Chapter 7
Light Sensitized Disinfection with Fullerene

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ABSTRACT

Fullerene has drawn wide interest across many fields due to its favorable electronic and optical properties, which has spurred its use in a myriad of applications. One of the hallmark properties of fullerene is its ability to act as a photosensitizer and efficiently generate $^1O_2$*, a form of Reactive Oxygen Species (ROS), upon visible irradiation when dispersed in organic solvents. However, the application of fullerene in environmental systems has been somewhat limited due to fullerene's poor solubility in water, which causes individual fullerene molecules to aggregate and form large colloidal species, quenching much of fullerene's* $^1O_2$ *production. This is unfortunate given that* $^1O_2$ *provides many advantages as an oxidant compared to ROS produced from typical advanced oxidation processes, such as OH radicals, due to* $^1O_2$*'s greater chemical selectivity and its ability to remain unaffected by the presence of background water constituents, such as natural organic matter and carbonate. Hence, fullerene materials may hold great potential for the oxidation and disinfection of complex waters. Herein, we chronicle the advances that have been made to propel fullerene materials towards use in emerging water disinfection technologies. Two approaches to overcome fullerene aggregation and the subsequent loss of* $^1O_2$ *production in aqueous systems are herein outlined: 1) addition of hydrophilic functionality to fullerene's cage, creating highly photoactive colloidal fullerenes; and 2) covalent attachment of fullerene to solid supports, which physically prevents fullerene aggregation and allows efficient* $^1O_2$ *photo-generation. An emphasis is placed on the inactivation of MS2 bacteriophage, a model for human enteric viruses, highlighting the potential of fullerene materials for light-activated disinfection technologies.*

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1. INTRODUCTION

The development of novel materials for the inactivation of waterborne pathogens is in a critical need for much of the world today. Materials that, in response to visible light, can efficiently inactivate viruses and spore forming bacteria that survive in dry conditions would be transformative for many of the challenges in developing countries and in the context of bioterrorism defense. In particular, such materials would be highly useful for advancing solar disinfection (SODIS) and antimicrobial/biocidal coating technology. Semiconductor photocatalysts, such as the archetypical TiO_2, have been intensely pursued in hopes of realizing such disinfection technologies,(Min Cho, Chung, Choi, & Yoon, 2005a; Lonnen, Kilvington, Kehoe, Al-Touati, & McGuigan, 2005) but have been somewhat hindered by their inefficient visible light utilization. Hence, many researchers have put forth serious efforts on expanding TiO_2's visible absorption through various doping schemes,(Pelaez et al., 2012; Rehman, Ullah, Butt, & Gohar, 2009) while others have focused on pursing new small-band gap semiconductors such as WO_3,(Kim, Lee, & Choi, 2010; Zhu, Xu, Fu, Zhao, & Zhu, 2007) graphitic carbon nitride,(H. Wang et al., 2014; Xu, Wang, & Zhu, 2013) and CdS quantum dots(Bessekhouad, Robert, & Weber, 2004). Moving beyond conventional inorganic photocatalysts, photosensitizing organic dye molecules, which efficiently harvest visible light, have gained recent attention with a particular focus on buckminsterfullerenes.(Chae, Hotze, & Wiesner, 2009; Jaesang Lee et al., 2009) Buckminsterfullerenes, or simply fullerenes, and their functional derivatives have been proposed by researchers as effective antimicrobial agents, via photocatalytic production of singlet oxygen ($^1O_2$) and subsequent microbial inactivation (Liyi Huang et al., 2010; Jaesang Lee et al., 2009; Q. Li et al., 2008; George P. Tegos et al., 2005). In contrast with the ubiquitous semiconductor photocatalysts, fullerenes have the advantage of being able to be activated by visible light, especially with functionalization, and covalently anchored to a host material such as polymers. Photocatalysts used for disinfection should be recoverable/reusable, completely conserved (no escape into the environment), activated by visible light, and able to retain their catalytic properties over repeated use. Fullerene derivatives are very attractive as photocatalysts because they can potentially exhibit these essential characteristics when properly functionalized and covalently anchored onto a supporting structure.

This chapter chronicles the advances of fullerene (specifically C_{60} and C_{70}) science and technology related to their application as photocatalysts for disinfection. Fullerene photocatalysis is affected by several factors that are relevant to disinfection applications. Functionalization of the fullerene cage directly impacts the electronic structure of the chromophore, altering the energy levels and efficiencies of the intermediate excited states and transitions. Further, aggregation of fullerenes in the aqueous phase can severely diminish the photoactivity of the fullerenes. Functionalization and immobilization of fullerenes can both lessen the degree or effects of aggregation of fullerenes on their photochemistry. The utility of fullerenes applied as photocatalysts is directly proportional to the efficiency of their photosensitization of $^1O_2$. The use of fullerenes as disinfection agents, and the methods of enhancing their capabilities, are discussed here with case examples from our past publications.

2. BACKGROUND

Comprised only of carbon atoms, caged fullerene molecules represent the third allotrope of carbon besides graphite and diamond.(Kroto, Heath, O'Brien, Curl, & E, 1985) Among fullerenes (e.g., C_{70}, C_{76}, C_{78}, C_{84} and C_{90}), C_{60} has been the most prolific in fullerene research and application as C_{60} is available with high

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