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FOREWORD

Alison Butler: papers in celebration of her 2018 ACS Alfred Bader Award in Bioorganic or Bioinorganic Chemistry

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Foreword

This special issue celebrates the scientific achievements of Alison Butler on the occasion of her receiving the American Chemical Society Alfred Bader Award in Bioorganic or Bioinorganic Chemistry at the 2018 American Chemical Society Spring National Meeting, held in New Orleans. Butler's Bader Award recognizes her research group's many significant contributions elucidating the bioinorganic chemistry of the marine environment, including the chemistry of siderophores and vanadium haloperoxidases. The unusual transition metal composition of surface seawater captured her interest early on [1]. She has investigated the chemistry of vanadium haloperoxidases from marine seaweeds and shown that the stereoselective bromocyclization of terpenes catalyzed by these enzymes is important in the biosynthesis of halogenated marine natural products [2]. While vanadium is the second-most abundant transition metal ion in seawater, iron levels are extremely low over much of the world's ocean. Given the scarcity of iron in surface ocean water, she has been interested in the siderophores produced by marine bacteria from different ocean environments. She and her collaborators have identified new classes of large suites of fatty acyl siderophores [3] as well as photoactive Fe(III)-siderophores with α-hydroxycarboxylic acid ligands in the form of citrate and β-hydroxyaspartate [4]. Recently, she has moved to a targeted approach to siderophore discovery through genome mining [5] and begun investigating wet adhesive properties of catechol siderophores and analogs [6].

Alison Butler has also immersed herself in service to the Bioinorganic Community. She has served as President of the Society of Biological Inorganic Chemistry (SBIC) as well as a member of the SBIC Council. Recently, she was appointed to serve on the nominations committee of SBIC. She has also served as Chair of the Bioinorganic Subdivision of the ACS Inorganic Division and as Chair of the Chemistry section of the American Association for the Advancement of Science (AAAS). Perhaps most importantly, she is thoroughly invested in promoting the next generation of bioinorganic scientists through these service contributions.

Siderophores

This issue begins with a collection of three review articles covering siderophore chemistry and is followed by three original papers on siderophores. In the reviews, the chemical biology, coordination chemistry and reactivity of β-hydroxyaspartate siderophores, bis-hydroxamate macrocyclic siderophores and catecholate siderophores are covered by Hardy and Butler [7], Codd et al. [8] and McRose, Seyedsayamdost and Morel [9], respectively. Groves and co-authors [10] find that the stability constant for the amphiphilic mycobactin J siderophore of *Mycobacterium paratuberculosis* is much higher than previously reported, which when considered along with the observed rapid exchange kinetics suggests that mycobactins are capable of removing iron quickly from very high-affinity siderophores in cellular environments. The mechanism of Fe(III)-amphi-enterobactin uptake by *Vibrio campbellii* and *V. harveyi* strains is shown by Naka et al. [11] to occur via a novel outer membrane receptor, FapA, which can also recognize enterobactin. Neumann and Nolan [12] report on the applicability of disulfide linkers for siderophore-mediated delivery of antibiotics.

Metalloproteins

The second grouping of articles is defined by new reports on metalloproteins. The importance of lanthanide-dependent enzymes is revealed by Deng, Ro and Rosenzweig [13]

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in their article on the 1.85-Å-resolution crystal structure of the La(III)-XoxF enzyme, a pyrroloquinoline quinone-dependent methanol dehydrogenase enzyme from methylotrophic bacteria. Einsle and co-authors [14] report the 2.2-Å-resolution structure of the iron protein of the alternative vanadium-dependent nitrogenase system, VnfH from *Azotobacter vinelandii*, in its ADP-bound state, which shows that it is structurally similar to NifH and is thus consistent with the cross-reactivity observed for both ATP-hydrolyzing reductases.

Turning to heme enzymes, Wilks and co-authors [15] report that disruption of an aspartic acid/arginine salt bridge in HemO, the heme oxygenase from *Pseudomonas aeruginosa*, leads to complete loss of enzymic activity. Bren, Elliott and co-authors [16] report on the heme ruffling and electronic structure in covalently attached heme in different motifs, for example, with three of four intervening residues, as in CX_3CH or CX_4CH, in variants of *Hydrogenobacter thermophilus* cytochrome c552. Sono, Dawson and co-authors [17] report spectroscopic evidence for the coordination of neutral thiol to Fe(II)-heme. The heme-containing multi-copper oxidase MnxG from marine *Bacillus* bacteria oxidizes Mn(II) to Mn(III) and plays an important role in biogeochemical cycling of Mn. Tebo, Spiro, Casey, Britt and co-authors [18] report on the enzyme-bound Mn(III) intermediate as interrogated by electron paramagnetic resonance spectroscopy. Dominguez-Calva, Quintanar and co-authors [19] show that Hg plays a role in the bioinorganic chemistry of cataract disease by inducing aggregation of the human lens proteins. Küpper et al. [20] report on volatile halogenated compounds, and the speciation and localization of bromine and iodine in the brown algal genome model *Ectocarpus siliculosus* and specifically that the vanadium haloperoxidase is unlikely to be involved in halogenation of the precursor substrates to the volatile halogenated compounds.

Metallodrugs and metals in medical imaging

Five articles comprise the last section, on metallodrugs and metals in medical imaging. Cohen and co-authors [21] show that hydroxypyridinethione isosteric ligands are useful metal-binding pharmacophores and hence useful scaffolds for inhibitors of disease-associated metalloenzymes. Donnelly and co-authors [22] report on new radioactive technetium-99 m complexes for single-photon emission computed tomography (SPECT) imaging, which is useful for assisting in diagnosis of cerebral amyloid angiopathy. Martinez and co-workers [23] report evidence for enhanced luminescence upon coacervation of a genetically engineered elastin-like polypeptide (ELP) and a ruthenium(II) polypyridyl complex, with application as reporters in optical sensing and bioimaging applications. Nazarov, Hartinger and co-authors [24] investigated interactions of diruthenium anticancer agents with amino acids with an aim towards developing high antiproliferative activity in cancer cells. Finally, Barba-Behrens and co-authors [25] describe the nuclease properties of a series of trinuclear and mononuclear Cu complexes of carboxylate benzimidazole ligands, through a proposed oxidative Fenton-type pathway.

In sum, this volume reflects the wide-ranging interests of the larger biological inorganic community. Coverage of new aspects of siderophore chemistry, new metalloenzymes and new metallodrug candidates or metallo-imaging agents is very much aligned with the research interests of Alison Butler's research program and synchronous with many of the previous Alfred Bader Award winners.

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