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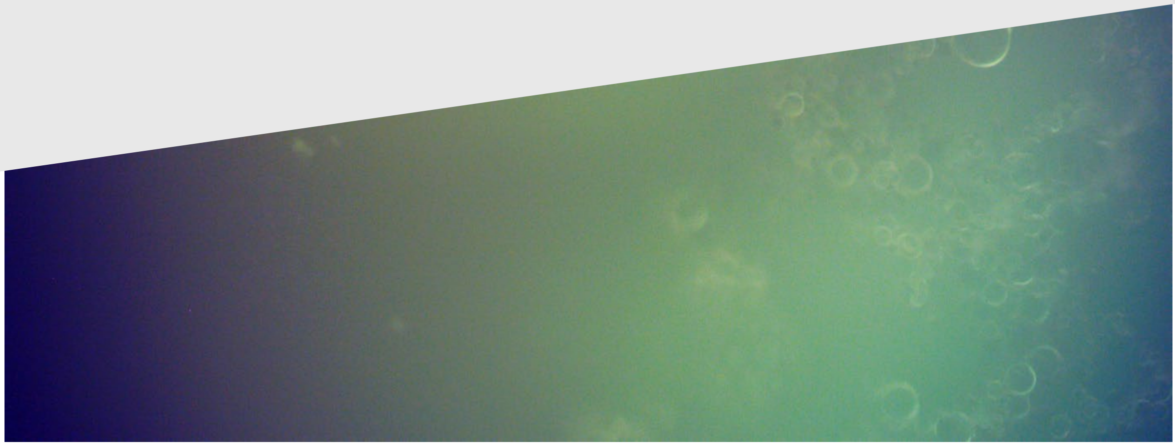

PNNL-37537

Imprinted Micelle Integration into a Commercial Platform

Progress Report

September 2024

Erin Fuller
Sarah Miley
Amy Sims
Caleb Allen
Samuel Morrison
Lance Hubbard

U.S. DEPARTMENT OF ENERGY

Prepared for the U.S. Department of Energy
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Richland, Washington 99354

Abstract

PNNL has successfully integrated a commercial aerosol detector and the imprinted micelle technology. The integrated systems have been shown to have a limit of detection between 33-47 particles with several options for data analysis presented that vary on computational requirements. It is possible to integrate these systems and receive response data on the second time scale. While more work is needed, these technologies are compatible, which opens up a large field of air sampling looking for specific contaminates.

Acknowledgments

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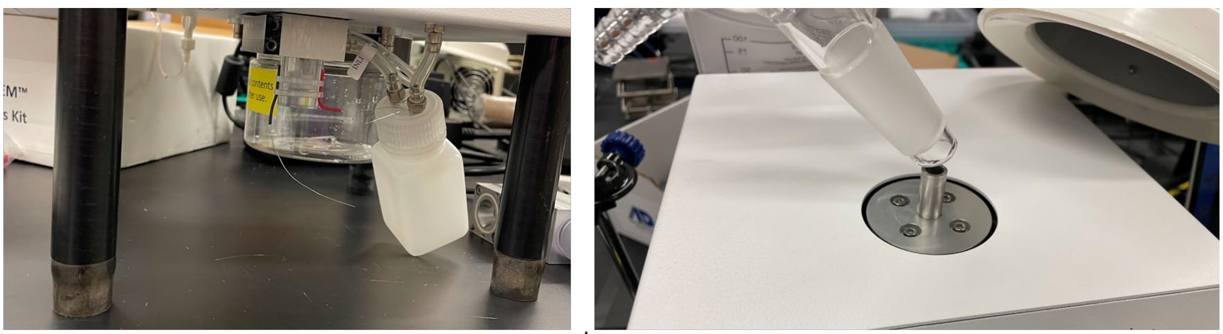
1.0 Introduction

PNNL has been tasked with the integration of a commercial aerosol sampler with the imprinted micelle technology. The work proceeded by verifying that the micelle synthesis was transferable between staff, as a new staff member created the micelles from the original notes. Then the functionality of the micelles was verified 1st optically and then electronically. With this, the platform was ready for integration into a commercial system.

The work proceeded with the creation of the custom particle collection cup/electrochemical cell which was shown attached instead of the vendor-supplied collection cup (Figure 1a). The platinum wires were hooked to the potentiostat assembly detailed in published literature (Hubbard et al., 2022).



a b

Figure 1. a) Photograph of the electrochemical cell attached to the bottom of the detector. b) Photograph of the outlet of the particle generator and the inlet of the aerosol collection unit, with building exhaust shown in the background.

Measurements occurred by direct injection of 10 microliters of particle suspensions into the collection unit. As well a particle generation setup producing between 1-10 million particles per minute was used to provide particles to the collector, Figure 1b. The collection of particles ensued, with collection into the electrochemical cell containing the imprinted micelles.

Several types of suspensions were tested. Deionized water was used as the overall control, with a mixture of 10%vol. of the micelle suspension in the cell. To this suspension of imprinted micelles in the integrated electrochemical cell and aerosol sampler 3 different types of particles were introduced:

1. The particles from the laboratory environment
2. The targeted 100 nm dia. spherical silica particles covered in vitamin C.
3. A spoof particle designed to mimic the target particle closely, 100 nm dia. spherical titania particles covered in fluorescein (Yellow #7)

By testing against a control and a very similar particle the detection limit of the integrated aerosol and micelle technology can be evaluated.

2.0 Methods

Please see (Hubbard et al., 2022; Sars-cov- et al., 2022) for synthesis methods of the micelles and various particles as well as a general overview of the functionality of the technology.

3.0 Results

3.1 Optical and Electronic

Optical imaging was used to identify the functionality of the micelles. As seen in Figure 2a, the micelles are mobile in their liquid environment. Upon the introduction of agglomerates of targeted particles, they mix and interact, Figure 2b. Lastly, after several seconds of interaction stable polymer waste begins to build up on the slide, indicative of the destruction of the micelles, Figure 2c. Electrically, these micelles were determined to give 1.08+-0.26 nA of signal in the setup (Hubbard et al., 2022) which is on par with the functionality seen in previous work. It should be noted that 3-year-old micelles that had been stored in a refrigerator produced 0.72+-0.19 nA, giving some indication of the shelf-life of the technology.

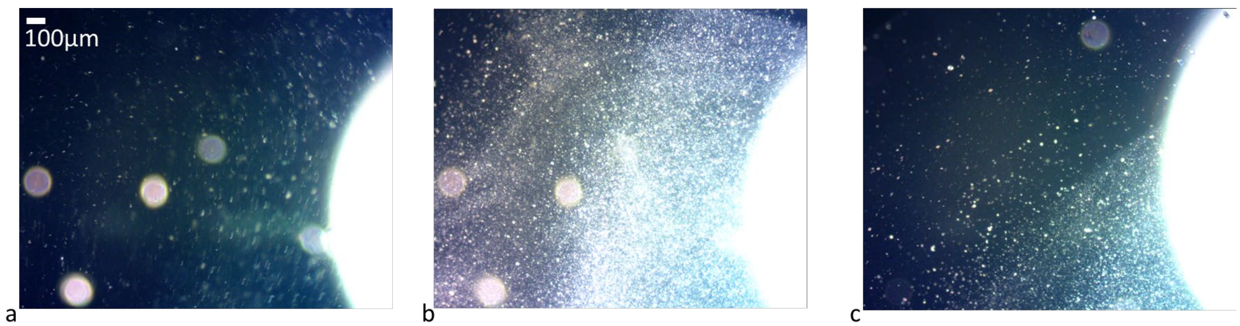


Figure 2. a) Darkfield micrograph, 500X, of micelles in deionized water. b) Darkfield micrograph, 500X, of the micelles mixed with the target particles. c) Darkfield micrograph, 500X, of the residual polymer/particle mixture adhered to the glass slide.

The raw data shows that similar to previous work, the micelles report groups of interactions, as shown in Figure 3a. When similar particles, are introduced the overall current is diminished, but occasional interactions do occur, Figure 3b. As a quick detection method, the standard deviation of the raw signal data as well as the 95% level calculation can be used to determine if response data warrants further investigation, as shown in Figure 3c.

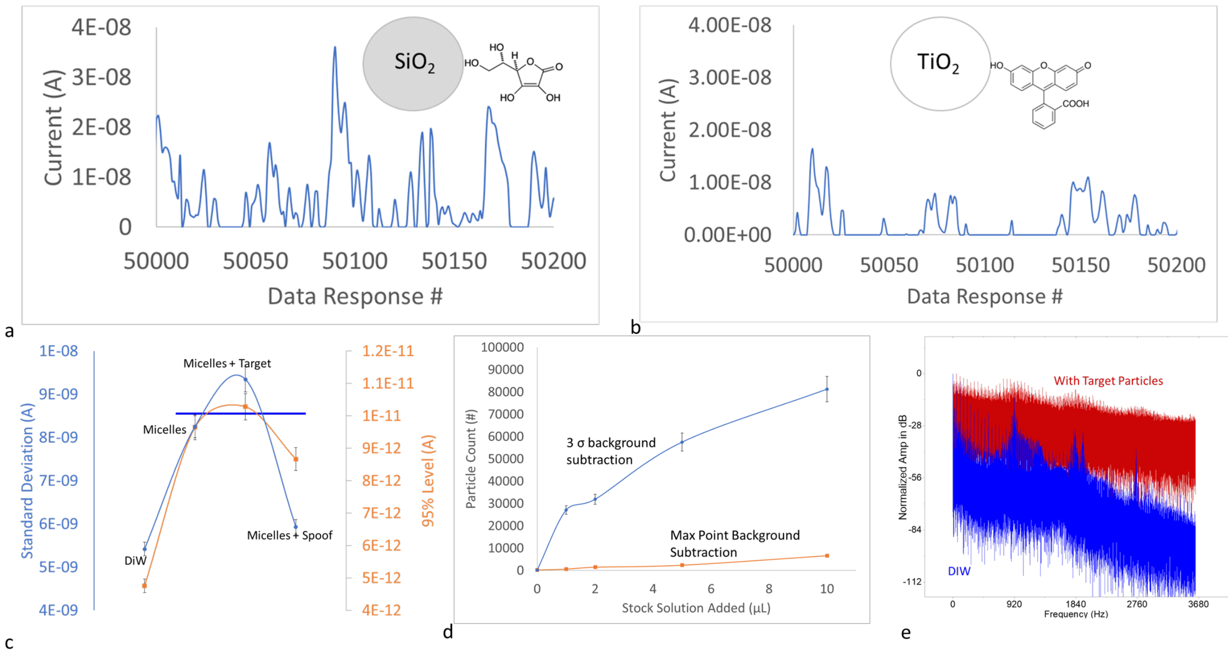


Figure 3. a) Two milliseconds of micelle electrical response data when exposed to the target particle. A diagram of the 100 nm dia. silica target particle is inset with ligand shown (not to scale). b) Two milliseconds of micelle electrical response data when exposed to a very similar particle. A diagram of the 100 nm dia. titania 'spoof' particle is inset with ligand shown (not to scale). c) Graph of the results of the quick detection method showing that the mixture of the micelle and target particles is the only one with the verified signal. d) Linear addition graph plotted with detected particles. e) Fast Fourier transform, of the background signal and the mixture of target particles and micelles.

If micelle-particle interactions are present, response curves can be used to determine the number of interactions based on applying various background subtraction techniques, as shown in Figure 3d.

Lastly, in post-processing Fast Fourier Transforms analysis can be used to ensure that the stochastic response of the micelle-particle interactions is riding on top of any cyclic noise in the system, as shown in Figure 3e. It should be verified that any motor noise from the collection hardware should be broadened but still represented by the micelle signal. This is a switch from the single-virus sensitivity shown in previous work, (Hubbard et al., 2022). With the signal verified as originating from samples collected by the aerosol collector and interacting the the micelles in the attached electrochemical cell, the limit of detection can be calculated.

The limit of detection can be calculated by several methods yielding results:

1. 33-47 particles
2. 0-137 particles
3. 0-1398 particles
4. 100000-110000 particles

The results presented above represent a diminishing amount of required computational power. Thus, there exist several methods to evaluate the data which can be tailored to the needs of the end user. While further work is needed, these methodologies can provide a basis for the development of automated control platforms with integrated data assurance in the form of feedback loops.

4.0 Conclusions

There has been a successful integration of a commercial aerosol detector and the imprinted micelle technology, developed at PNNL. The integrated systems have been shown to have a limit of detection between 33-47 particles with several options for data analysis presented that vary on computational requirements. It is possible to integrate these systems and receive response data on the second time scale. While more work is needed, these technologies are compatible, which opens up a large field of air sampling looking for specific contaminates.

5.0 References

Hubbard, L. R., Allen, C. J., Sims, A. C., Engbrecht, K. M., Hara, M. J. O., & Johnson, J. C. (2022). Detection of SARS - COV - 2 by functionally imprinted micelles. *MRS Communications*, *XX*(xx), 1–8. https://doi.org/10.1557/s43579-022-00242-0

Sars-cov-, T. R., Hubbard, L., & Morrison, S. (2022). *COVID-causing virus in air detected with high-tech bubbles*. *October*, 1–2. https://doi.org/10.1557/s43579-022-00242-0

Pacific Northwest National Laboratory

902 Battelle Boulevard
P.O. Box 999
Richland, WA 99354

1-888-375-PNNL (7665)

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