

Bacterial synthesis of unusual sulfonamide and sulfone antibiotics by flavoenzyme-mediated sulfur dioxide capture.

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Abstract

Sulfa drugs, such as sulfonilamide and dapsone, are classical antibiotics that have been in clinical use worldwide. Despite the relatively simple architectures, practically no natural products are known to feature such aromatic sulfonamide or diarylsulfone substructures. We report the unexpected discovery of three fully unprecedented, sulfonyl-bridged alkaloid dimers (sulfadixiamycins A-C) from recombinant *Streptomyces* species harboring the entire xiamycin biosynthesis gene cluster. Sulfadixiamycins exhibit moderate antimycobacterial activities and potent antibiotic activities even against multidrug-resistant bacteria. Gene inactivation, complementation, and biotransformation experiments revealed that a flavin-dependent enzyme (XiaH) plays a key role in sulfadixiamycin biosynthesis. XiaH mediates a radical-based, three-component reaction involving two equivalents of xiamycin and sulfur dioxide, which is reminiscent of radical styrene/SO₂ copolymerization.

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