Physical Characterization of Solid-Liquid Slurries at High Weight Fractions Using Optical And Ultrasonic Methods (# 81964)

University of Washington Team: Lloyd Burgess, Anatol Brodsky, Summer Randall (Graduate Student)

PNNL Team: Paul Panetta, Dick Pappas, Salahuddin Ahmed, Brian Tucker, Judith Bamberger, and Leonard Bond

Outline

- Objectives and benefits
- Ultrasonic measurements and theory
- Optical measurements and theory
- Conclusions
- Path forward

Objectives and Benefits

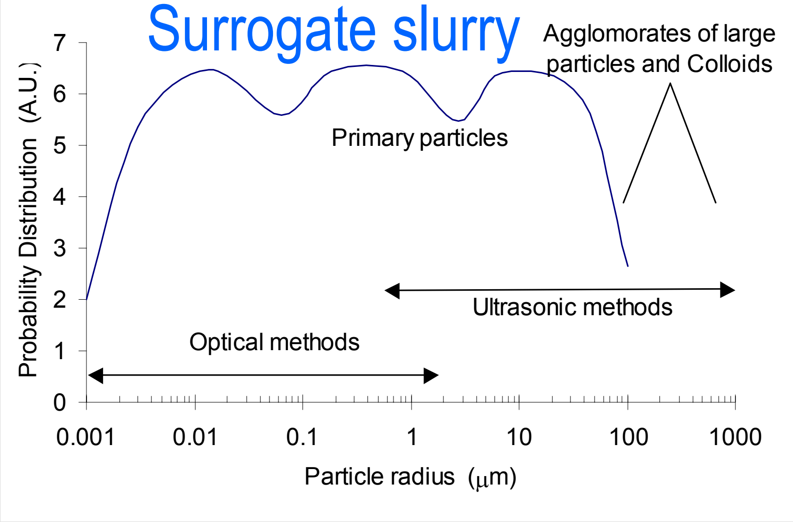
Objective

- Physically characterize HLW properties at high concentration over a wide range of particle sizes
  - Particle size distribution (nm to mm)
  - Concentration
  - Agglomeration
  - Gelation

Benefits

- Accurately characterize HLW at high concentrations
  - Avoids dilution
  - Saves cost due to lower volumes of waste to process
  - More accurate process control
  - Avoids pipeline clogging
  - Accelerates clean up
  - Beginning discussions with SRS

Optical and Ultrasonic Plan



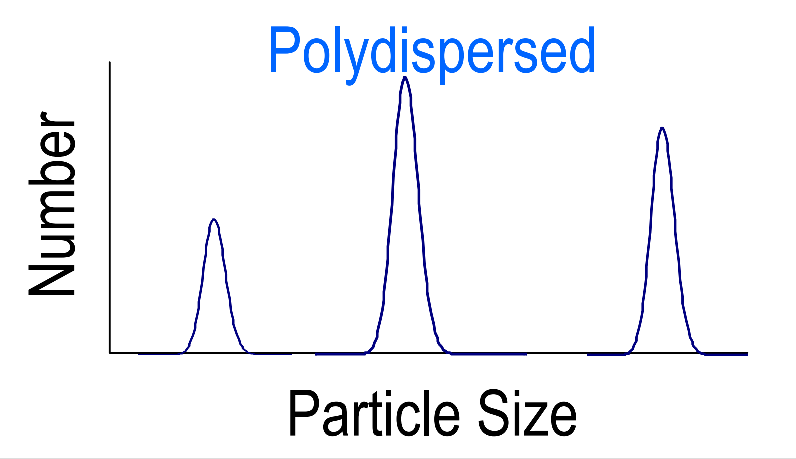
<u>Monodispersed and Polydispersed Particles</u>

- Concentration
- Size (nm to mm)
- Type of Material (Scattering strength)

Theoretical developments

Combined measurements

Design combined optical and ultrasonic system



Objective:

Online physical characterization of HLW to accelerate clean up

Samples measured

Methodology development

- Silica
- Polystyrene
- Well characterized slurry – attrition process

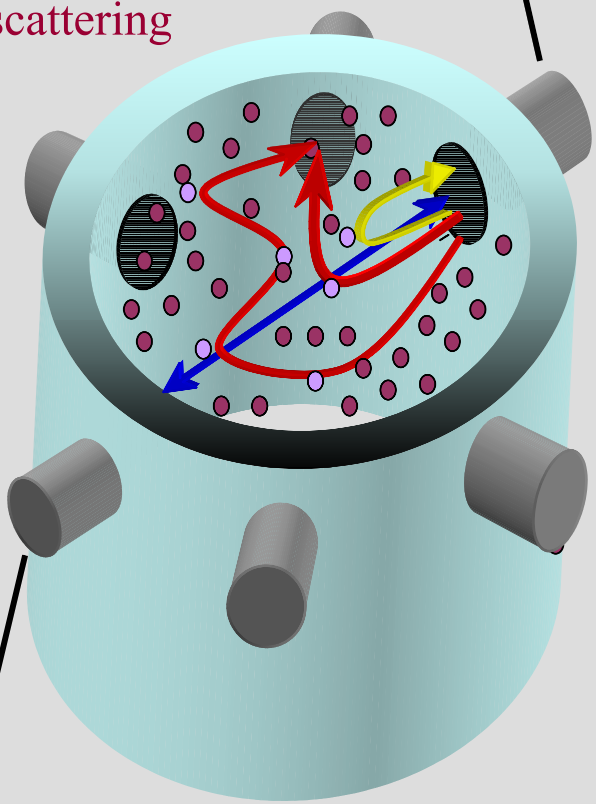
“Real” Specimens

- AZ 101/102 Tank simulant

Ultrasonic measurements

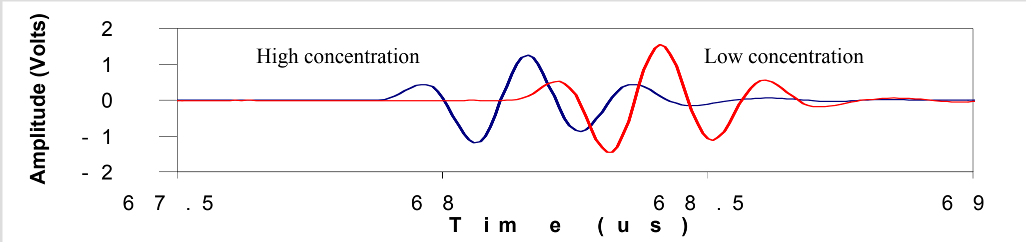
Receiver for diffuse field and off axis backscattering

Transmitter/ Receiver

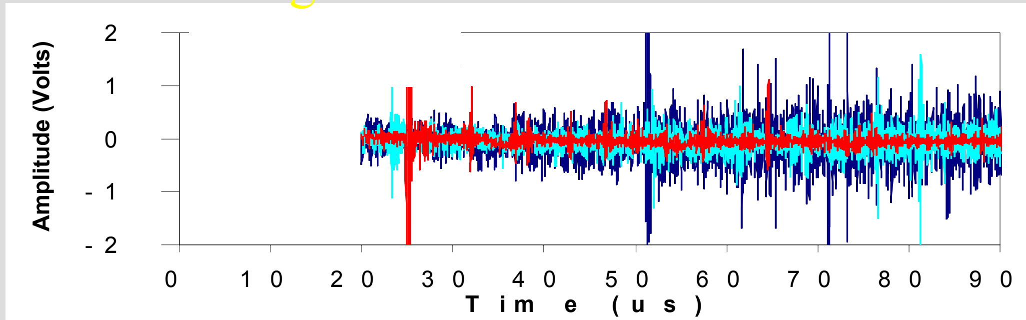


Attenuation, Velocity

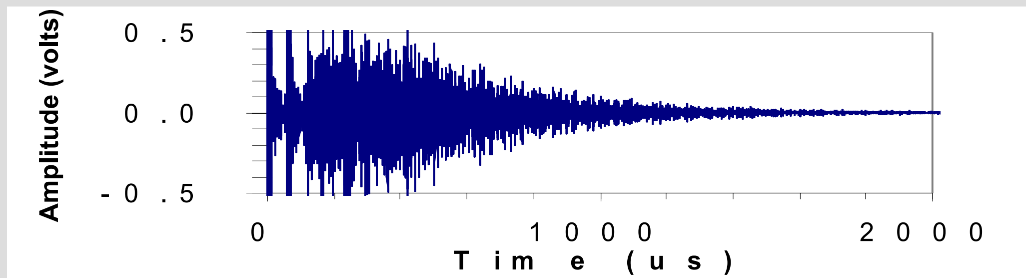
Attenuation and Velocity



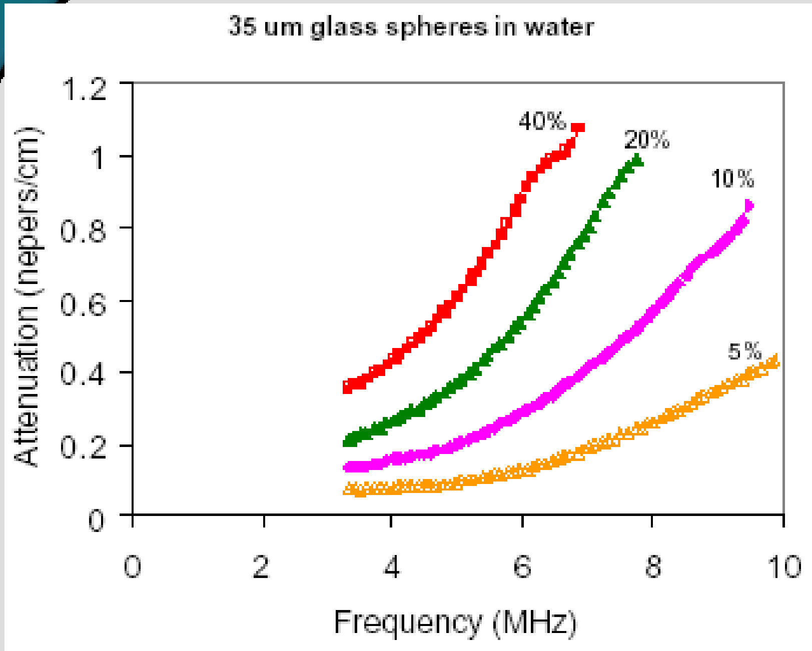
Backscattering

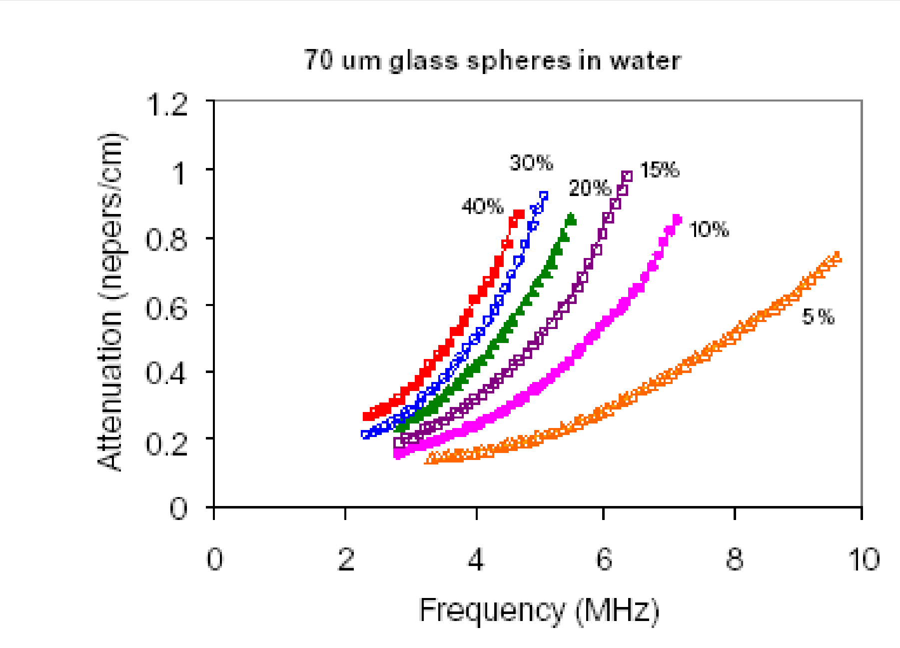


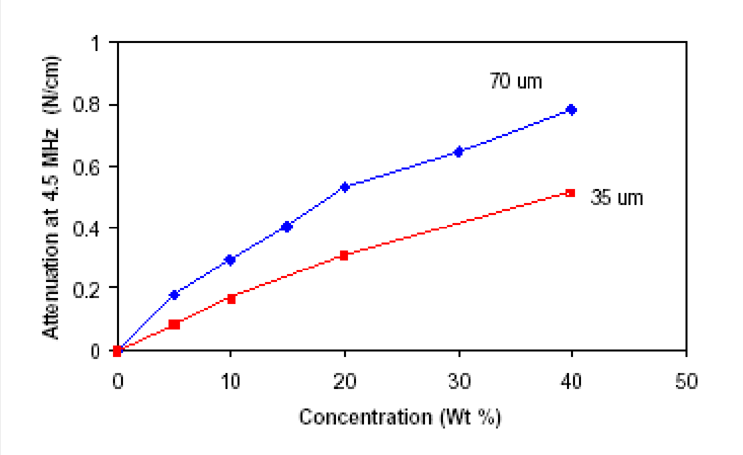
Diffuse field

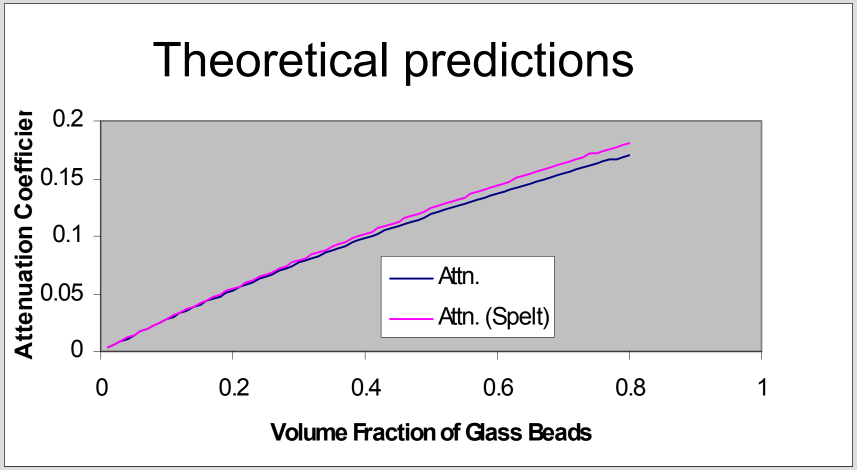


Glass spheres



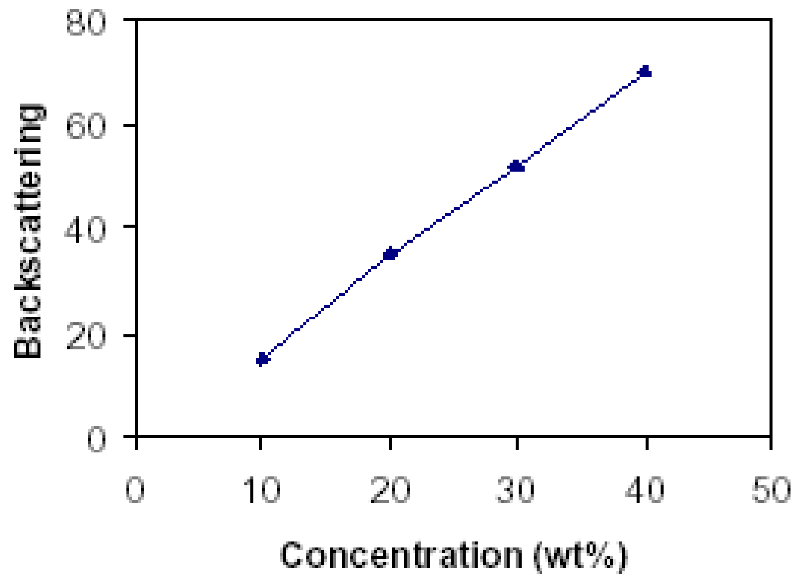




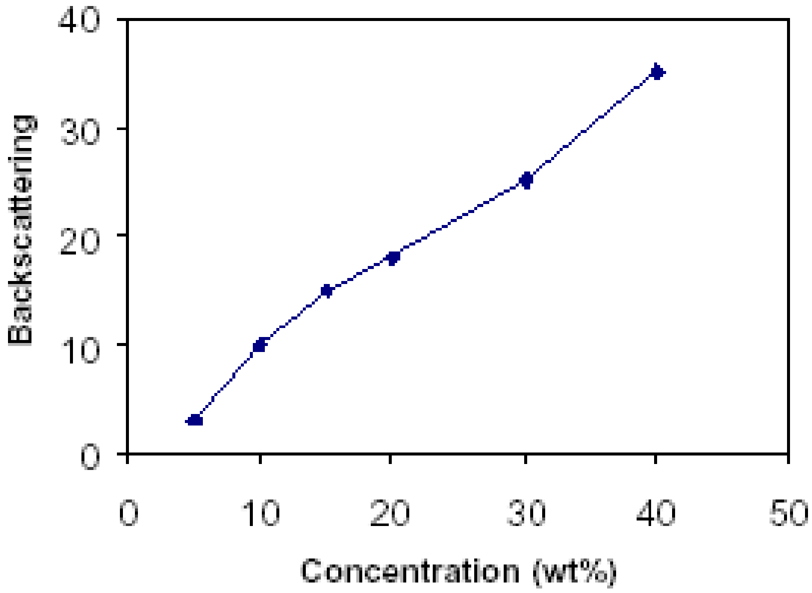


Backscattering

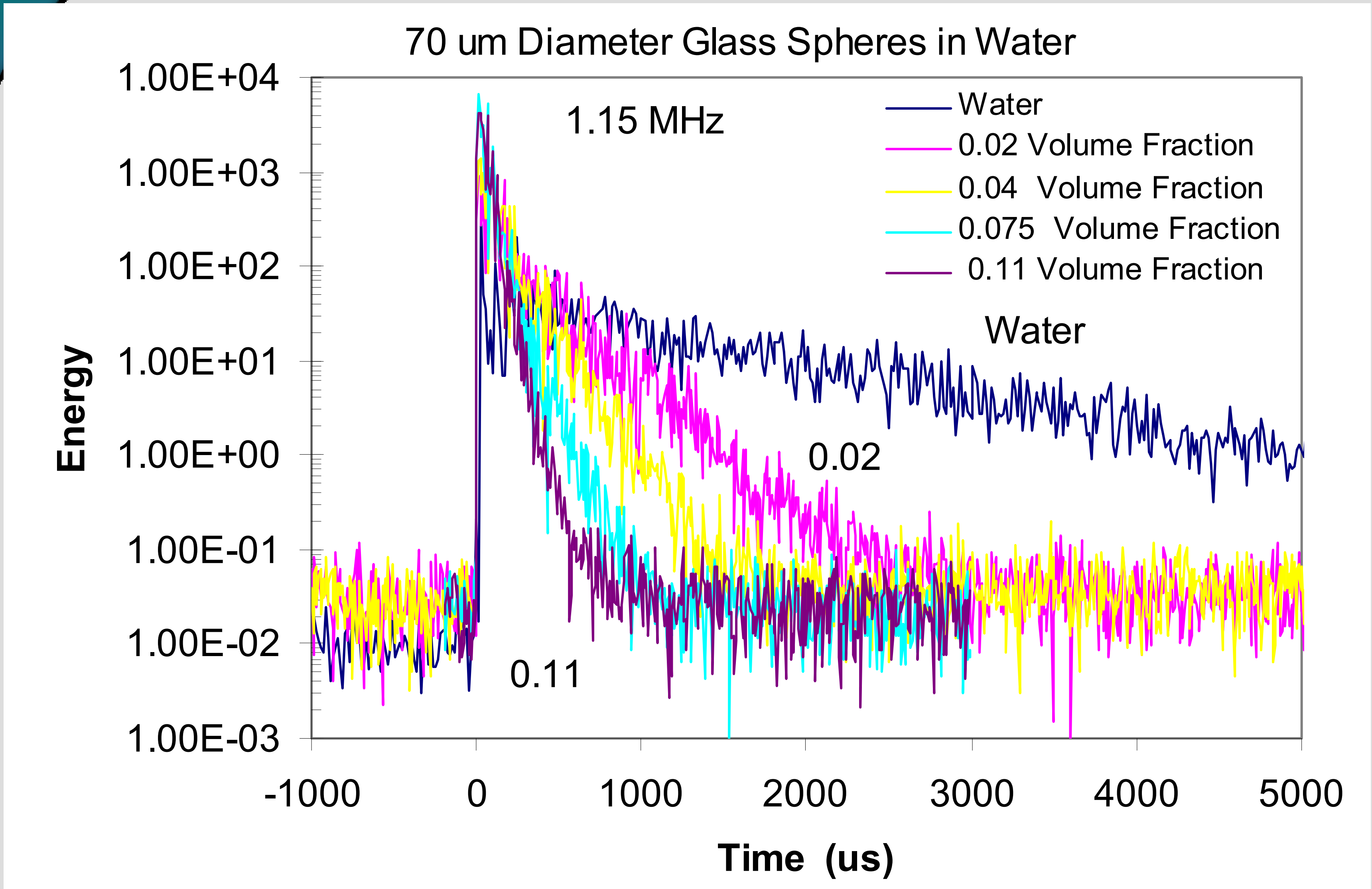
35 um glass spheres



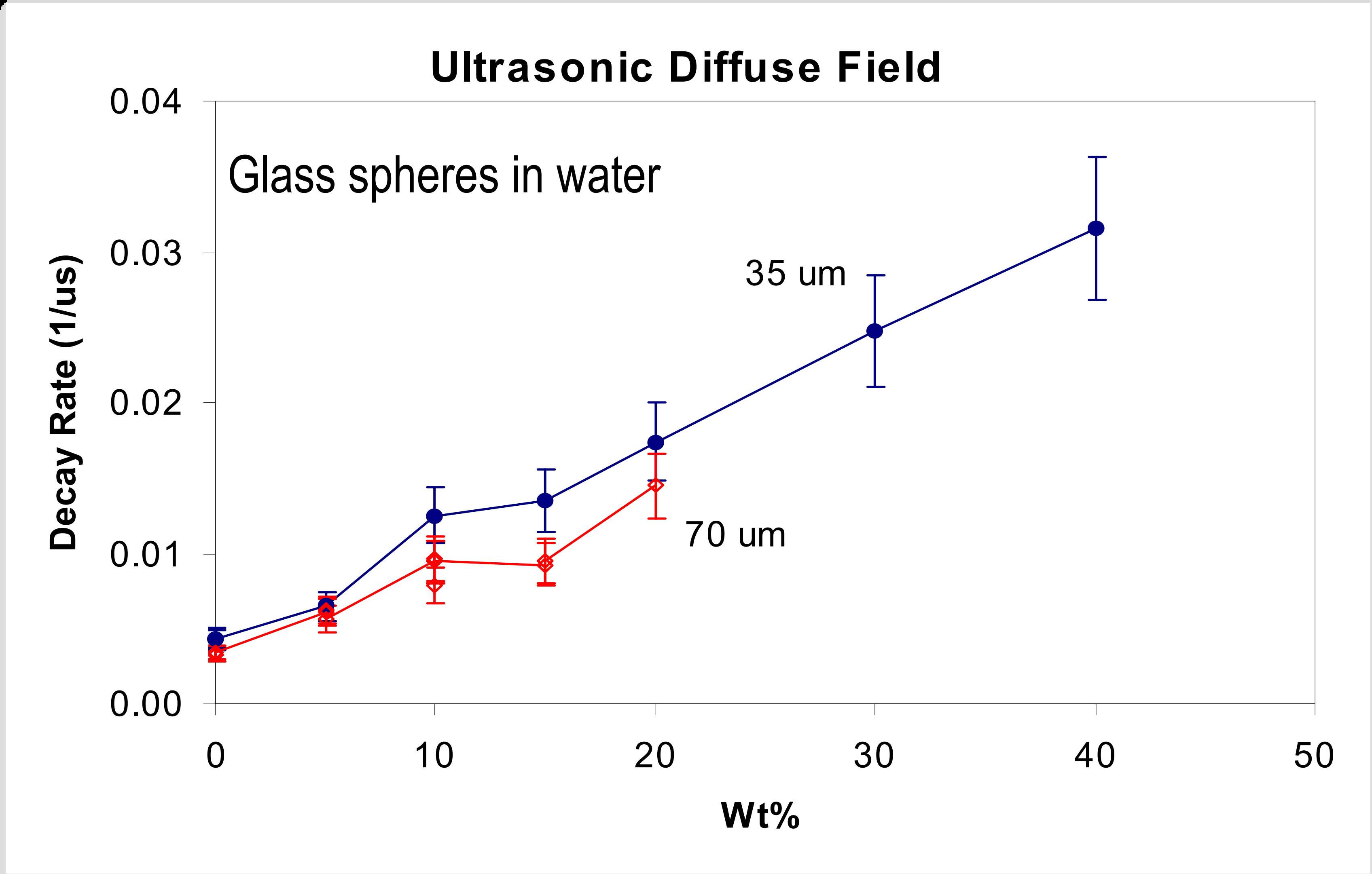
70 um glass spheres



Ultrasonic Diffuse Field



Diffuse field decay rate vs. concentration



Regimes of Sound Propagation

- **Viscous regime**: Boundary layer surrounding the particle is thick compared to the particle radius.

$$\alpha \propto a^2 f^2 / \mu$$

- **Inertial regime:** Boundary layer thickness is very small compared to the particle radius.

$$\alpha \propto (\mu f)^{1/2} / a$$

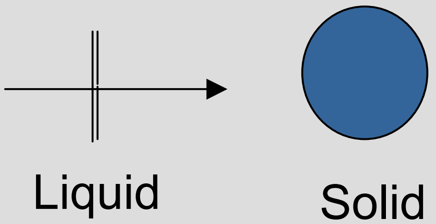
- **Multiple scattering regime**: Particles are large compared to the wavelength of sound waves.

$$\alpha \propto f^4$$

Mathematical Description

- Allegra-Hawley Equations:

Wave equation



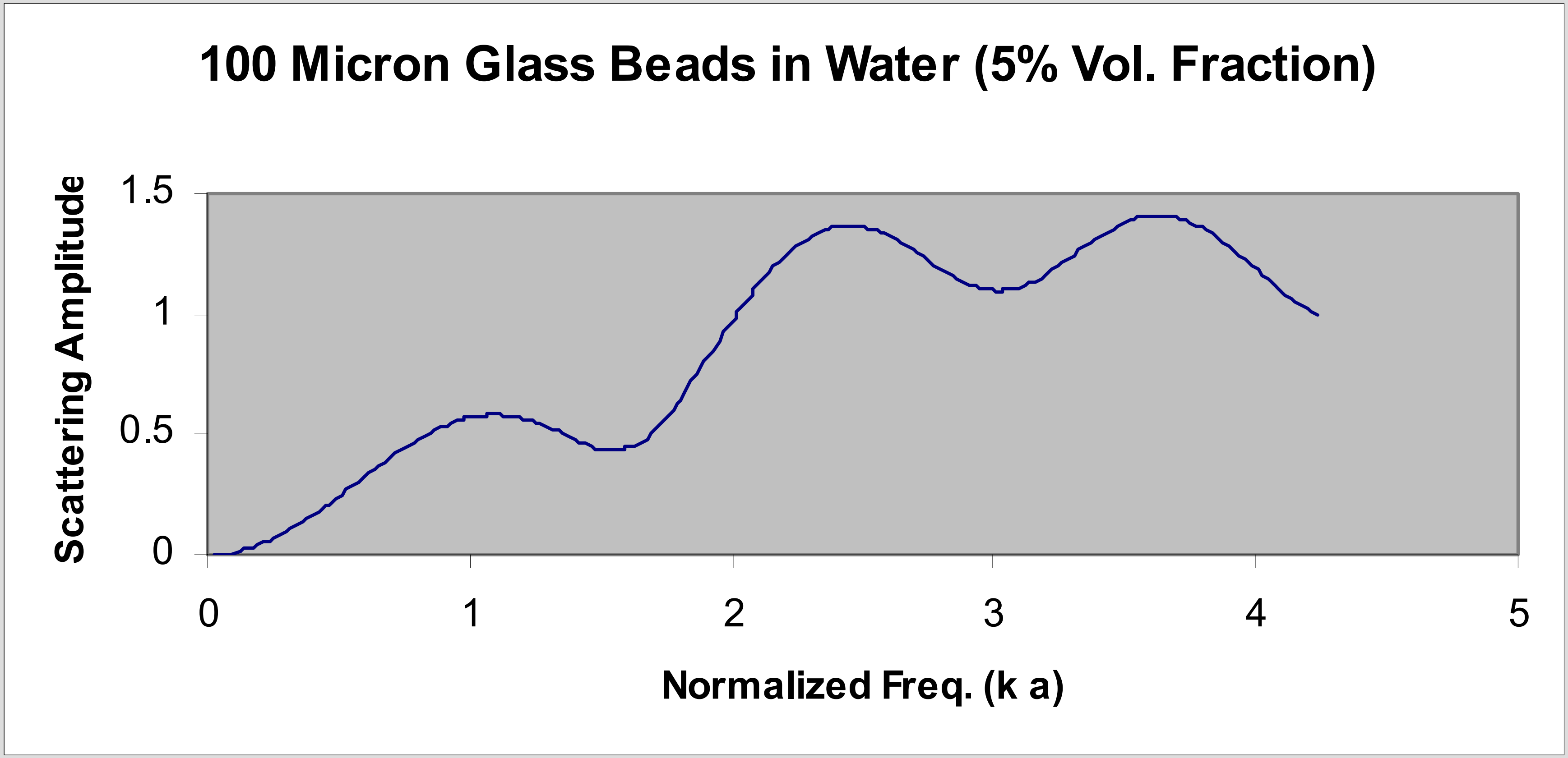
$$\vec{v} = -\nabla(\phi_c + \phi_t) + \nabla \times \vec{A}$$

$$(\nabla^2 + k_c^2)\phi_c = 0$$ **Compressional**

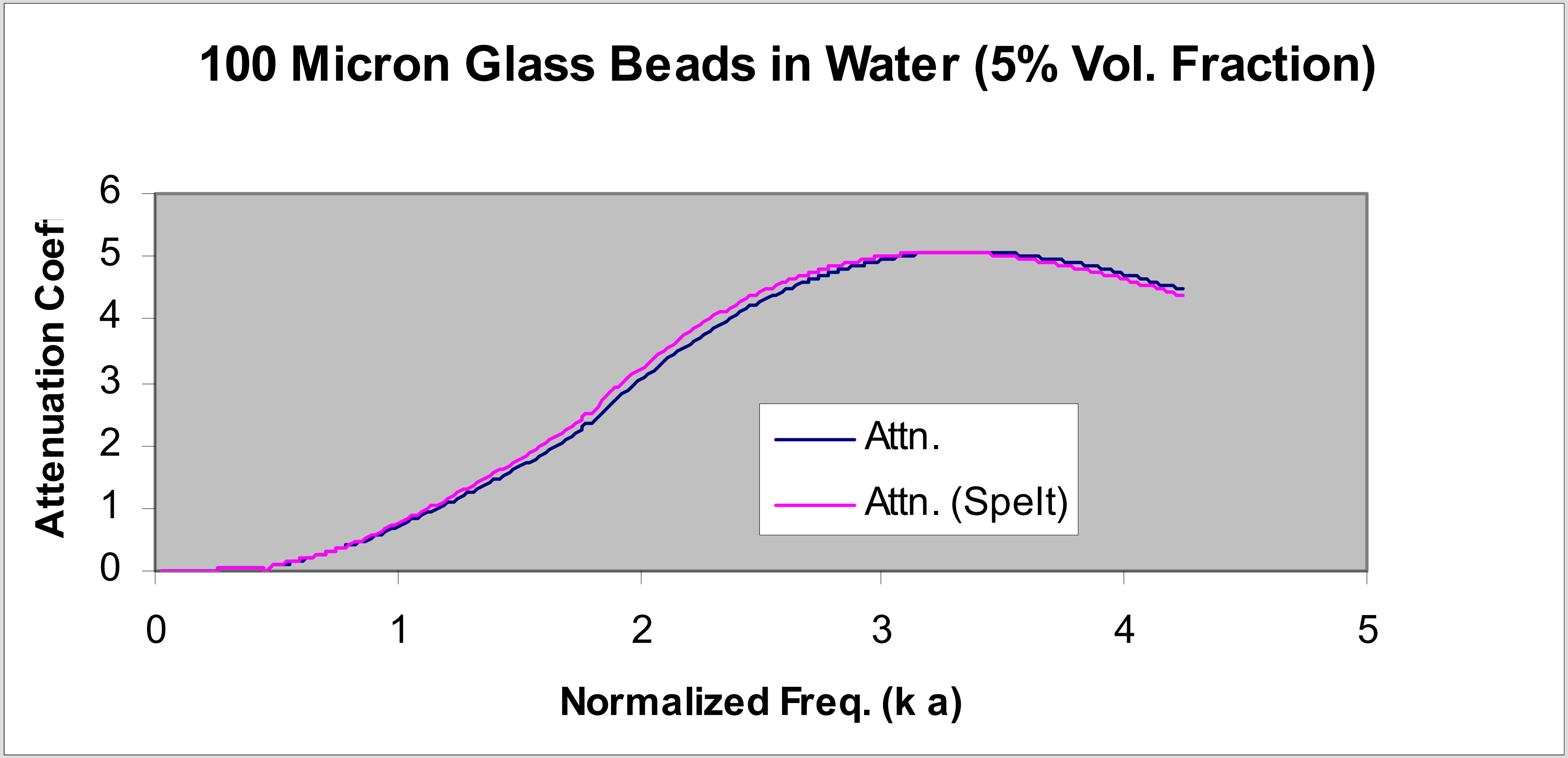
$$(\nabla^2 + k_c^2)\phi_t = 0$$ **Thermal**

$$(\nabla^2 + k_c^2)\vec{A} = 0$$ **Shear**

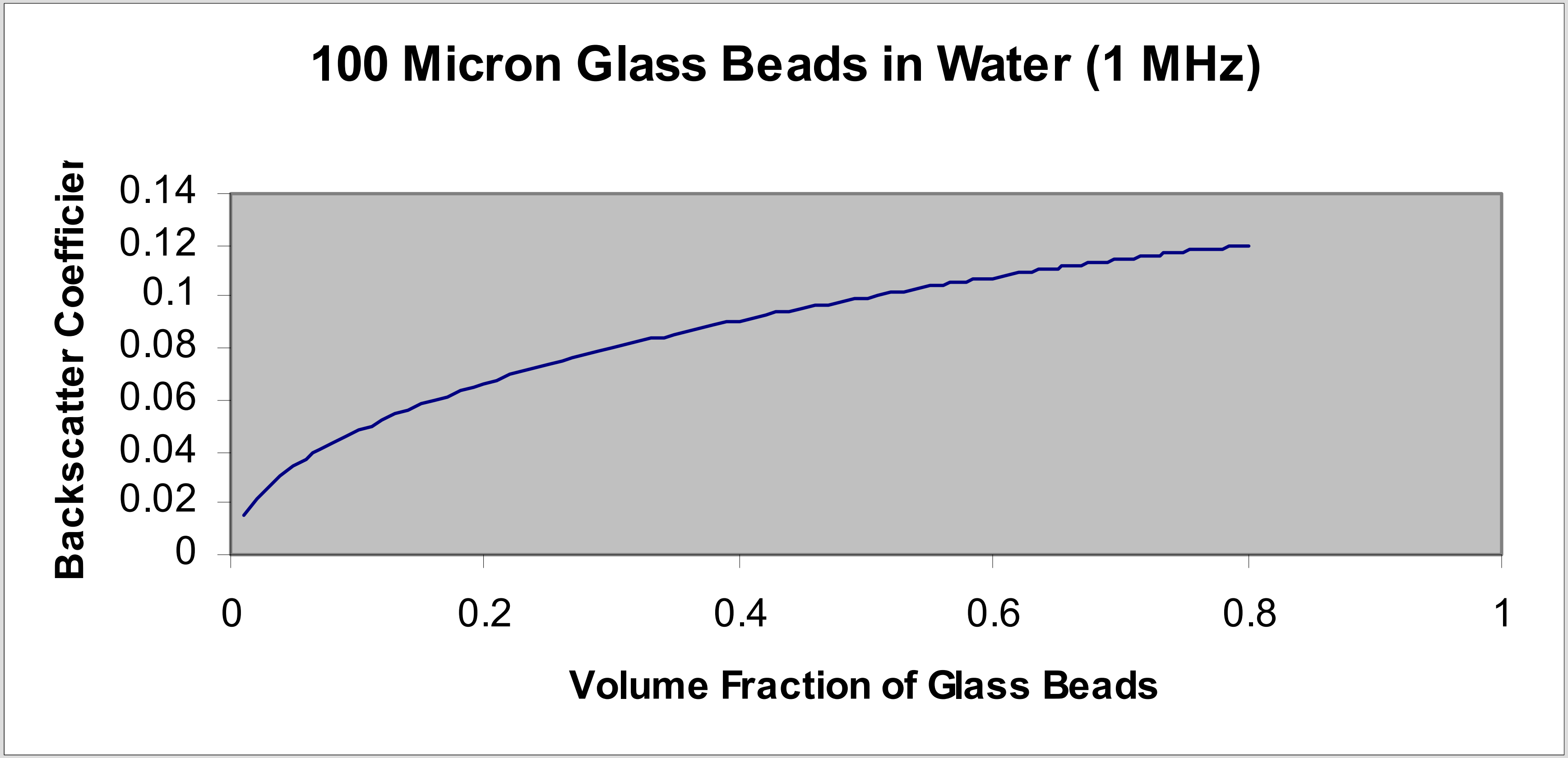
Glass Beads in Water



Glass Beads in Water



Glass Beads in Water

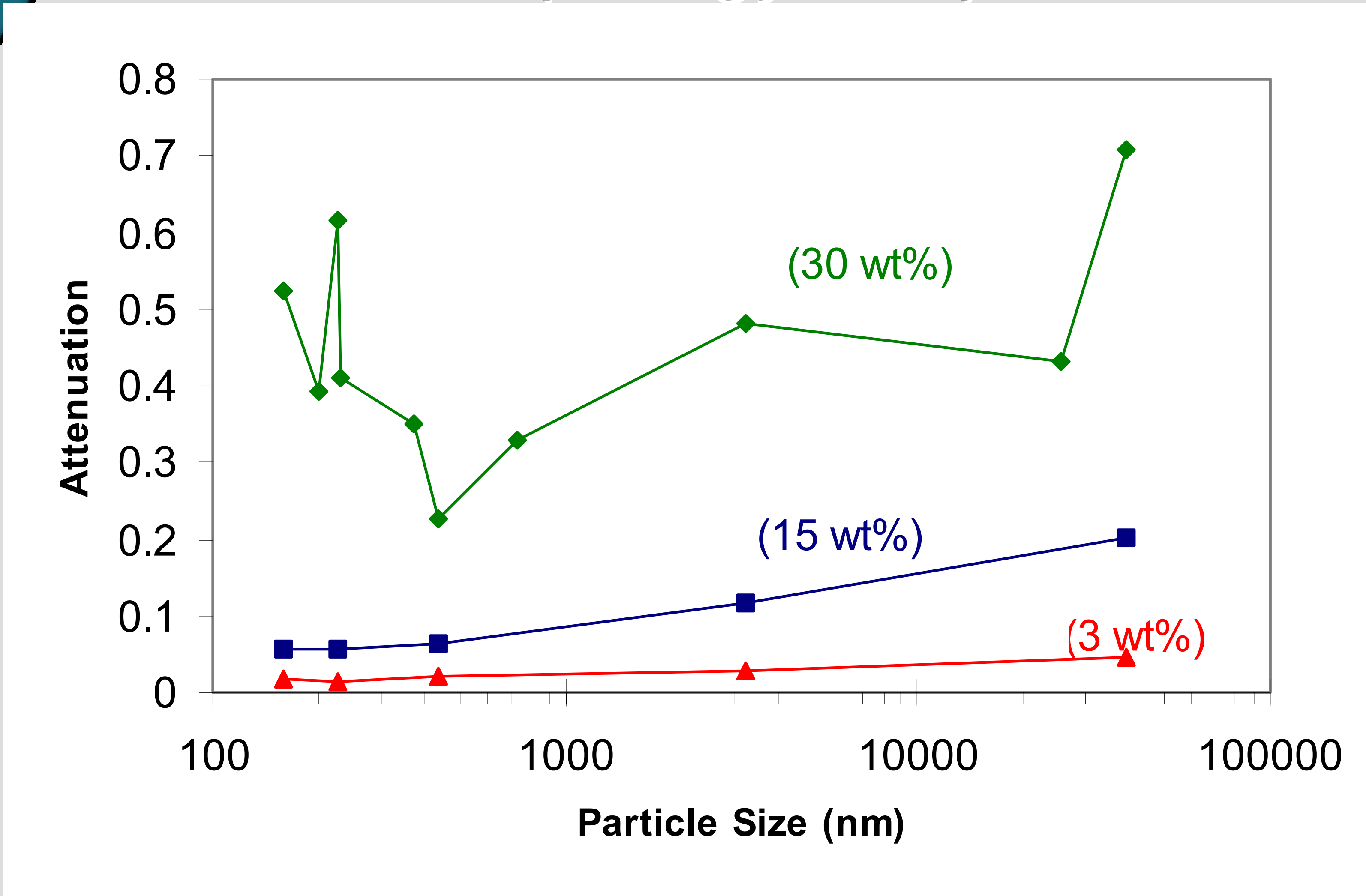


Characteristics of Attrition Samples 30%, 15%, 1% w/w

Time Point (hr)	Particle Size (μm)
0	39.270
1	25.500
2	3.263
4	0.731
6	0.438
8	0.369
10	0.232
12	0.228
14	0.199
16	0.158

- Methodology development
- Follow dynamic process
- Multiple scattering regimes
- Known concentration
- Known optical and acoustic properties

Typical measurement of attenuation (energy loss)



Energy loss is concentration dependant
Problems at high concentration

Problems with attenuation

- **Problem:** Attenuation has problems determining particle size at high concentrations – multiple scattering
- **Solution:**

Alternative interpretations

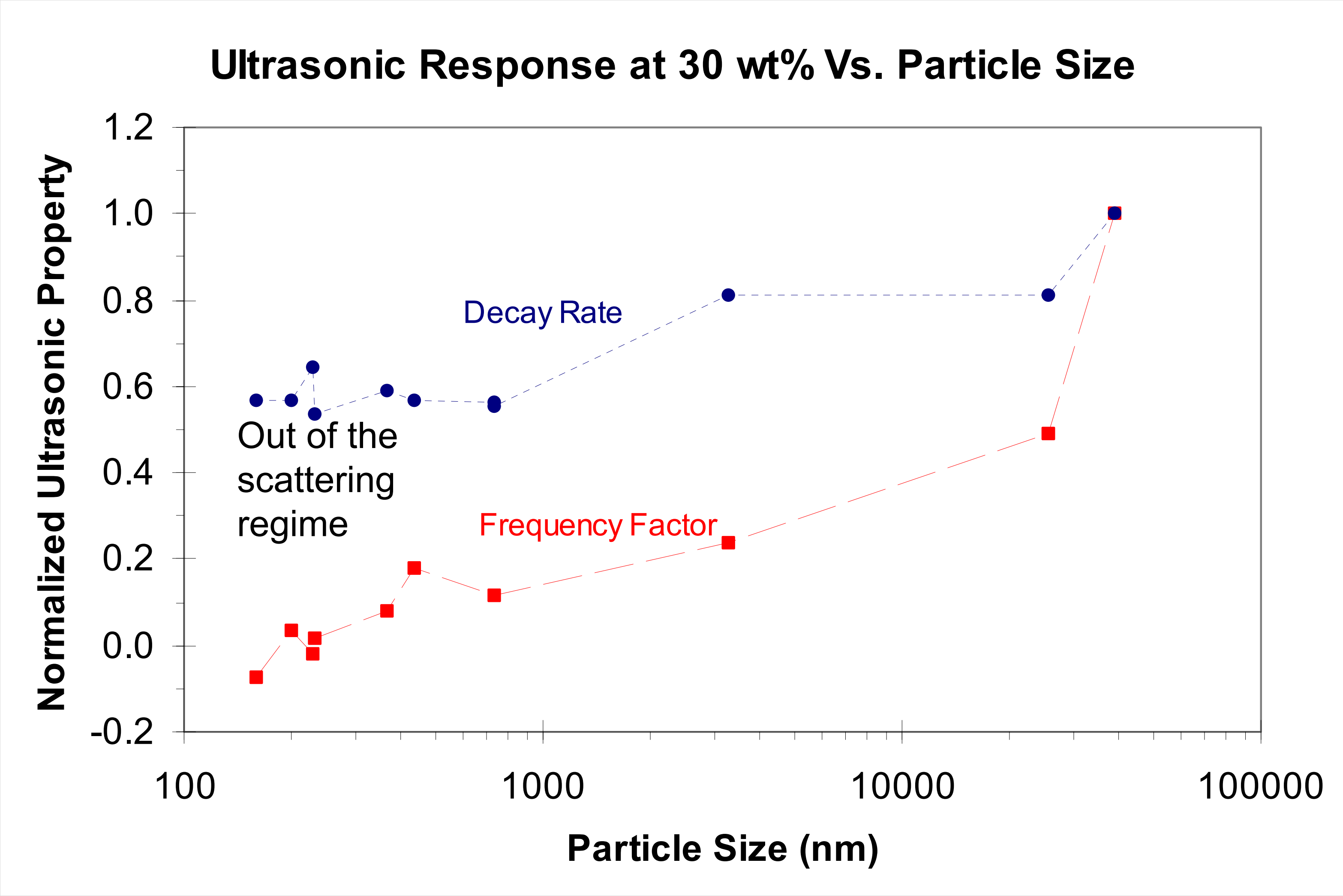
- Frequency response is a function of particle size
- Small particles → Large particles

  Viscous Inertial Multiple scattering

Alternative measurements

- Backscattering response and decay rate

Ultrasonic response as a function of Particle size



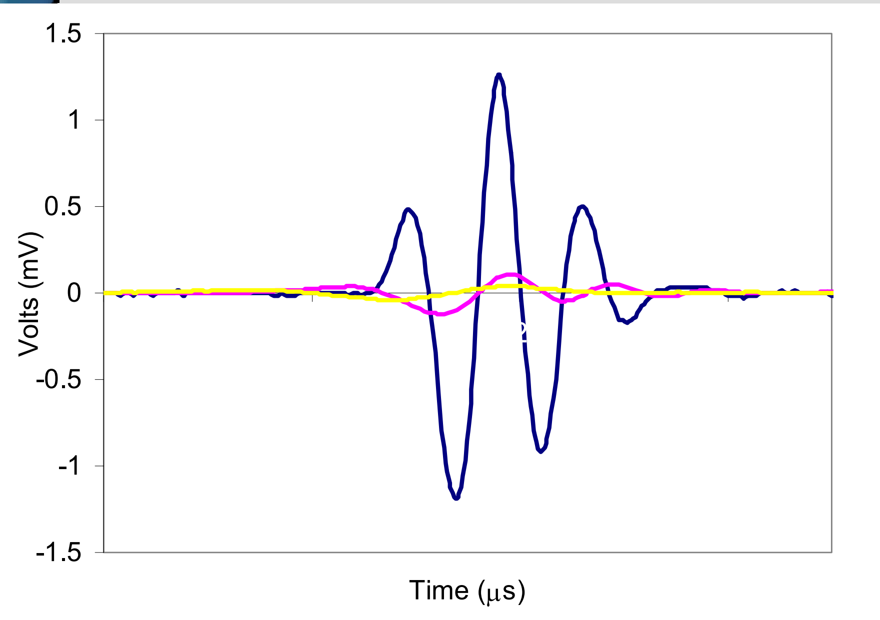
AZ 101/102 tank simulant

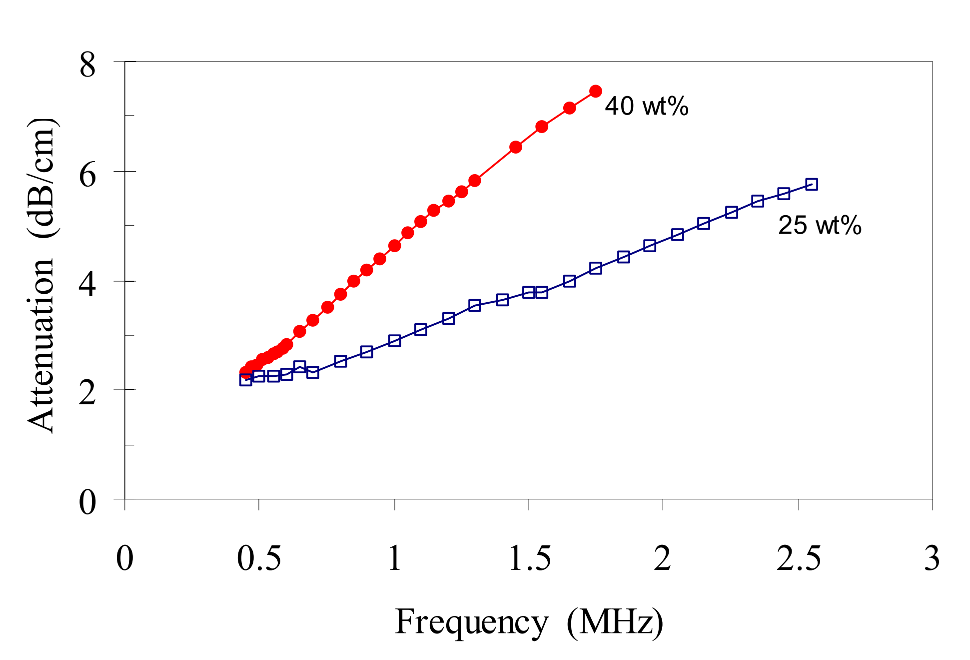
Table S.1. Inactive AZ-101/102 Filtration Simulant Composition

Solids Components					
Compounds Bearing:	**Wt %**	**Mineral Phase**	**Powder Grade**	**Mean Volume PSD (Distribution)**	**Wt %**
Iron	58	Hematite	Iron Oxide No: 07-5001	22 µm	17.400
			Red Iron Oxide No: 07-3752	2–3 µm	29.000
			Synthetic Red Iron Oxide No: 07-2568	0.6 µm	11.600
Aluminum	24	Beohmite	HiQ-10 Alumina	0.0028–0.004 µm	7.200
		Gibbsite	C-231 Ground White Hydrate	14 µm (broad)	8.400
			SpaceRite S-23 Alumina	7.5 µm (broad)	5.040
			SpaceRite S-11 Alumina	0.25 µm (narrow)	3.360
		Gibbsite/Beohmite Ratio: 2.33			
Zirconium	13	Zirconium Hydroxide	Zirconium Hydroxide; Product Code: FZO922/01	15 µm	13.000
Silicon	5	Nepheline	Spectrum A 400 Nepheline Syenite	10 µm	5.000

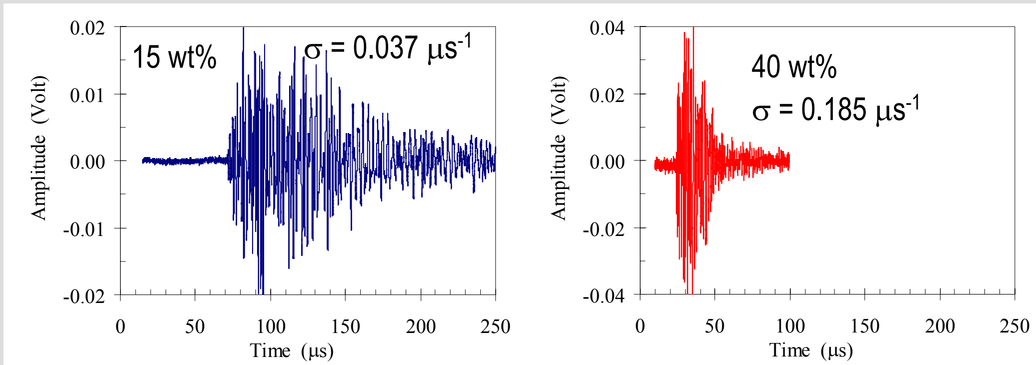
Supernatant Components		
Component	**Concentration (M)**	**Concentration (g/L)**
NaOH	0.8	32
$NaNO_3$	1.0	85

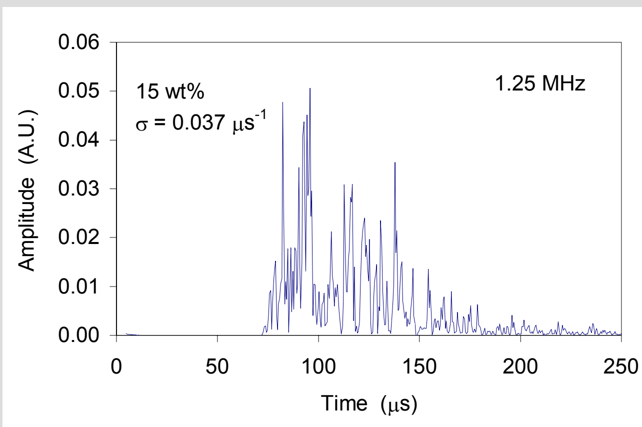
Ultrasonic Attenuation (Surrogate Slurry)

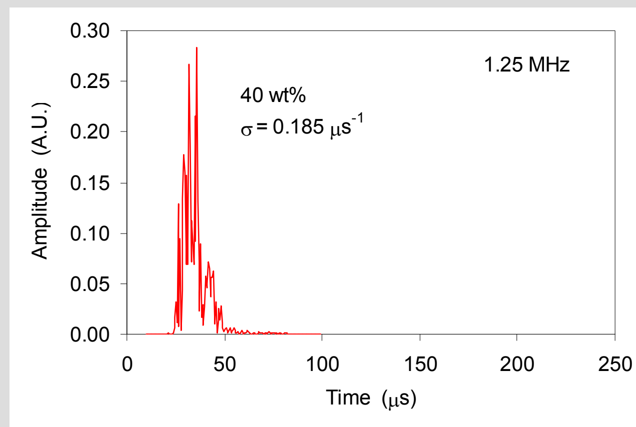




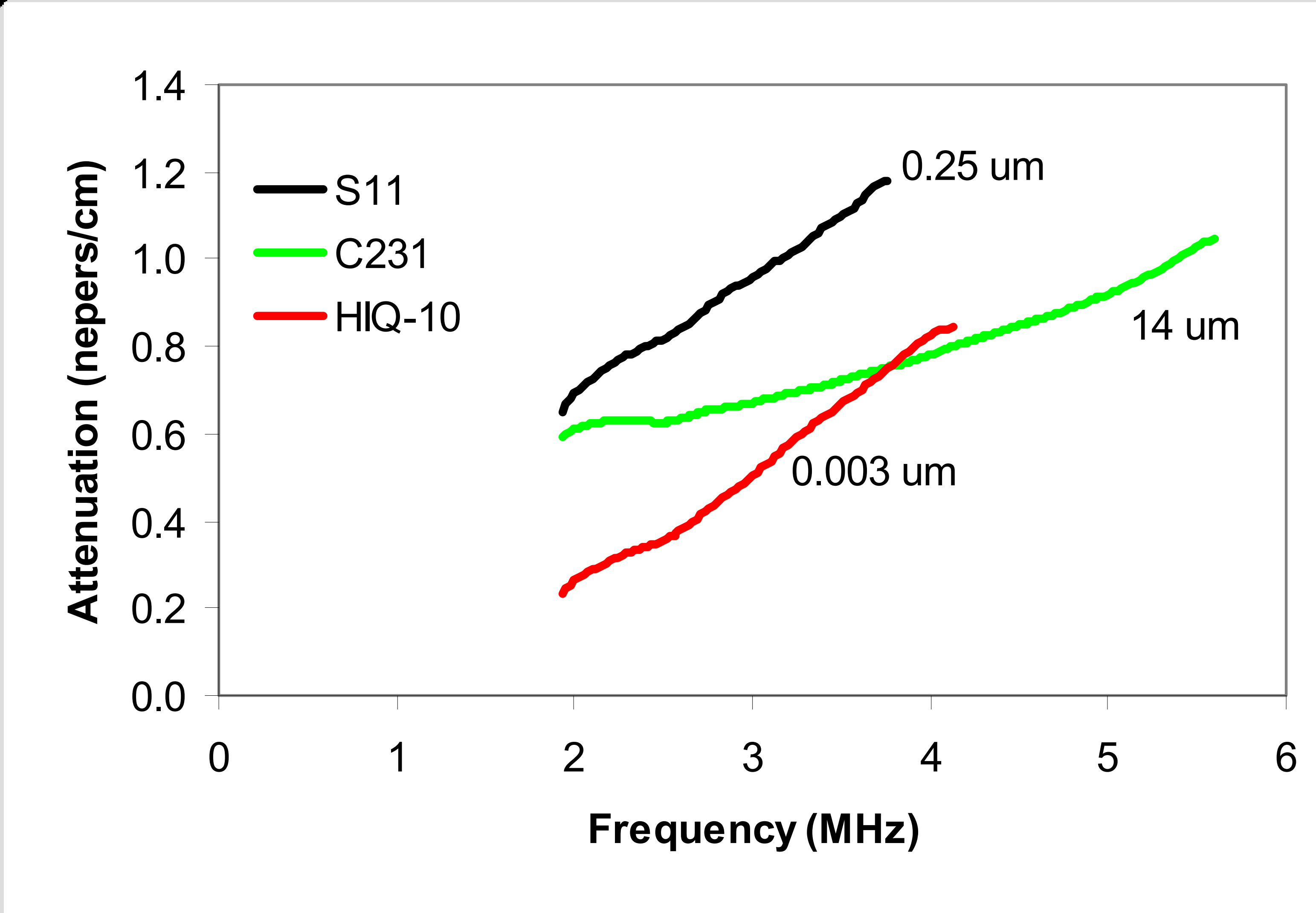
Surrogate Slurry (Ultrasonic Diffuse Field Results)







Alumina subset of AZ 101/102 tank simulant

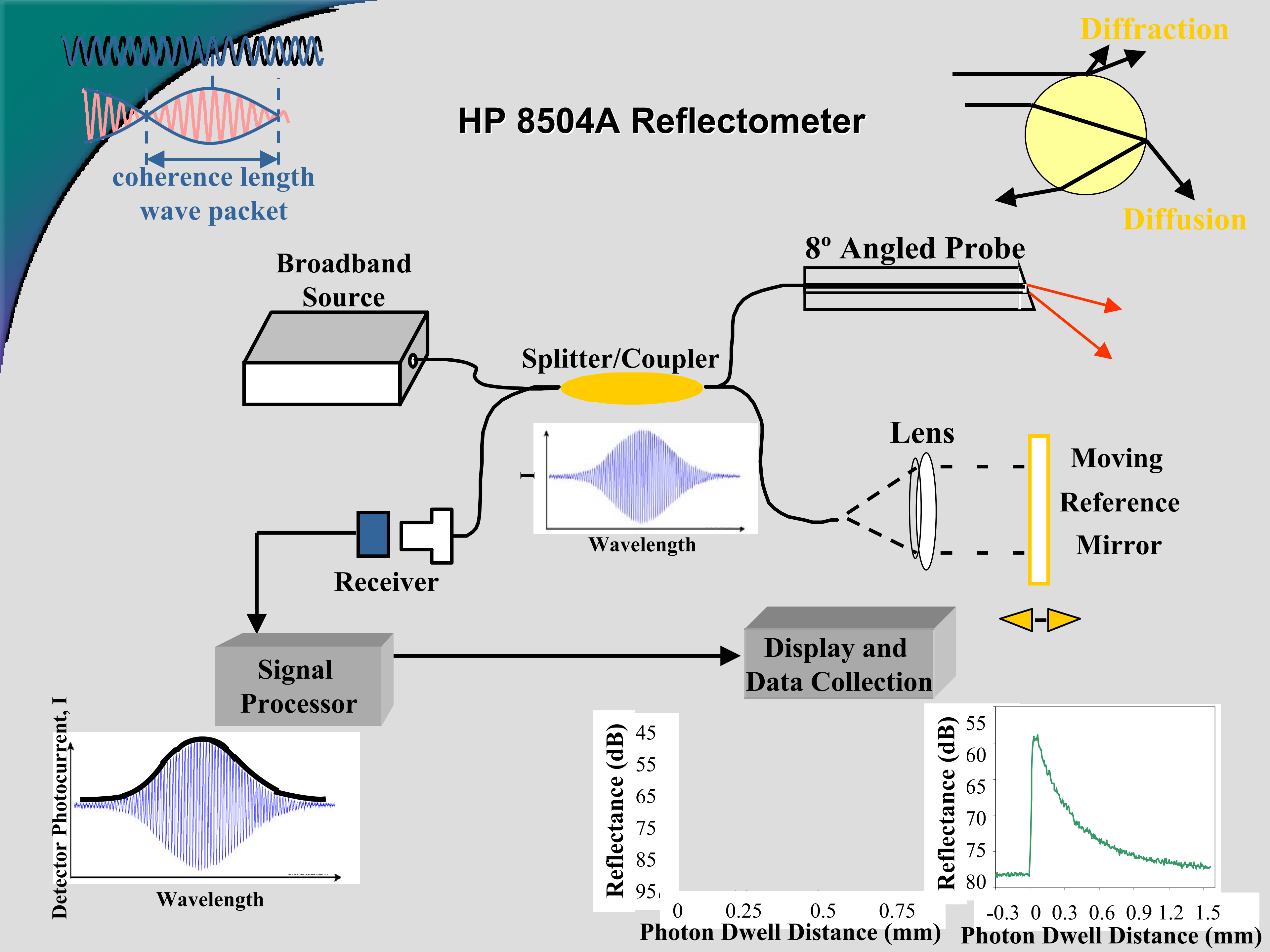


Unexpected behavior – possibly indicates agglomeration
Consistent with optical results

UW Optical Team

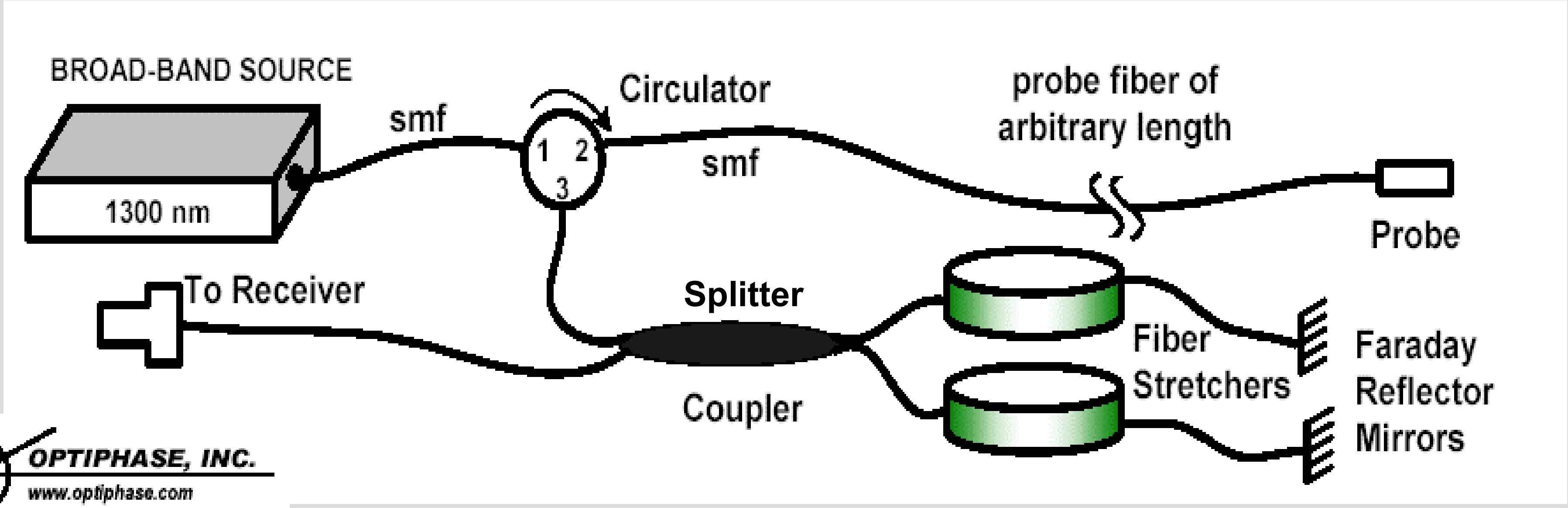
Lloyd Burgess, Anatol Brodsky, Summer Randall

HP 8504A Reflectometer



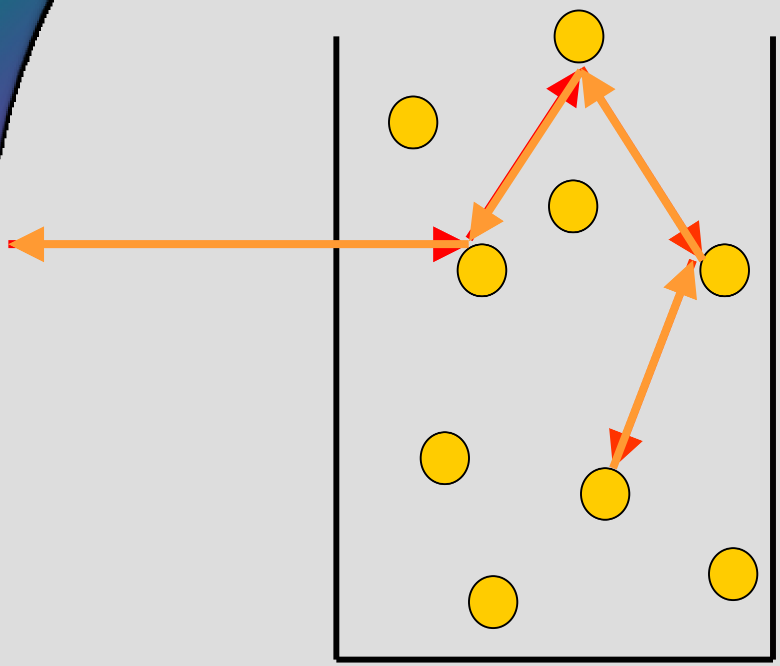
Autocorrelator Instrument

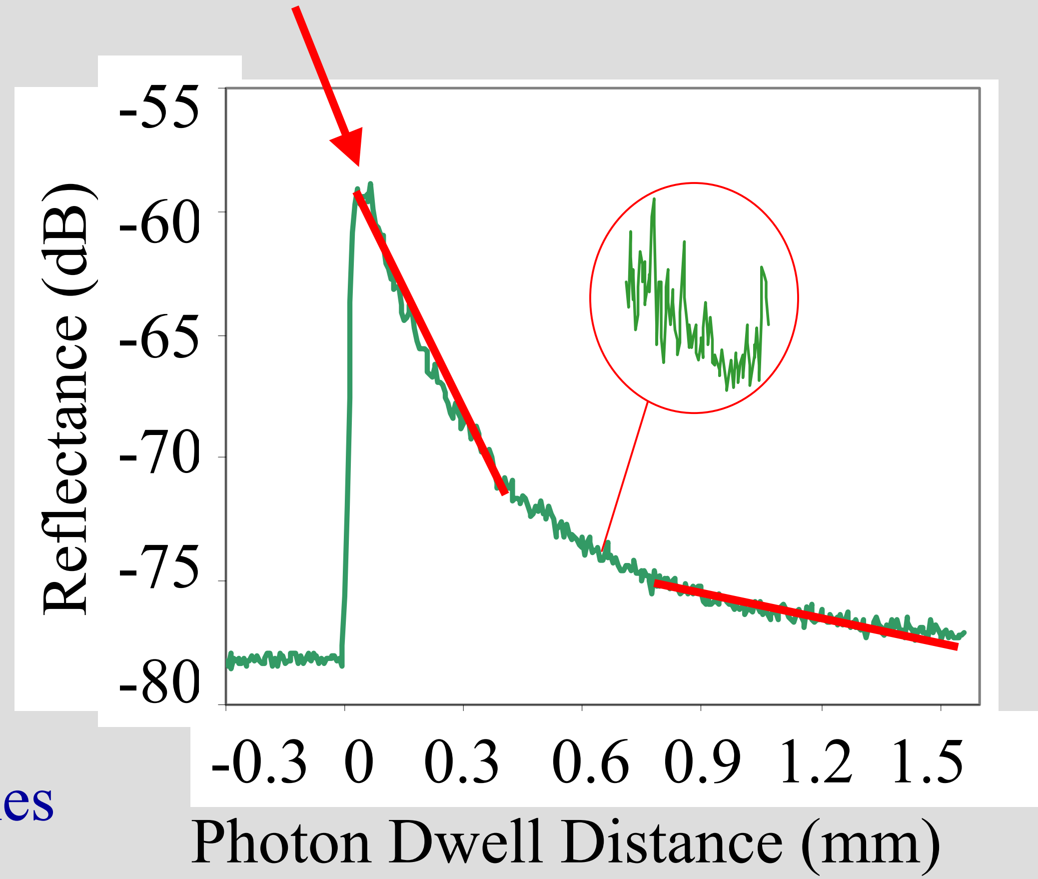
- Probe arm can be any length
- Complex probe design
- Software development for data analysis
- PZT, No mechanically moving parts
- Higher power sources
- Required reference reflection for normalization



OPTIPHASE, INC.
www.optiphase.com

Scattering Materials
Liquids and Solids

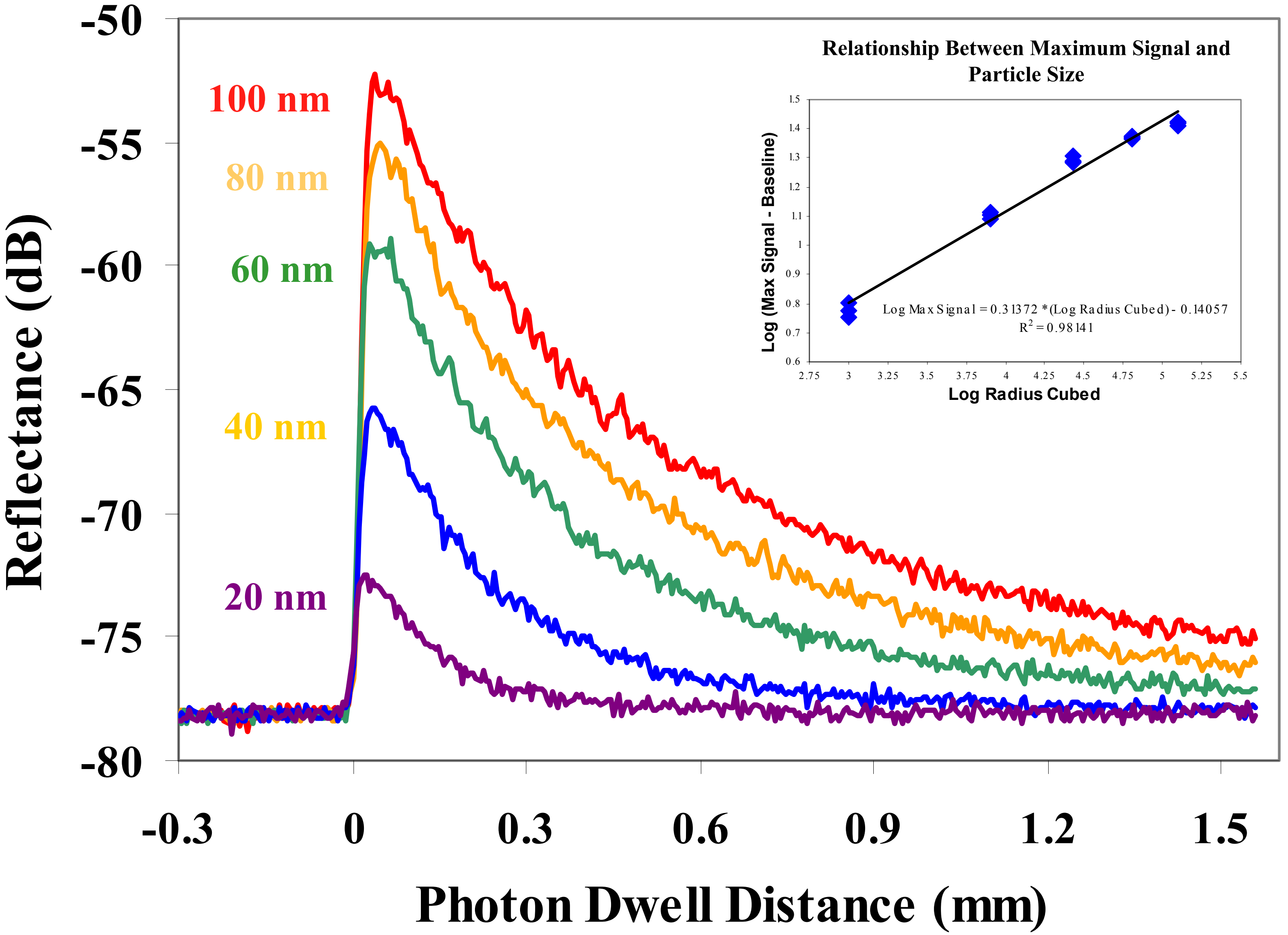




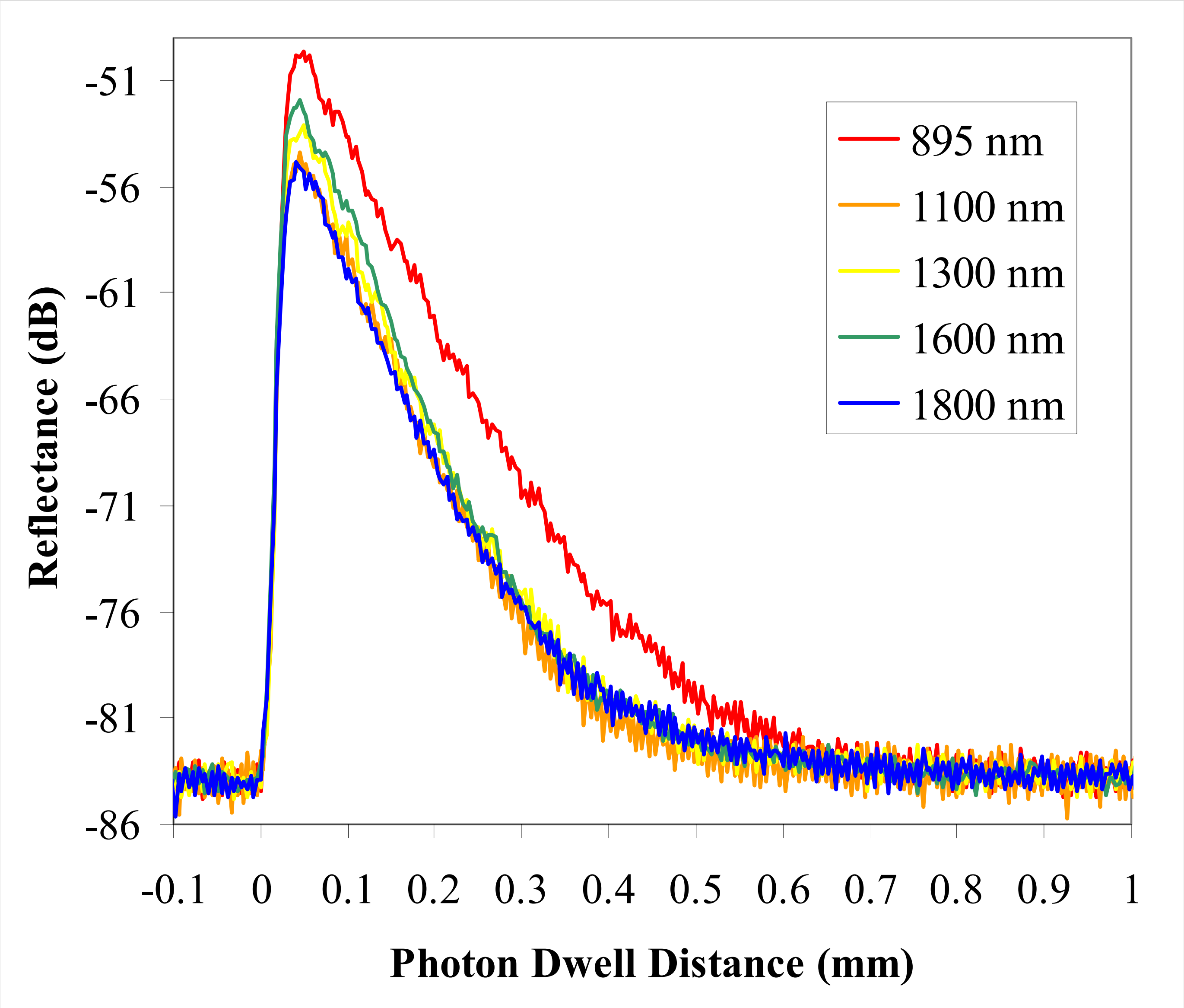
- Paints
- Primers
- Tablet coatings
- Catalyst beads
- Membranes
- Paper
- Emulsions
- Suspensions

Particle Size << λ

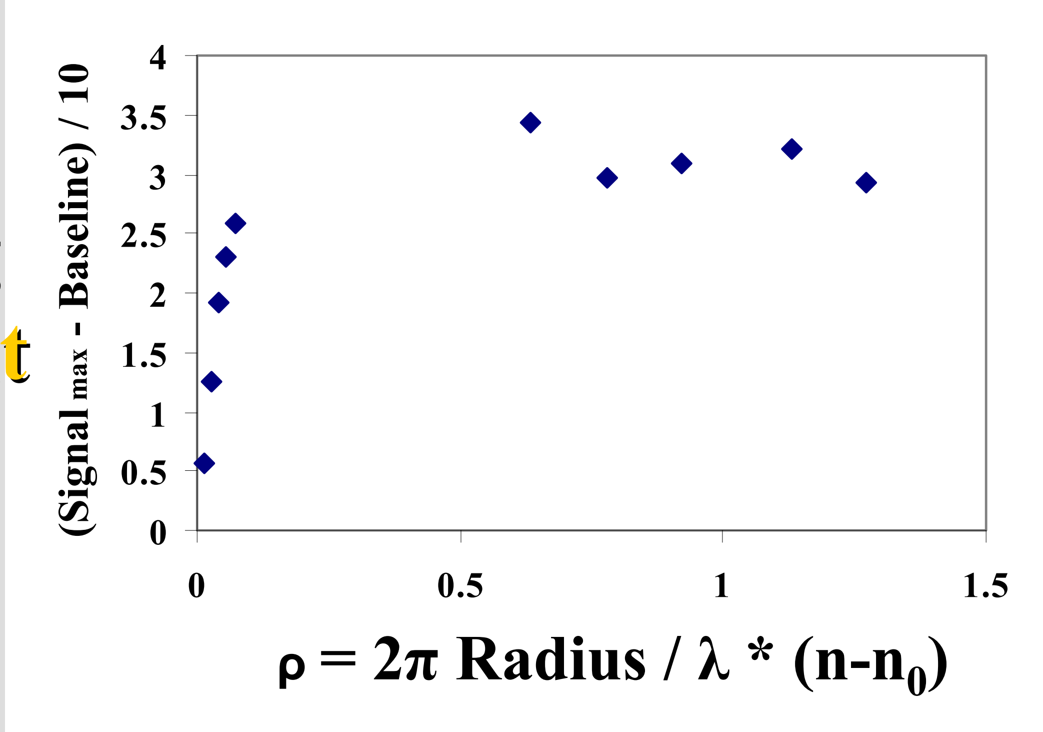
1% Monodispersed Polystyrene Nanospheres



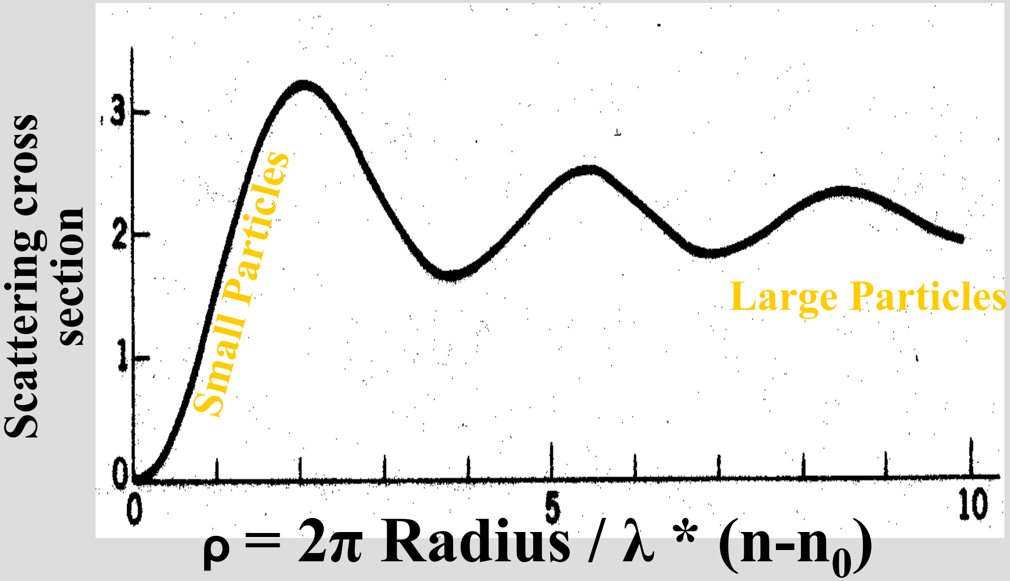
1% Polystyrene Nanospheres
Below, At, and Above the Wavelength



Multiple Scattering Theory Development



Van de Hulst Single Scattering Approximation



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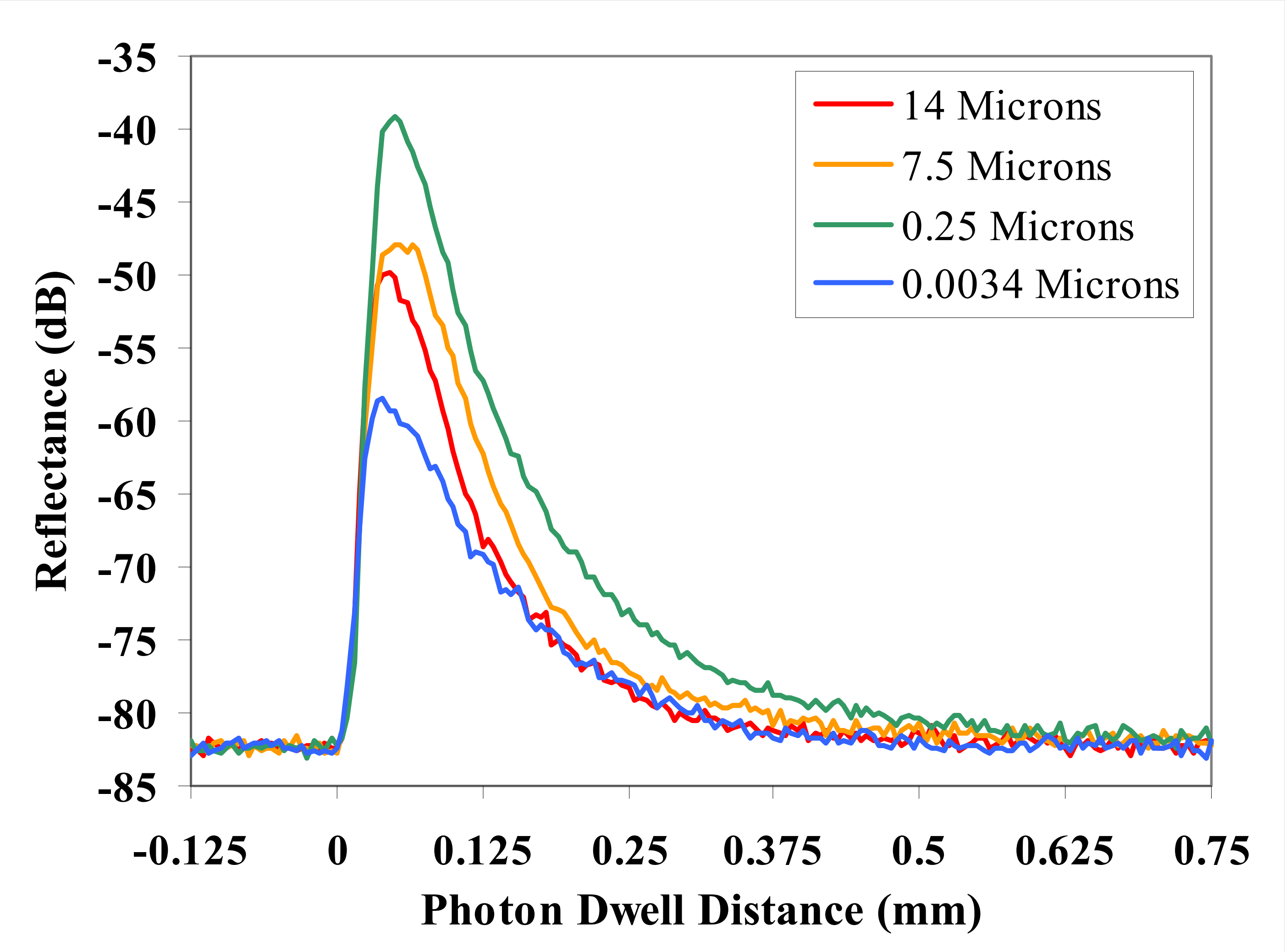
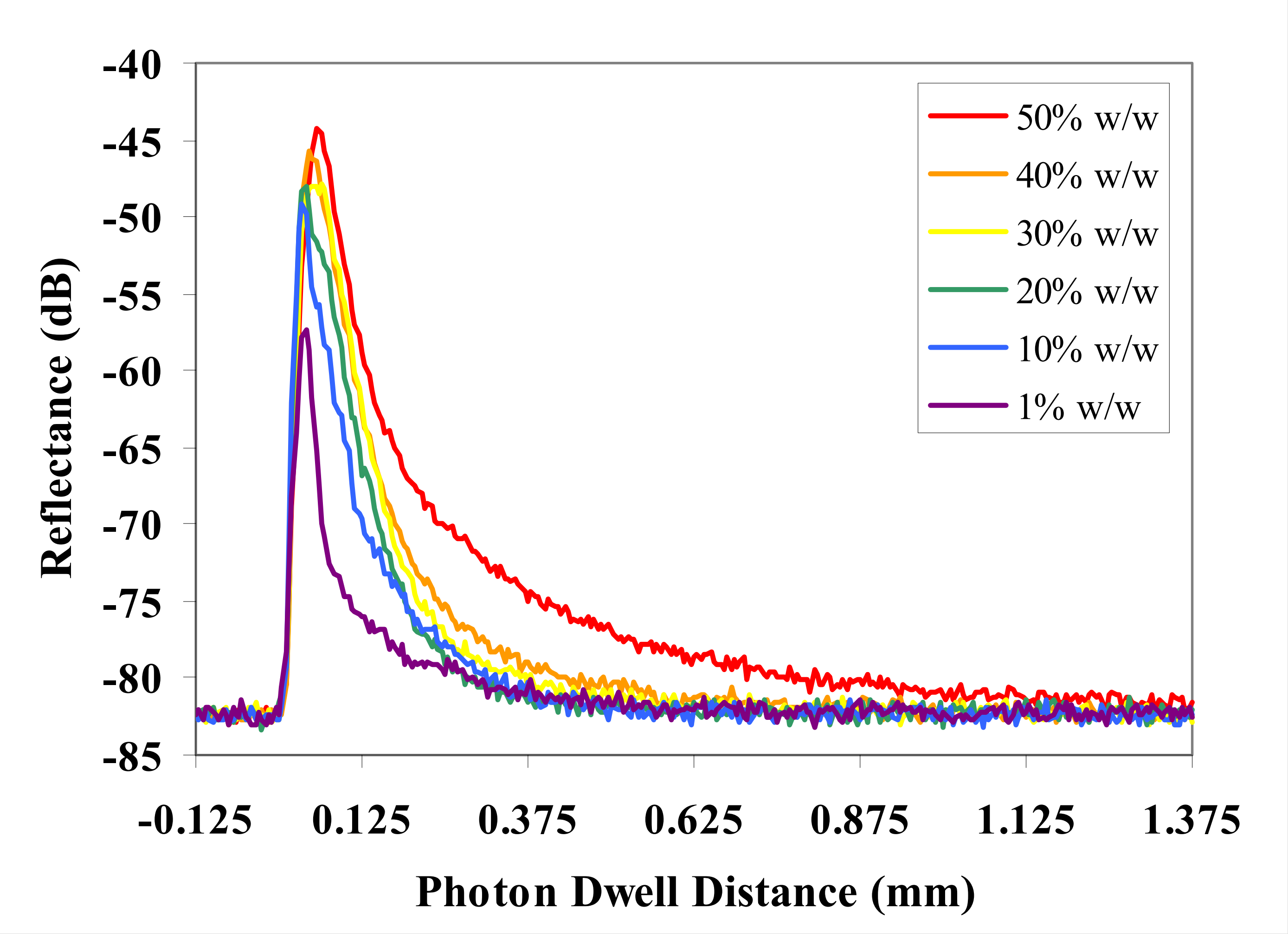
30% w/w Surrogate Alumina Series

AZ 101/102



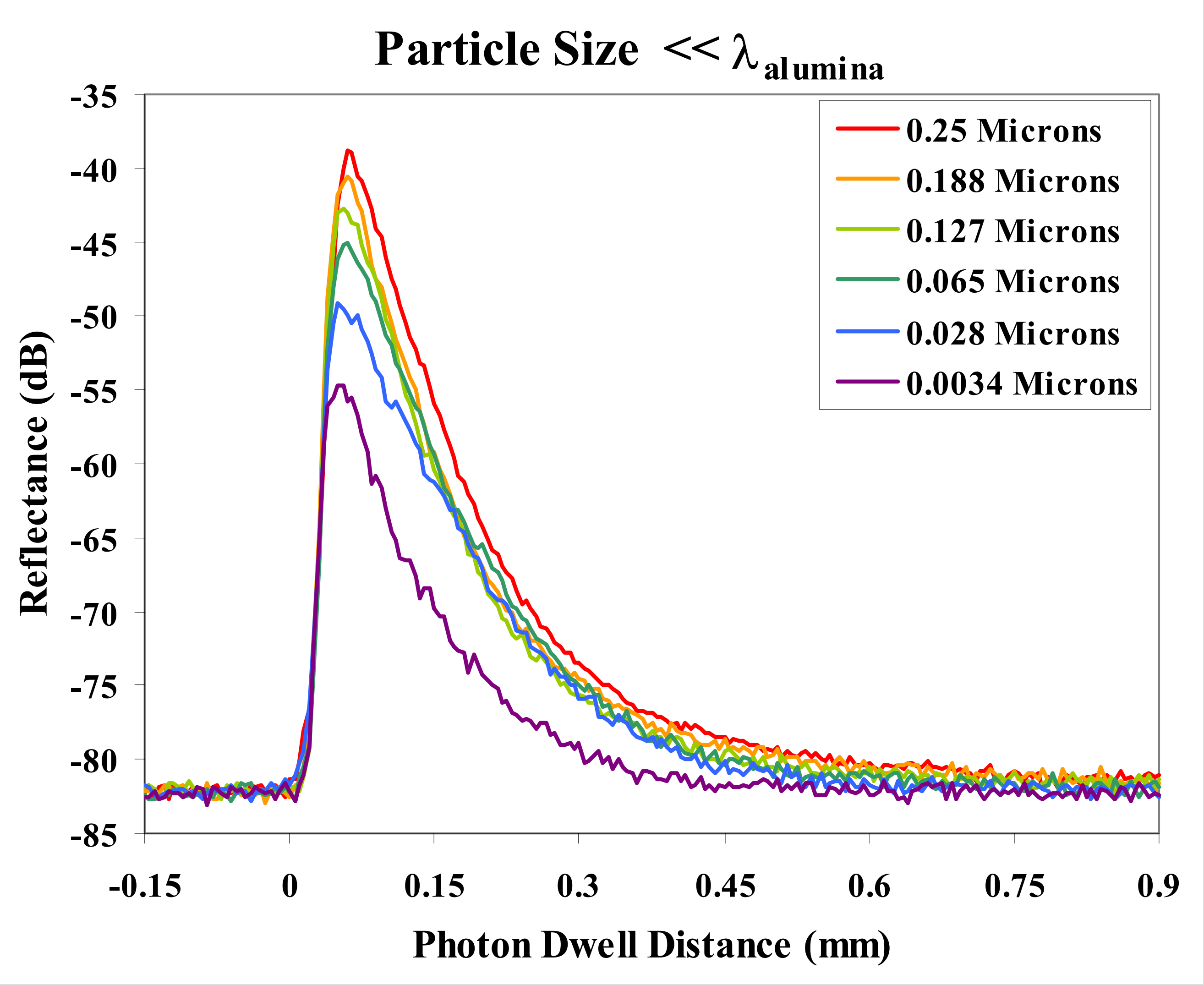
7.5 μm Surrogate Alumina Series

AZ 101/102



DOE Surrogates--Alumina

30% w/w Polydispersed



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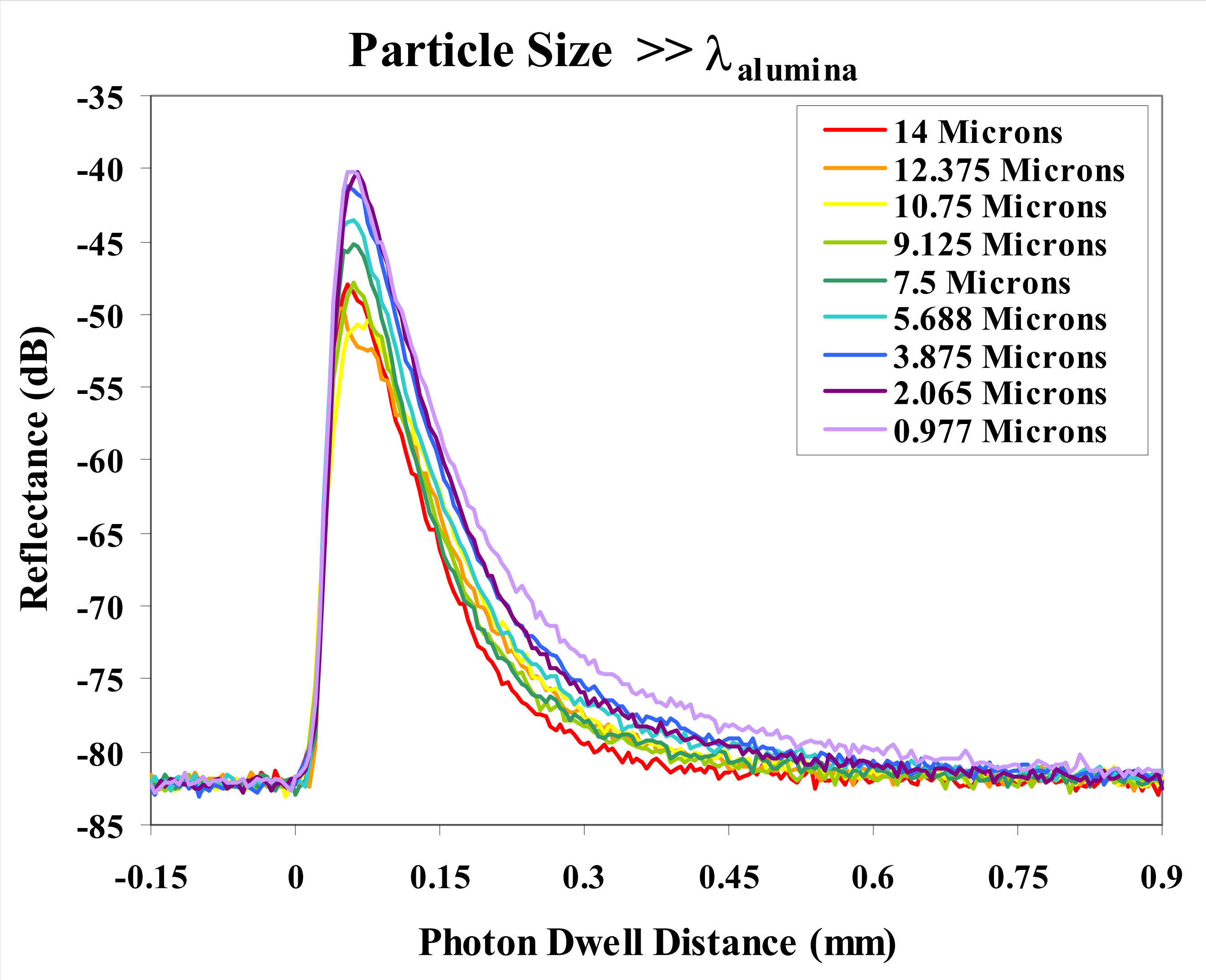

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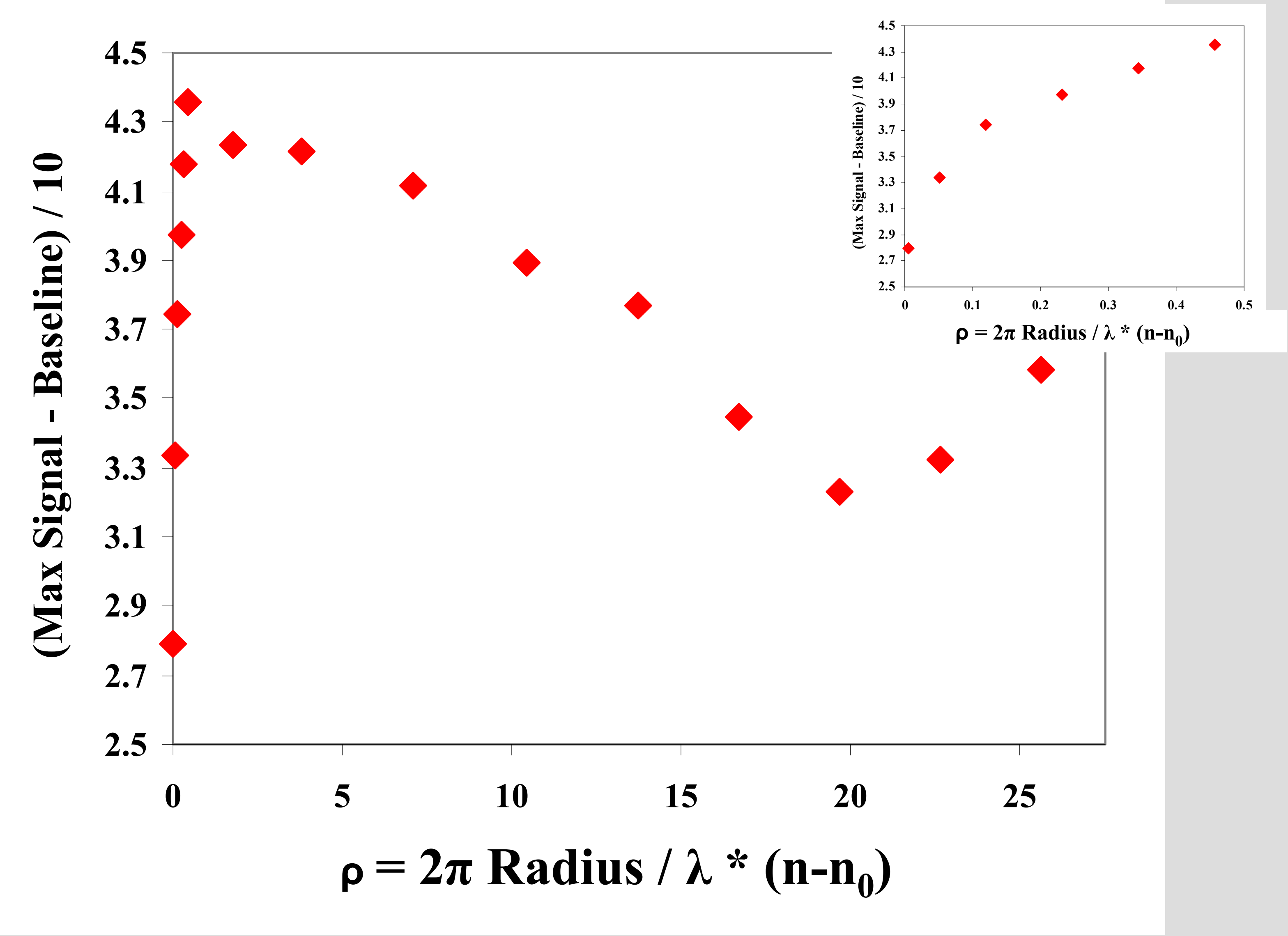

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- Methodology development
- Follow dynamic process
- Multiple scattering regimes
- Known concentration
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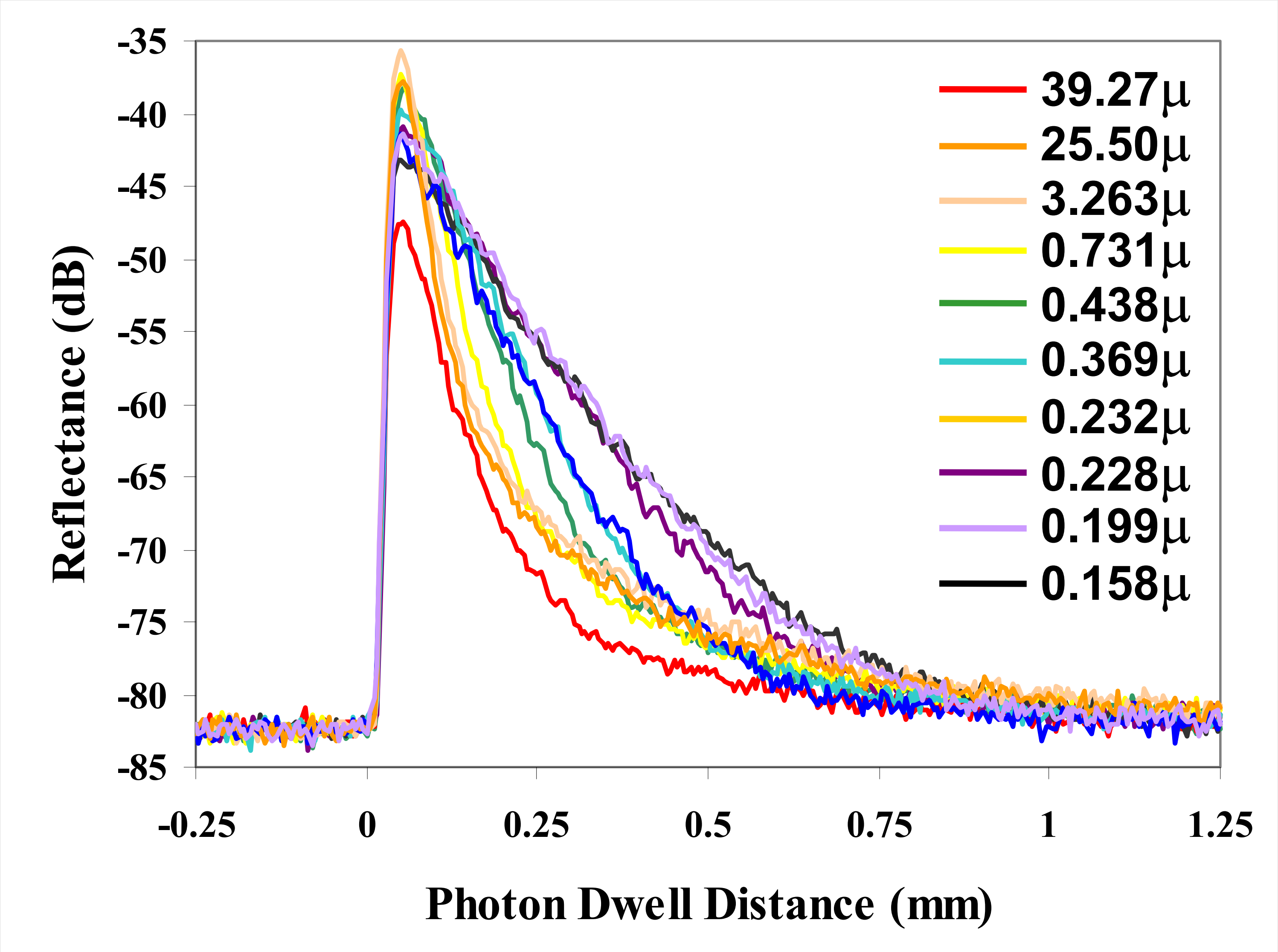
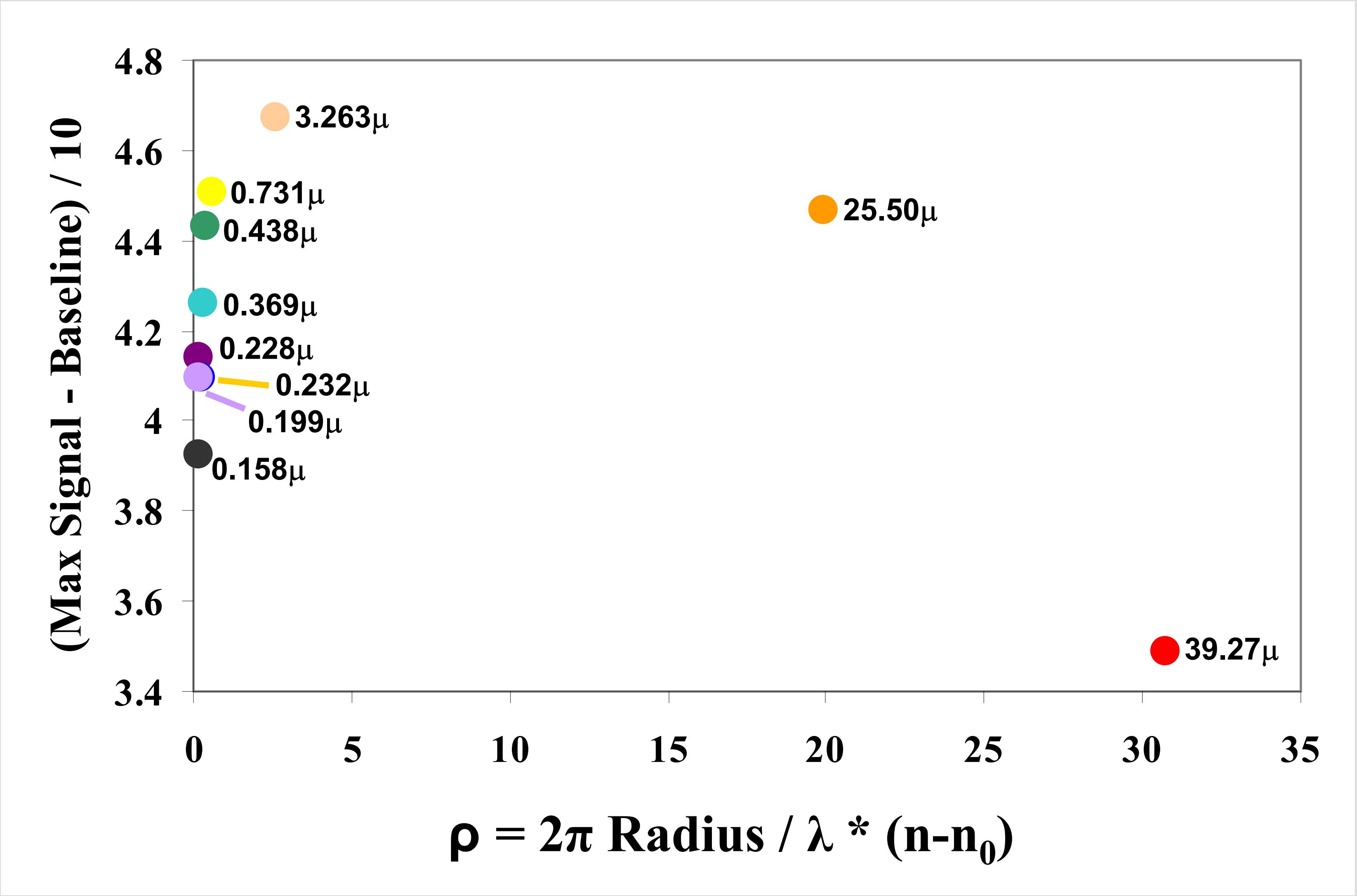
Attrition Mill Monitoring with OLCR



OLCR Multiple Scattering Theory



Publications

Optical Publications

•Brodsky, A. and L. Burgess. “Theoretical Study of the Coherent Backscattering of Light by Disordered Media,” *J. Modern Physics B*, (2003 *in press*).

• Randall, S. L., A. M. Brodsky, L. W. Burgess, and R. L. Green. “Optical Low-Coherence Reflectometry for Nondestructive Process Measurements,” 29th Annual Review of Progress in Quantitative Nondestructive Evaluation (QNDE) - 2003.

Ultrasonic Publications

• P. D. Panetta, B. Tucker R. A. Pappas, and S. Ahmed, “Characterization of Solid Liquid Suspensions Utilizing Ultrasonic Measurements” IEEE Instrumentation and Measurement Technology Conference (in press)

• P. D. Panetta, B. J. Tucker, R. A. Pappas, and S. Ahmed,” Characterization of solid liquid suspension utilizing ultrasonic measurements”, 29th Annual Review of Progress in Quantitative Nondestructive Evaluation -2003

Conclusions

- Developing the science base to characterize HLW to benefit the cleanup program - tank and pipeline applications
  - Particle size
  - High Concentration
  - Agglomeration
  - Gelation

- Benefits of combined measurements
  - Complimentary information
  - Large particle size range
  - Rapid measurements
  - On-line configurations
  - Large Penetration
  - High concentration

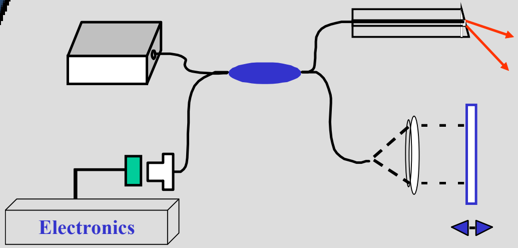
Path forward

- Continued Methodology Development
- Continue Theoretical development
- Regime characterization -- Monodispersed
- Further bimodal and polydispersed studies
  - Experimentation and Theory, Phase Transitions
- Dynamic Systems
  - Agglomeration and gelation of simulant
- Synergistic measurements
- Continue discussions with sites (SRS, Hanford, etc.)

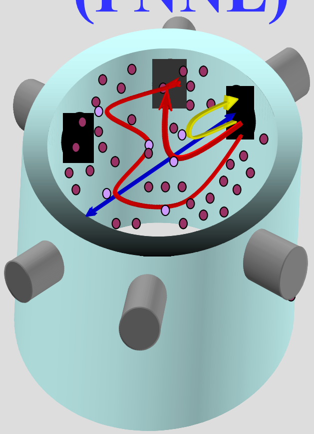
Synergism between EMSP Projects for HLW Clean up

Burgess – Panetta EMSP

OLCR (UW)



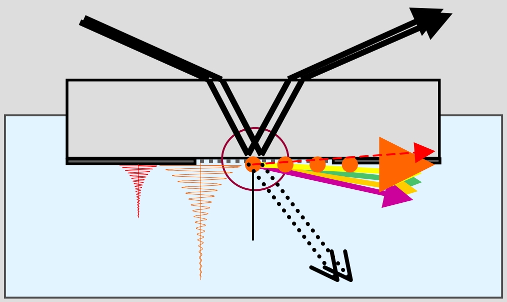
Ultrasonics (PNNL)



HLW Waste Characterization

Greenwood – Burgess EMSP

GLRS (UW)



UDGS (PNNL)