



Review

Mechanisms for rhamnolipids-mediated biodegradation of hydrophobic organic compounds



Zhuotong Zeng^{a,1}, Yang Liu^{b,1}, Hua Zhong^{b,c,1}, Rong Xiao^{a,1}, Guangming Zeng^{a,b,*}, Zhifeng Liu^b, Min Cheng^b, Cui Lai^b, Chen Zhang^b, Guansheng Liu^b, Lei Qin^b

^a Department of Dermatology, Second Xiangya Hospital, Central South University, Changsha 410011, Hunan, PR China

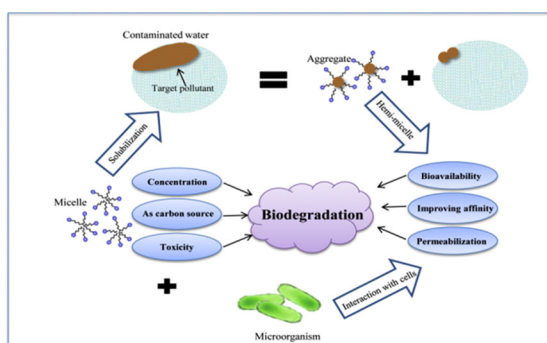
^b College of Environmental Science and Engineering, Hunan University and Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha 410082, PR China

^c State Key Laboratory of Water Resources and Hydropower Engineering Science, Wuhan University, Wuhan 430070, PR China

HIGHLIGHTS

- Rhamnolipids have excellent solubilization activity even below CMC concentration.
- Micellar HOCs bioavailability is based on hemi-micelle formation on cell surface.
- Rhamnolipids-induced release of LPS and rhamnolipids adsorption can change CSH.
- Rhamnolipids permeabilization facilitates the biodegradation of HOCs.
- Biodegradation and toxicity of rhamnolipids is important for their application.

GRAPHICAL ABSTRACT



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ABSTRACT

The widespread existence of hydrophobic organic compounds (HOCs) in soil and water poses a potential health hazard to human, such as skin diseases, heart diseases, carcinogenesis, etc. Surfactant-enhanced bioremediation has been regarded as one of the most viable technologies to treat HOCs contaminated soil and groundwater. As a biosurfactant that has been intensively studied, rhamnolipids have shown to enhance biodegradation of HOCs in the environment, however, the underlying mechanisms are not fully disclosed. In this paper, properties and production of rhamnolipids are summarized. Then effects of rhamnolipids on the biodegradation of HOCs, including solubilization, altering cell affinity to HOCs, and facilitating microbial uptake are reviewed in detail. Special attention is paid to how rhamnolipids change the bioavailability of HOCs, which are crucial for understanding the mechanism of rhamnolipids-mediated biodegradation. The biodegradation and toxicity of rhamnolipids are also discussed. Finally, perspectives and future research directions are proposed. This review adds insight to rhamnolipids-enhanced biodegradation process, and helps in application of rhamnolipids in bioremediation.

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* Corresponding author at: Department of Dermatology, Second Xiangya Hospital, Central South University, Changsha 410011, Hunan, PR China.

E-mail address: zgming@hnu.edu.cn (G. Zeng).

¹ These authors contribute equally to this article.

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1. Introduction

The anthropogenic environmental pollution by hydrophobic organic compounds (HOCs) is well documented (Luo et al., 2014; Wang et al., 2010). The widespread existence of HOCs in soil and water causes serious problems to ecosystem and human health, and thus has drawn increasing attentions (Cheng et al., 2016; Lee et al., 2014; Lin and Gan, 2011). It is reported that these compounds are carcinogenetic and teratogenetic, and could cause allergy, skin diseases, heart diseases, etc., after long-term exposure (Cheng et al., 2018; Xiong et al., 2018; Daifullah and Girgis, 2003; Standeker et al., 2007). The sound and effective techniques to treat HOCs contaminated sites have been proposed, and bioremediation is considered to have higher ecological significance and greater promise (Budd et al., 2009; Cheng et al., 2017b; Zhu et al., 2010). However, due to the hydrophobicity, most of HOCs either exist as non-aqueous phase liquids (NAPLs) or strongly adsorb onto soil matrix, which greatly decrease the bioremediation efficiency (de la Cueva et al., 2016; Ren et al., 2018).

Various studies have shown that the addition of surfactants facilitates removal of HOCs from contaminated soil and water (Mao et al., 2015; Trellu et al., 2016; Zhong et al., 2017). They are able to decrease the surface/interfacial tension of immiscible phase, increase the apparent solubility of HOCs, and thereby enhance the bioremediation (Cheng et al., 2017b; Zhang et al., 2015). Compared to chemical surfactants, biosurfactants have higher solubilizing ability towards hydrophobic pollutants (Barnadas-Rodríguez and Cladera, 2015; Yu et al., 2015). In addition, biosurfactants are more eco-friendly than most chemical synthetic surfactants (De et al., 2015; Yadav et al., 2016). As a result, biosurfactants have been promising alternatives in surfactant-based bioremediation (Zhong et al., 2017). Rhamnolipids, as a class of anionic glycolipid biosurfactant, have attracted particular interest. They present the maximum number of patents and publications among biosurfactants. According to Müller et al. (2012), >200 patents were registered for biosurfactants until 2012, and 50% of them are related to rhamnolipids. At the end of 2017, the numbers of publications on rhamnolipids and biosurfactants have reached 2100 and 4500, respectively.

Rhamnolipids are the most extensively studied and used biosurfactant in bioremediation area (De et al., 2015; Kim et al., 2015). They are biodegradable, less toxic, and can be produced from renewable resources (Gudiña et al., 2015; Ramírez et al., 2015). Studies also suggested that rhamnolipids are as good or better than synthetic surfactants (e.g., Tween 80 and Triton X-100) in enhancing aqueous solubility of HOCs, such as alkanes (Kiran et al., 2016), polycyclic aromatic hydrocarbons (PAHs) (Mahanty et al., 2011), polychlorinated biphenyls (PCBs) (Chakraborty and Das, 2016), and pesticides (Singh

et al., 2016). Moreover, it has been demonstrated that the presence of rhamnolipids could decrease the energy consumption of biodesulfurization by resting cells in biphasic O/W systems with hydrocarbon as the oil phase (Raheb et al., 2012). Due to these advantages, many studies have been performed on rhamnolipids-enhanced bioremediation in recent years (Lladó et al., 2012; Tahseen et al., 2016).

Some review papers (Bai et al., 2017; Hošková et al., 2013; Lamichhane et al., 2017; Shao et al., 2017) and few book chapters (Galabova et al., 2014; Leitermann et al., 2010) have summarized data on the application of rhamnolipids in bioremediation. To the best of our knowledge, however, these articles are mainly focused on biosynthesis and characteristics of rhamnolipids (Bai et al., 2017; Hošková et al., 2013), influence of rhamnolipids on microbial metabolism process (Shao et al., 2017), or simply the remediation efficiency (Lamichhane et al., 2017). To date, a comprehensive overview on mechanisms for rhamnolipids to enhance biodegradation of HOCs from a microscopic view point of interactions between rhamnolipids, HOCs, and microorganisms, are still in scarce. However, such an overview is important to fill the knowledge gap and definitely required, and thus is the focus of this article.

2. Rhamnolipids

As a biosurfactant produced by *Pseudomonas aeruginosa*, rhamnolipids were first reported in 1949 (Jarvis and Johnson, 1949). They are composed of L-rhamnose and β -hydroxy fatty acids moieties (Kiran et al., 2016). Up to date, over 60 congeners and homologues of rhamnolipids have been reported in literatures (Kourmentza et al., 2018). They are different in the number of rhamnose rings, chain length, and the saturability of fatty acid moiety (Lovaglio et al., 2015). Four common rhamnolipid homologues are Rha-C₁₀-C₁₀, Rha-C₁₀, Rha₂-C₁₀-C₁₀ and Rha₂-C₁₀, respectively (Liu et al., 2017).

It was reported that rhamnolipids can lower the interfacial tension of hexadecane/water from 43 to below 1 mN/m, decrease the surface tension of water from 72 to <30 mN/m, and have critical micelle concentration (CMC) value in the range of 10 to 200 mg/L (Dubeau et al., 2009; Hörmann et al., 2010; Müller et al., 2012). CMC is an important characteristic for surfactants, defined as the concentration of surfactants at which micelles begin to form and corresponds to the point at which the surfactant achieves the lowest stable surface/interfacial tension (Santos et al., 2016). Surface activity of rhamnolipids can be maintained even under extreme conditions of temperature (able to withstand 90 °C up to 120 min, and even 120 °C for 15 min) and pH (range from 3 to 11) (Hošková et al., 2015; Jackson et al., 2015; Pornsunthorntawe et al., 2008). Patel and Desai (1997a) and Patel and Desai, (1997b) reported that the hydrophilic/lipophilic balance (HLB) is 13 for

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