Characterization of Silicon Dioxide Food Additives by Single Particle Inductively Coupled Plasma Mass Spectrometry

Monique E. Johnson,[1] Sadia Afrin Khan,[2] Timothy R. Croley,[2] Antonio R. Montoro Bustos,[1] Ingo H. Strenge,[1] and Karen Murphy[1]

[1] Chemical Sciences Division, Material Measurement Laboratory, ***National Institute of Standards and Technology***, *Gaithersburg, MD 20899*

[2] Center for Food Safety and Applied Nutrition (CSFAN), ***U.S. Food and Drug Administration***, College Park, MD 20740

Introduction

- Silicon dioxide (SiO_2), in its amorphous form, is an approved food additive (21 CFR 172.480) in the U.S., mainly used as an anticaking agent in powdered food products, a chill proofing agent in malt beverages, and a filter aid in wines, beer, and juices. During production, particles of SiO_2 aggregate to form nano-sized and larger particles, however, there is currently no data concerning the particle size distributions of food grade SiO_2. This particle size metric is important in evaluating the potential transformations of nanoparticles (NPs) in food products.
- In 2006, the U.S. Food and Drug Administration (FDA) initiated a Nanotechnology Task Force to address knowledge gaps associated with the use of nanoscale materials in FDA-regulated products. In this effort, FDA's Center for Food Safety and Applied Nutrition (CFSAN) has begun collaborative work with the Inorganic Measurement Science Group in National Institute of Standards and Technology (NIST) to develop reliable analytical strategies for the characterization of metal oxide NPs in food derivatives. Six commercially available food grade SiO_2 will be characterized by a suite of analytical techniques.

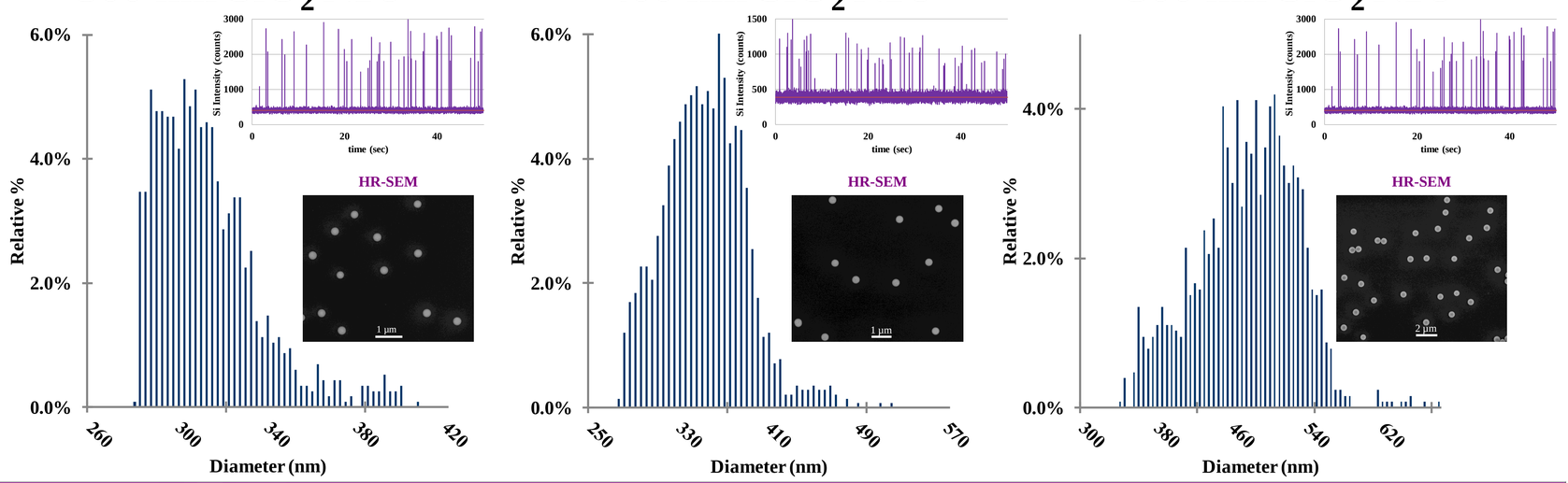
Research Goals

- Silicon content will be determined in test materials by inductively coupled plasma mass spectrometry (ICP-MS) following acid-assisted digestion.
- Particle size distribution information will be obtained using dynamic light scattering, scanning electron microscopy, and single particle (sp)ICP-MS analysis.
- Commercially available spherical SiO_2 NPs, ranging from 300 nm to 500 nm in size, will be used as control materials to optimize the challenging spICP-MS analysis.

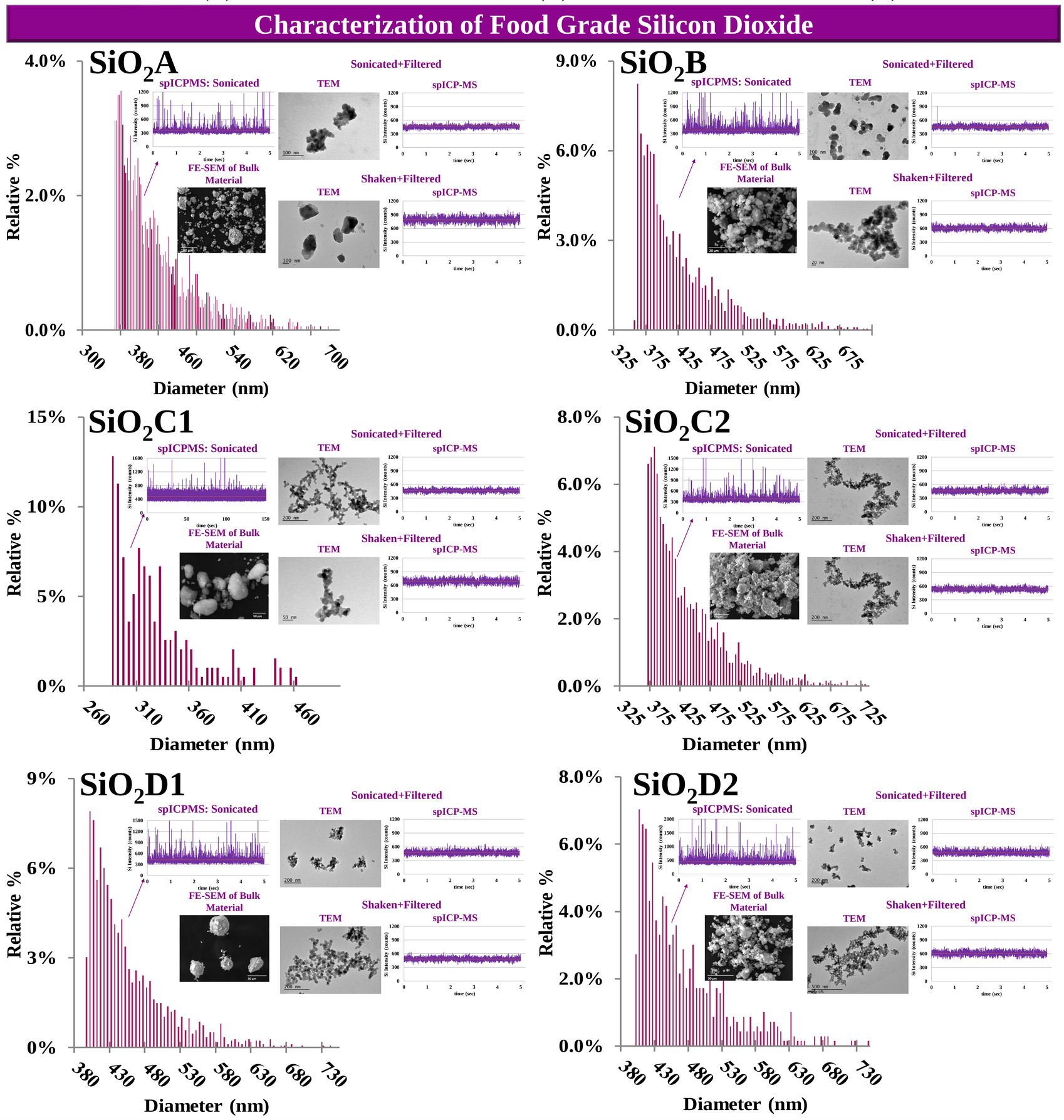
Sample Preparation and Analysis

- ✓Six food grade SiO_2 samples were procured from four commercial distributors/manufacturers. These samples were given unique names to ensure confidentiality.
- ✓0.1 % w/v dispersions of each food grade SiO_2 material were prepared in ultrapure water (18 MΩ) by three methods:
  - ✓Sonication: 10 min, 55 % pulse amplification, pulse mode: 20 s on and 20 s off
  - ✓Shaken + filtration: 15 min at 250 RPM, followed by filtration through a 0.22 µm filter.
  - ✓Sonication + filtration: sonication procedure mentioned above followed by filtration through a 0.22 µm polyethersulfone filter membrane.
- ✓Characterization of the bulk food additive material and dispersions were carried out using a JEOL JEM-1400 Transmission Electron Microscope (TEM) and TESCAN Mira3 Field Effect-Scanning Electron Microscope (FE-SEM).
- ✓Particle size distributions were measured *via* Malvern Zetasizer Nano ZEN3600.
- ✓spICP-MS measurements were carried using a PerkinElmer NexION 350D ICP-MS
- ✓Key instrument parameters: signal at *m/z* 28, 0.10 mL min^{-1} flow rate, 1 ms dwell time, 600 s measurement time

Characterization of Commercial Silicon Dioxide Nanoparticles



Characterization of Food Grade Silicon Dioxide



Sample*	High Resolution SEM (nm)	Average Diameter *via* spICP-MS (nm)	# of Particles Analyzed *via* spICP-MS (nm)	Particle Number Concentration Recovery (%)
300 nm SiO_2*	296 ± 16	312 ± 24	1,154	102 %
400 nm SiO_2*	367 ± 11	350 ± 33	1,413	97 %
500 nm SiO_2*	564 ± 22	473 ± 53	1,263	100 %

Sample	Z-Average (nm) *via* DLS	Average Diameter *via* spICP-MS (nm)	# of Particles Analyzed *via* spICP-MS (nm)
SiO_2-A sonication	1362	419 ± 66	1,799
SiO_2-B sonication	495	421 ± 65	2,209
SiO_2-C1 sonication	156	323 ± 41	195
SiO_2-C2 sonication	469	431 ± 63	2,011
SiO_2-D1 sonication	277	451 ± 55	1,746
SiO_2-D2 sonication	440	468 ± 65	697

Conclusions

- ✓Transmission- and scanning electron microscopy confirmed that the food grade silicon dioxide samples measured in this study contained nanosized particles when the materials were prepared by a combination of sonication+filtration or shaken+filtration.
- ✓spICP-MS results show that probe sonication of the six food additive materials produced nanosuspensions containing particles in a size range from 300 nm to 700 nm. These sizes differ significantly from the size of particles in the original material, demonstrating substantial transformations that silicon dioxide undergoes when they are used as food additives.
- ✓spICP-MS preliminary results also indicate that the filtration step caused a significant retention of nanoparticles larger than the 0.22 µm filter cutoff. Future work will seek to expand the linear range of spICP-MS for SiO_2 NP measurements in an effort to quantify and size NPs in the nanorange.