

Fundamental Aspects of the Science and Engineering of Oil Spill Dispersant Systems: An Overview of Recent Research Activities of the Gulf of Mexico Research Initiative-funded CMEDS Project

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ABSTRACT 285695:

Upon consideration of dispersant-related research, both before and after the Macondo Well oil release, it can be divided into two general categories: (1) the fundamentals of how dispersants work and the effects that may result from their use (e.g., physicochemical and transport characteristics of drops, bubbles, hydrates, surfactants), and (2) an applied focus that has emphasized the design of new dispersants or an enhancement of the performance of those products that are currently available.

While there is an extensive amount of data relating to dispersants, a main focus has been on the demonstration of their effectiveness in bench tests and examination of the toxicity of dispersants and dispersed oil. As a result, there is a need for an enhanced understanding of dispersant and dispersed oil thermodynamics and their fate and transport, with a goal to translate the science and engineering to the development of new, effective dispersant systems. The focus of the work to be discussed addresses the following areas:

Formation of small oil droplets: Widely dispersed stable oil droplets in the water column are easily accessible to microbes and therefore highly susceptible to degradation. It is important therefore, to understand the fundamental mechanisms of oil breakup and colloidal stabilization in order to develop new and effective dispersants.

Dispersant-related processes under deep sea conditions: Current dispersants have been developed for surface spills. The efficacy of such formulations when applied at the high pressures and low temperatures representative of deep ocean release has not been systematically studied. Because of concomitant gas release at the discharge point, and the pressures involved, the liquid droplet is essentially a gas-expanded liquid which could behave quite differently when treated with dispersant components depending upon how they partition at the phase interfaces, i.e., gas/water, gas/oil, oil/water.

Fluid mechanics of stabilized oil droplets: Droplet transport, as influenced by all thermodynamic variables of relevance under deep sea conditions, is being studied.

Droplet interactions with solid particulates: A better understanding of these processes, either in marine sediments or in the water column, will help predict the environmental fate of the droplets.

Development of alternative dispersants: Based on the knowledge gained with respect to the fundamentals, a key goal is the systematic translation of that understanding to the development of new and improved materials.

This paper summarizes recent work of a collaborative research effort involving investigators from 22 universities, with particular emphasis on increasing the understanding of the science and engineering of oil spill dispersants.

INTRODUCTION:**The Gulf of Mexico Research Initiative**

On May 24, 2010, shortly after the Macondo Well release, BP announced a commitment of up to \$500 million over 10 years to fund an independent research program designed to study the impact of the oil spill and its associated response on the environment and public health in the Gulf of Mexico. Following initial funding of five studies to establish critical baseline data as the foundation for subsequent research, the Gulf of Mexico Research Initiative (GoMRI) announced additional funding of eight research consortia on August 30, 2011 over three years to study the effects of the oil spill on the Gulf of Mexico. All lead institutions for the consortia are located in Gulf States.

The ultimate goal of GoMRI is to improve society's ability to understand, respond to and mitigate the impacts of petroleum pollution and related stressors of the marine and coastal ecosystems, with an emphasis on conditions found in the Gulf of Mexico (GoMRI, 2011). The main research themes of the effort include the physical distribution, dispersion and dilution of petroleum, the chemical evolution of petroleum/dispersant systems, and the fundamental science and technology development for remediation associated with oil spills and gas releases.

DISCUSSION:

The Consortium for the Molecular Engineering of Dispersant Systems (CMEDS) was one of the eight groups funded in 2011. The consortium involves eight institutions from the five Gulf of Mexico states with 22 participants, and 14 institutions from the other states with 20 participants. The Consortium has an Advisory Board with distinguished representatives from industry and the federal laboratories.

The Consortium reflects a focused effort to understand the role of dispersants and related chemical compounds in mitigating the effects of deep-sea hydrocarbon releases, and in designing the next generation of dispersants. From the thematic perspective of GOMRI, the Consortium is fully invested in the topics of Research Theme 4 (fundamental science and technology development for remediation associated with oil spills and gas releases). Key focus areas include understanding the role of dispersants in mitigating the effects of oil releases and designing the next generation of dispersants. The main questions that are being considered by the research group are:

- What are the fundamental factors that rapidly produce droplets that will be sustained in the water column?
- How can dispersant formulations be designed to minimize oil-water interfacial tension and stabilize droplet coalescence?
- How can an understanding of the chemistry and physics of droplet behavior lead to the design of more effective environmentally benign dispersants?
- What are fate and transport characteristics of dispersed oil droplets?

In keeping with GoMRI's expectations, the CMEDS principal investigators are comprised of a mix of established researchers and promising junior faculty members. Most

investigators supervise graduate students working toward a doctoral degree. In addition to its research mission, the consortium has a strong education mission to train students in advanced science and technology related to the mitigation of oil spills and there is a strong expectation that Ph. D. dissertations will result from the research investment, a very positive potential outcome aimed at the production of a generation of researchers with an awareness of oil spill response-related issues. This is intended to establish and nurture the careers of the researchers themselves and to provide opportunities for students to develop fruitful scientific careers. Additional research-related aspects of this include the goals to:

- Create fundamental science that can be extended beyond oil spill response and mitigation
- Develop technology that is translatable to industrial practice, with a focus on long-range, high impact work
- Provide opportunities for the establishment of long-lasting collaborations

As an example of broad collaboration, a recent publication, “Attachment of a Hydrophobically Modified Biopolymer at the Oil-Water Interface in the Treatment of Oil Spills,” was the result of work by scientists at Tulane University, University of Maryland, University of Louisiana and the University of Rhode Island (Venkataraman, 2013).

Additionally, CMEDS has had regular interactions with scientists outside of academia, either by teleconferencing, face-to-face meetings on a specific topic, or providing opportunities to meet during regular scientific conferences. All of these activities have been facilitated by having an external advisory board which has a large percentage of its members from industry and the national laboratories.

Research

CMEDS takes the long range perspective of fundamental research that is use-inspired in the development of new technologies for oil spill remediation. The following paragraphs outline research accomplishments that are related to resolving specific problems.

The current use of dispersants is almost entirely focused on COREXIT 9500™ which was extensively used in the Macondo Well release. While COREXIT™ 9500 contains nonionic sugar based surfactants such as the Tween™ and Span™ products, widely considered to be nontoxic due to their biodegradable sugar moieties, it also contains a double-tailed anionic surfactant, dioctylsodium sulphosuccinate (or DOSS), an FDA approved compound and generally considered to be safe for human contact and consumption in moderate quantities. Additionally, COREXIT™ 9500 contains solvents, the use of which may be able to be reduced in alternative, new technology-based formulations. From a technological perspective, while COREXIT™ 9500 significantly lowers the oil-water interfacial tension and is an effective dispersant, its use in deep-sea thermodynamic conditions where high pressures and low water temperatures exist is not definitively established. Newer concepts in colloid and surface chemistry and in polymer science and particulate technology may be exploited effectively to improve dispersant design.

CMEDS research is therefore focused on finding greener alternatives for COREXIT™ 9500 that involve new concepts taken from the field of colloid and interfacial science. In addition

to using small molecule surfactant formulations, researchers at CMEDS have made significant scientific and technological advancements in the use of novel materials including polymers and submicron particles to form and stabilize oil droplets, and gels to immobilize surface oil. We describe some of these new developments and research accomplishments below.

CMEDS researchers have shown that particle stabilized emulsions (Pickering emulsions) can be effectively deployed to stabilize oil droplets against coalescence (He, 2013; Saha, 2013; Katepalli, 2013). The fundamental science of such particle stabilized emulsions are being explored with extensions to novel particle morphologies (flat sheets of clay and graphene, hollow particles, functionalized particles with strong surface affinities etc.) to produce highly efficient amphiphilic particles that disperse oil at low concentrations (Yoon, 2013; Sarkar, 2013A; Sarkar 2013B). This work is of particular relevance to observations following the Macondo Well release of oil droplets with attached sediment particulates. The origin, fate and transport of these oil mineral aggregates (OMA) are of considerable interest. CMEDS researchers are studying these systems to seek analogies to Pickering emulsion formation where the energetics of particles at oil-water interfaces can be clearly understood through considerations of amphiphilicity. The designed stabilization of such particle based emulsions may obviate the need for surfactant based emulsifiers, especially at deep sea conditions of high turbulence. Particle based emulsion systems also provide opportunities to control the dynamics of oil droplets through morphology and density adjustments of the particles. Interfacially adherent particles with encapsulated magnetic materials or adsorbed fluorescent materials can be used in the development of imaging technologies to locate oil through MRI imaging and fluorescence detection. Such technologies may be relevant to imaging surface spills at night and in the imaging of deep sea spills.

The use of polymers to stabilize the oil-water interface is also being explored (Venkataraman, 2013). These include biopolymers with pendant side chains that anchor to the oil-water interface, naturally derived polymers with interfacial activity, and synthetic amphiphilic polymers. There is a significant rationale behind the use of polymers that adsorb and stabilize oil-water interfaces. Because there are so many amphiphilic moieties on appropriately designed polymer chains, binding to the oil-water interface may be considered to be virtually irreversible through the multiplicity of contact points of the polymer at the interface. The self-assembly of such polymers could lead to the formation of essentially uni-molecular micelles with extremely high partition coefficients, as opposed to surfactants which eventually desorb from interfaces when bulk solution concentrations decrease below the critical micelle concentration. Whether-or-not this is a significant advantage remains to be explored.

In a similar vein, CMEDS researchers have found new classes of siloxane and sugar based organogelators (Mallia, 2013). These small molecules self-assemble to fibrils using non covalent interactions and lead to the selective gelation of oil, preventing oil slick spreading (Mallia, 2013; Balachandran, 2013). Concepts of such phase selective gelation can be effectively employed in exploiting the Marangoni effect to herd oil for controlled burns and skimming (Venkataraman, 2013). In this technology, environmentally benign surfactants are first used to create surface convection through gradients of surface tension and thus corral oil. Gelling agents are then applied to the corralled oil to keep the oil stationary for skimming (smaller spills) and burning (larger offshore spills).

CMEDS researchers are also involved in the development of green molecular dispersants, using protein based surfactants (the class of hydrophobins) and phospholipids. The self-assembly of these materials and their ability lowering the oil-water interfacial tension are being explored. Results on these materials will be published soon. Along such directions, the generation of self-assembling gels of surfactants allows reduction of the organic solvents used in current technologies, and may also facilitate integration of particle based systems.

The technological directions of CMEDS are strongly complemented by fundamental research. Research has led to the development of two novel experimental techniques to measure fast dynamics of surfactant adsorption to the oil-water interface, using a microtensiometer and fluorescence quenching (Reichert, 2013). A remarkable and rather counterintuitive finding is that the nonionic surfactant Tween™ appears to have a level of irreversibility associated with its adsorption at the oil-water interface. A third novel technique that is being developed is the use of cantilever deflections in atomic force microscopy to measure the forces restraining particles to the oil-water interfaces. More traditional but highly sophisticated methods using cryo-electron microscopy to visualize particles at interfaces are being employed. CMEDS is also undertaking basic studies of the phase behavior of COREXIT™ 9500 components to understand the relationship between interfacial tension and phase composition.

CMEDS has a distinctive focus on chemical computation and theory to guide the experimental work. Molecular dynamics simulations of surfactants at interfaces guide the understanding of dynamic interfacial tension and the work is being extended to understanding polymer conformation at the oil-water interface. Recent accomplishments include the finding that Tween™ adsorbs strongly on the interface, and that critical micelle concentrations go through a maximum with pressure (Meng, 2013). This understanding can serve to provide additional insight as dispersant development strategies are considered, especially with respect to the conditions encountered in the event of a deep water sub-sea release.

Ongoing fate and transport studies within CMEDS complement the new technologies that are being developed. For example, CMEDS researchers have studied partitioning of hydrocarbons between the bulk phase and aerosol droplets generated through wave action and the transport of non-volatile oil fractions and dispersants to the aerosol phase is noted (Liyana-Arachchi, 2013A). The research is extended to cases with different dispersants at the air-water and the oil-water interface (Liyana-Arachchi, 2013B). CMEDS is also conducting studies on the effects of mass transfer on the hydrodynamics of oil droplets and bubbles. The implications of oil adsorption on plant materials are studied through the development of leaf mimics to guide the development of new dispersants that mitigate the impact of oil on marshland. Similarly, the bacterial movements towards oil/dispersants within sediment pore spaces are being studied to understand oil/dispersant/bacterial interactions. CMEDS research has led to the development of technologies that have ramifications beyond oil spill remediation and mitigation. The use of particles to stabilize emulsions has led to systems with potential applications in imaging and detection using electromagnetic and magnetic resonance imaging (MRI) based techniques (Bagaria, 2013A; Bagaria, 2013B; Bagaria, 2013C). These technologies have implications to oil mobilization in reservoirs and the imaging of oil in reservoir settings. The stabilization of particles and polymers at interfaces are also leading to new technologies in energy related materials for the development of fuel cell and lithium ion batteries. Development of new micro-

fabrication and microfluidic technologies to understand molecular scale dynamics is another potential outcome of the work. Thus, the long range science associated with CMEDS will have implications in a variety of new technologies.

Future Proposed Research

For the next phase of CMEDS research, the vision is to focus along the following three topical areas:

- The molecular and nano-scale design of new dispersant systems – experiment and computation.
- Understanding dispersants and their impacts at the molecular and nano-scale level – experiment and computation.
- Translational dispersant science – from the molecular scale to practice.

Community Outreach

While much of the CMEDS focus is on science and engineering, a significant component that is stressed by GoMRI revolves around an outreach mission and conducting meaningful outreach that will benefit local communities. For CMEDS, the emphasis has been on communicating and providing information on the role of dispersants in oil spills. With this in mind, the CMEDS Outreach Initiative has several components designed to enhance the dissemination of knowledge to the academic community as well as to the general population. For example, a joint effort between Tulane and LSU CMEDS faculty and students has resulted in the pairing of 21 undergraduate students with faculty mentors and their engagement in 10 weeks of focused research. Each summer culminated with a research symposium in which each intern delivered a presentation of his/her research. The symposiums were held in conjunction with an NSF Undergraduate Research Mentoring (NSF-URM) initiative – Enhancing Diversity in Environmental Biology (Figure 2).¹ In addition to the undergraduate interns, a local middle-high school science teacher was involved in the outreach initiative. A one-day summer science academy was held for a number of local elementary students where the students were engaged in a “round-robin” rotation of age appropriate, scientific experiments. Outreach activities were expanded during the academic year when CMEDS interns continued science-based discussions with high school students with a focus on academic careers, Tulane University, the Macondo Well oil release, and individual research projects.

Efforts to reach out to more community members directly affected by the Macondo Well release were realized when CMEDS partnered with Tulane’s School of Public Health and Tropical Medicine (SPHTM) and hosted a forum in Houma, LA. In addition to the CMEDS steering committee chair, the forum was facilitated by a toxicologist and a SPHTM outreach coordinator. The forum, entitled “Is it safe? Oil spill dispersants and frequently asked questions,” provided the Vietnamese fishing/shrimping community with an opportunity to ask questions

¹ The goal of the Undergraduate Research and Mentoring in the Biological Sciences (URM) program is to increase the number and diversity of individuals pursuing graduate studies in all areas of biological research supported by the NSF Directorate for Biological Sciences. Support is provided to academic institutions to establish innovative programs to engage undergraduates in a year-round research and mentoring activity.

from researchers. Questions included those related to long term health care issues related to the spill as well as those surrounding the stability of the seafood industry post spill.

Since its creation, other CMEDS investigators have conducted distinct individual outreach activities. Each investigator has a portion of their budget allocated to facilitate an undergraduate research experience in addition to other unique outreach opportunities which are often leveraged through other grant support. Examples include:

- An integrated science and education program through the Tampa Museum of Science and Industry
- A service learning course at the New Orleans Charter Science and Mathematics High School
- A workshop on oil spill dispersants during a “Tech Savvy” conference organized by the American Association of University Women (AAUW, 2013)²
- An “Art-Meets-Science” Program at Glasgow Middle School in Baton Rouge, LA

A supplemental outreach program is planned for 2014 which will emphasize the public dissemination of research through journalist training. A representative of the Metcalf Institute for Marine & Environmental Reporting will work with CMEDS principal investigators to develop and implement an oil spill science workshop for journalists focused on the science and technology of deep-sea dispersants at the 2014 Gulf of Mexico Oil Spill & Ecosystem Science Conference in Mobile, Alabama. The purpose is to summarize and clarify the latest scientific findings from the conference, and to support improved coverage of oil spills and their effects (Metcalf, 2014).³

SUMMARY:

Through the support of the Gulf of Mexico Research Initiative (GoMRI) the Consortium for the Molecular Engineering of Dispersant Systems (CMEDS) has focused the efforts of academic researchers from a number of universities on the topic of oil spill dispersants. Following the Macondo Well oil spill, a large amount of attention has been paid to these materials and it was recognized that there was an opportunity to explore new science and engineering advances in the area of oil spill response and to communicate them to the public and the press.

Significant novel approaches have been undertaken by the researchers of the CMEDS group and the findings are in the early stages of publication. Industrial scientists have been

² Tech Savvy is a daylong science, technology, engineering, and math (STEM) career conference designed to attract girls in sixth through ninth grade to these fields and to inform families about STEM education and careers.

³ The Institute is part of the University of Rhode Island (URI) Office of Marine Programs, based at the URI Graduate School of Oceanography

actively involved in discussions about a number of these research efforts with an eye toward evaluating their applicability in real world oil spill scenarios.

Additionally, the consortium has focused efforts on a substantial outreach program aimed at improving the scientific literacy of those who may be affected by or otherwise involved with a possible oil spill and its aftermath. An improved public understanding of oil spill responses potentially involving the possible use of dispersants has been a key part of CMEDS through local educational, community, and journalist-related efforts.

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FIGURES:

Figure 1 CMEDS Participant Universities

Tulane Univ.	Louisiana State Univ.	Univ. Texas at Austin
Univ. Rhode Island	Arizona State Univ.	Auburn Univ.
Brown Univ.	Carnegie Mellon	City Univ., NY.
Florida Intl. Univ.	Georgetown Univ.	Univ. Maryland
Univ. Michigan	Univ. Minnesota	North Carolina State
Princeton Univ.	Univ. Colorado	Univ. Mass, Amherst
Univ. South Florida	Univ. Buffalo, SUNY	Univ. Houston
Univ. Southern Mississippi	http://dispersant.tulane.edu website	

Figure 2 CMEDS 2012 Summer Interns Following the Summer Research Academy Symposium Held in Conjunction with the NSF-URM Initiative



2014 INTERNATIONAL OIL SPILL CONFERENCE

Figure 3 CMEDS Summer Interns and 2013 Summer Campers and Staff Following the Summer Science Outreach Day and Tulane University

