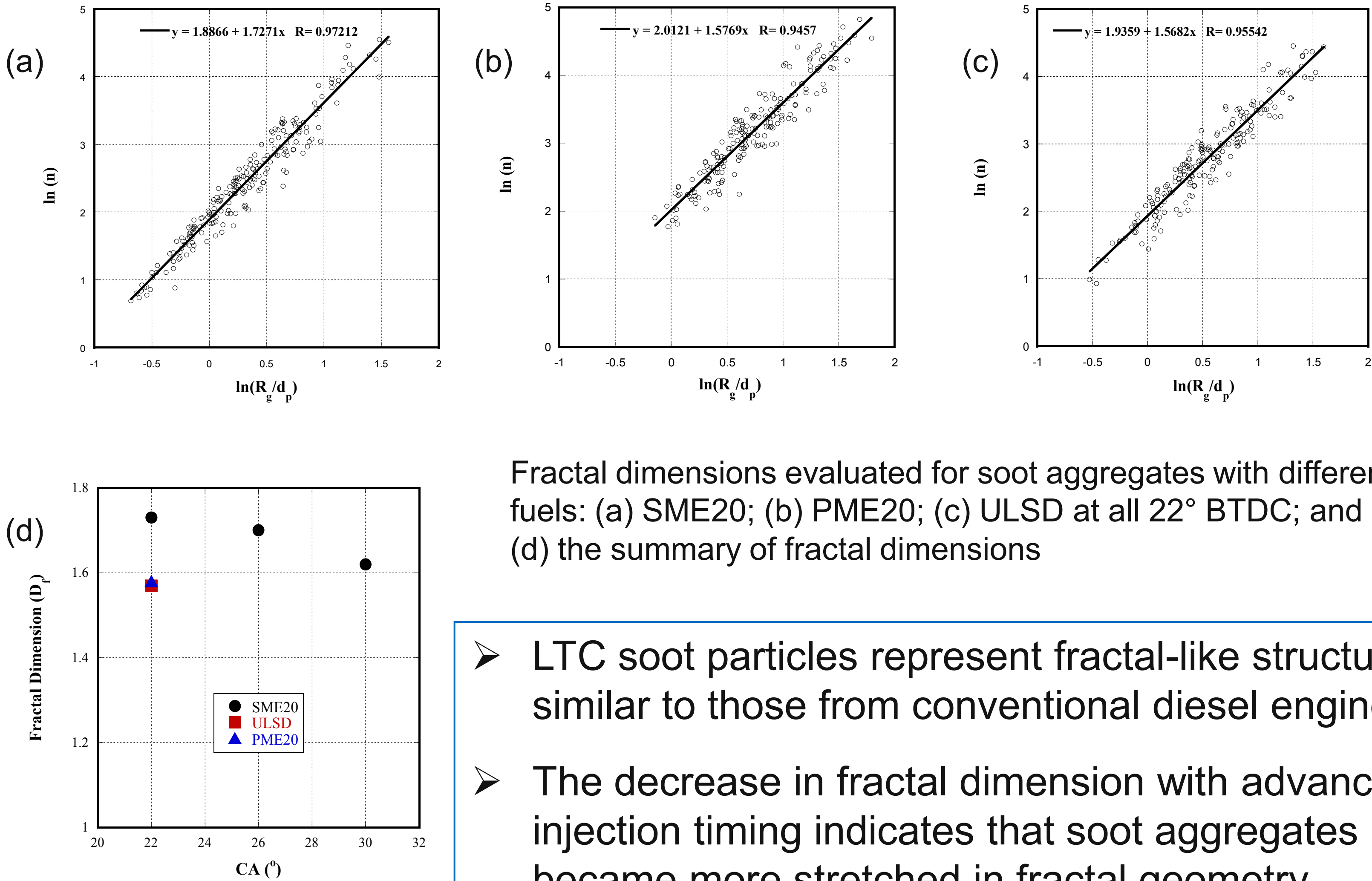
- ➢ The majority of soot particles from LTC represented chain-like structures.
- ➢ SME20 soot appeared to be smallest in aggregate size.
- ➢ The population of large particles increased with advancement in injection timing.

Summary of particle physical dimensions

Fuel	Injection timing (° BTDC)	Primary particle (D_p), nm	Radius of gyration (R_g), nm	Projected area equivalent diameter (D_{proj}), nm
SME20	22 26 30	14.4 ± 7.7 13.2 ± 3.7 16.9 ± 5.2	22.1 ± 11.4 28.2 ± 11.1 31.0 ± 15.1	47.2 ± 19.5 55.2 ± 17.5 62.4 ± 23.5
PME20	22	10.9 ± 4.0	25.0 ± 10.8	49.4 ± 16.0
ULSD	22	14.8 ± 3.5	28.0 ± 13.1	56.1 ± 19.3

- LTC soot particles are smaller in D_p and R_g than those from conventional engines, which are in the ranges of 20 – 50 nm and 40 – 80 nm, respectively.
- Average aggregate size (SME20) increases with advancement in injection timing, in contrast to SMPS data
  → Contribution of volatiles for SMPS data
- Both primary and aggregate particles appeared to grow in size with advancement in injection timing
  → Effects of increased temperatures and longer residence time

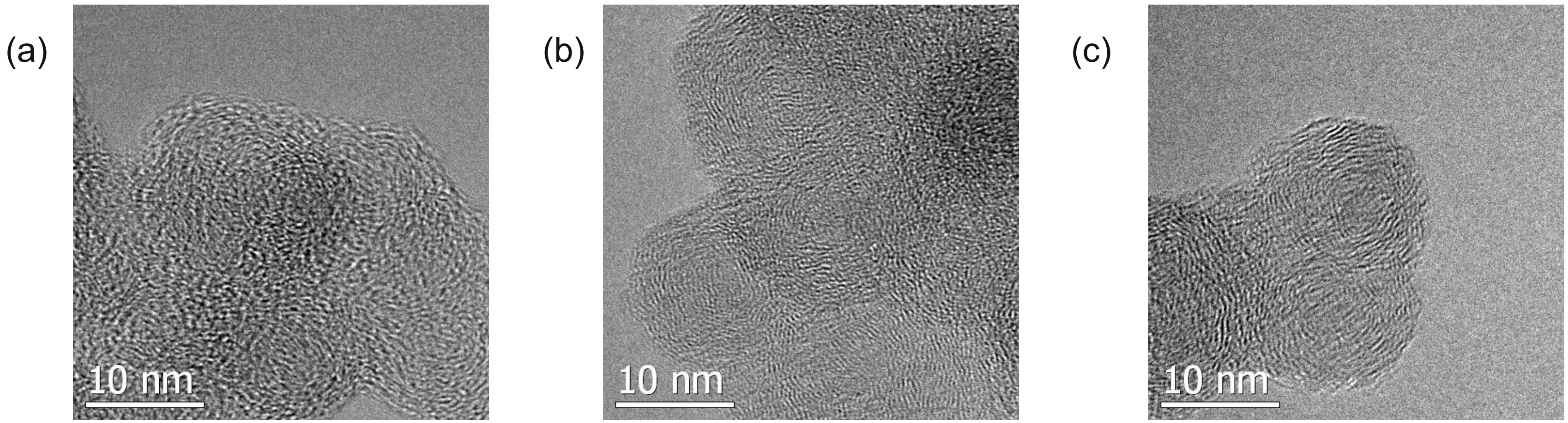
Fractal dimensions



Fractal dimensions evaluated for soot aggregates with different fuels: (a) SME20; (b) PME20; (c) ULSD at all 22° BTDC; and (d) the summary of fractal dimensions

- LTC soot particles represent fractal-like structures, similar to those from conventional diesel engines.
- The decrease in fractal dimension with advanced injection timing indicates that soot aggregates became more stretched in fractal geometry.

Nano-structures of primary particles



HRTEM images of soot particles: (a) SME20 at 22º BTDC, (b) SME20 at 30º BTDC, and (c) ULSD at 22º BTDC

- Both SME20 and ULSD soot samples showed a higher degree of disorder in nanostructures.
  - Short-range graphene segments within primary particles
  - Rough outer surfaces of primary particles
- We speculate that soot oxidation was insignificant with advancement in injection timing, due to the LTC conditions.
  - Insignificant difference in the interlayer spacing: 0.364 nm (22º BTDC), 0.361 nm (30º BTDC)
  - Increasing trends in both primary and aggregate sizes

Future Work

- LTC soot properties will be further examined.
  - Evaluation of soot oxidative reactivity (kinetic parameters) using a TGA
  - Examination of soot nano-structures (soot crystalline structures) using Raman spectroscopy

- Detailed assessment of particulate emissions will be made for spark ignited direct injection (SIDI) engines.
  - Engines: Hyundai 2.0L SIDI engine (ANL) and GM 1.9L SIDI engine (Univ. of Wisconsin)
  - Scope of work: examination of soot properties from SIDI engines
  - Deliverables: particle size distributions, PM morphology, particulate nano-structures, and crystalline structures, and kinetic parameters of particulates (e.g., activation energy, reaction order).
  - Measurement instruments: SMPS, TEM, Raman spectroscope, and TGA

Summary

- SMPS data showed a large population of nucleation mode particles in diameters below 10 nm .
- Based on the TEM examination where aggregate particles smaller than 10 nm were rarely observed, the nucleation particles measured by the SMPS are to be aerosols from volatile organics.
- The LTC soot particles are found to be smaller in both primary and aggregate particle sizes than those from conventional diesel engines.
  - LTC soot particles represent chain-like fractal geometry, similar to those from conventional diesel engines.
  - Results from the present study support the proposed model (a).
- Examinations of nano-structures as well as morphology propose that soot oxidation was insignificant during the combustion process. Therefore, the degree of graphitic structures should be relatively low.
- Biodiesel soot appears to be smaller in both primary and aggregate sizes than does ULSD soot, which results in the reduced total soot mass.