DOE GRANT FINAL REPORT

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"Cavitational Hydrothermal Oxidation: A New Remediation Process"

PI Name: **Kenneth S. Suslick**

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PI Institution: **University of Illinois at Urbana-Champaign**

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PI Address: **Department of Chemistry**
University of Illinois at Urbana-Champaign
600 S. Mathews Av.
Urbana, IL 61801

tel: 217-333-2794
fax: 217-333-2685
email: ksuslick@uiuc.edu

I certify that to the best of my knowledge (1) the statements herein (excluding scientific hypotheses and scientific opinions) are true and complete, and (2) the text and graphics in this report as well as any accompanying publications or other documents, unless otherwise indicated, are the original work of the signatories or individuals working under their supervision.

PI Signature:

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Kenneth S. Suslick

1. Summary

During the past year, we have continued to make substantial scientific progress on our understanding of cavitation phenomena in aqueous media and applications of cavitation to remediation processes. Our efforts have focused on three separate areas: sonoluminescence as a probe of conditions created during cavitational collapse in aqueous media, the use of cavitation for remediation of contaminated water, and an addition of the use of ultrasound in the synthesis of novel heterogeneous catalysts for hydrodehalogenation of halocarbons under mild conditions.

In order to gain further understanding of the conditions present during cavitation, we have continued our studies of sonoluminescence. We made a breakthrough in the use of emission spectroscopy for temperature and pressure measurement of cavitation events, which we expect to publish shortly. We were able to measure *for the first time* the temperature of cavitation *in water* during multi-bubble cavitation in the presence of aromatic hydrocarbons. The emission from excited states of C_2 in water gives temperatures that are consistent with adiabatic compressional heating, with maximum temperatures of 4300 K. Our prior measurements of cavitation temperatures in low vapor pressure nonaqueous media gave somewhat higher temperatures of 5000 K. This work lays permanently to rest exotic mechanisms for cavitational chemistry, at least for cavitation fields.

In a new avenue of research, we explored the generation of cavitation by hydrodynamic techniques and demonstrated for the first time that chemical reactions occur during turbulent flow of water and of water containing chlorocarbons. This work received substantial publicity. While the chemical effects of *acoustic* cavitation (i.e., sonochemistry and sonoluminescence) have been extensively investigated during recent years, little is known about the chemical consequences of *hydrodynamic* cavitation created during turbulent flow of liquids. Hydrodynamic cavitation is observed when large pressure differentials are generated within a turbulent liquid flow and is accompanied by a number of physical effects, erosion being most notable from a technological viewpoint. It has the potential advantage of facile scale-up: there already exist commercial high pressure flow mixing units capable of generating hydrodynamic cavitation with flows of 50 gpm (Microfluidics, Inc., Newton, MA). However, prior reports of hydrodynamically induced chemistry or luminescence and direct comparisons to sonochemistry or sonoluminescence have been extremely limited. The oxidation of iodide to triiodide in aqueous solutions containing halocarbons (e.g., $CHCl_3$ or CCl_4) under hydrodynamic cavitation conditions was examined in order to determine the origin of this hydrodynamic chemical reaction. This reaction also serves as a model for remediation of halocarbon contaminated water using hydrodynamic cavitation, a previously unexplored approach to remediation. The effects of several experimental variables on the I_3^- production rate were investigated. Increasing the reaction temperature inhibits the I_3^- production due to increased vapor pressure, which leads to reduced efficacy during cavitational collapse; the rate decreases exponentially with vapor pressure. The production of I_3^- is sensitive to the nature of the dissolved gas and decreases exponentially with increasing gas thermal conductivity. Thus, the chemistry observed and its response to external parameters is remarkably similar to ultrasonically induced cavitation, albeit the sonochemical rates are significantly higher at least with our current experimental configurations.

2. Output From The Research

Five major reviews and eight peer reviewed scientific publications.

Scientific Reviews:

1. Suslick, K. S.; Crum, L. A. "Sonochemistry and Sonoluminescence," in *Encyclopedia of Acoustics*; Crocker, M. J., ed.; Wiley-Interscience: New York, 1997; vol. 1, ch. 26, pp. 271-282.
2. Suslick, K. S.; Crum, L. A. "Sonochemistry and Sonoluminescence" in *Handbook of Acoustics*; Crocker, M. J., ed.; Wiley-Interscience: New York, 1998; pp. 243-253.
3. Suslick, K. S. "Sonochemistry" in *Kirk-Othmer Encyclopedia of Chemical Technology*; 4^{th} Ed. J. Wiley & Sons: New York, 1998, vol. 26, 517-541.
4. Suslick, K. S.; Matula, T. J. "Acoustic Cavitation, Sonochemistry, and Sonoluminescence" in *Wiley Encyclopedia of Electrical & Electronics Engineering*; Webster, J.G., ed.; Wiley-Interscience: New York, 1999, vol. 22, pp. 646-657.
5. Suslick, K. S.; Didenko, Y.; Fang, M. M.; Hyeon, T.; Kolbeck, K. J.; McNamara III, W. B.; Mdleleni, M. M.; Wong, M. "Acoustic Cavitation and Its Chemical Consequences" *Phil. Trans. Roy. Soc. London A*, **1999**, *357*, 335-353.

Publications (peer reviewed):

6. Suslick, K. S.; Mdleleni, M. M.; Ries, J. T. "Chemistry Induced by Hydrodynamic Cavitation" *J. Am. Chem. Soc.*, 1997, *119,* 9303-9304.
7. Suslick, K. S.; Didenko, Y. T.; McNamara III, W. B. "Conditions during Multi-Bubble Sonoluminescence" *Proc. 16^{th} Intl. Conf. Acoustics* Acoust. Soc. Am.: Seattle, 1998, pp. 2577-79.
8. Suslick, K. S.; McNamara III, W. B.; Didenko, Y. "Hot Spot Conditions During Multi-Bubble Cavitation" in *Sonochemistry and Sonoluminescence,* Crum, L. A.; Mason, T. J.; Reisse, J.; Suslick, K. S., eds. Kluwer Publishers: Dordrecht, Netherlands, 1999, pp. 191-204.
9. Suslick, K. S.; Didenko, Y.; Fang, M. M.; Hyeon, T.; Kolbeck, K. J.; McNamara, W. B. III; Mdleleni, M. M.; Wong, M. "Acoustic Cavitation and Its Chemical Consequences," *Phil. Trans. Roy. Soc. London A*, 1999, *357*, 335-353.
10. McNamara III, W. B.; Didenko, Y.; Suslick, K. S. "Hot Spot Conditions During Cavitation in Water" *J. Am. Chem. Soc.*, **1999**, *121*, 5817-5818.
11. Didenko, Y.; McNamara III, W. B.; Suslick, K. S. "The Temperature of Multi-Bubble Sonoluminescence in Water" *J. Phys. Chem. A,* **1999,** 103, 10783-10788.
12. McNamara III, W. B.; Didenko, Y.; Suslick, K. S. "Sonoluminescence Temperatures During Multibubble Cavitation" *Nature*, **1999**, *401*, 772-775.
13. Suslick, K. S.; McNamara III, W. B.; Didenko, Y. "Conditions During Multibubble Cavitation" " in *Nonlinear Acoustics at the Turn of the Millennium*, Lauterborn, W.; Kurz, T., eds. Amer. Inst. Physics: Melville, NY, 2000, pp. 463-466.

3. Education and Human Resource Development.

Yury Didenko
B.S., M.S., Moscow State University, 1970-75.
Ph.D., Moscow State University, 1985.
Head of Laboratory, Pacific Oceanological Institute, Vladivostok, Russia.
Research Associate, Univ. of Illinois, 1995-97.

William B. McNamara, III
B.S., University of California, Santa Barbara, 1992.
Ph.D., UIUC, 2000.

Millan M. Mdleleni
B.Sc., University of Fort Hare (S. Africa), 1987.
Ph.D., Univ. of California at Santa Barbara, 1995.
Postdoctoral Associate, Univ. of Illinois, 1995-97.