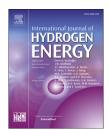
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# Biogranules applied in environmental engineering

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### ABSTRACT

The efficiency of wastewater treatment with renewable energy generation has been greatly improved with the development of biogranules. In this review article, various types of biogranules (methanogenic, hydrogenic, aerobic, anaerobic ammonium oxidation, and oxygenic photogranules) applied in environmental engineering are introduced along with their history, theories on how they are formed, physico-chemical and morphological characteristics, and the effects on enhanced performance. Although each individual granule has its own characteristics, there might be something in common that the formation is related with high production of extracellular polymeric substances, and they all have high hydrophobicity, settling velocity, and density. To our knowledge, this is the first review article dealing with various types of biogranules. The information given herein will provide a chance for a deep understanding on biogranules in both fundamental research and engineering point of views.

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### Introduction

In the last decades, the generation of wastewaters (domestic and industrial) has been rapidly increasing with population growth, urbanization, and industrialization, and the amounts produced are already far beyond the self-cleaning limit of natural aquatic systems. Thus, it is essential to treat such waste streams before discharge to protect human populations and eco-systems, as well as to improve our environmental quality [1]. Carbon, nitrogen, and phosphorous are, in general, the main targets to be treated, and are removed through

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### Abbreviations

AmGs	Anammox granules
Anammox Anaerobic ammonium oxidation	
ArGs	Aerobic granules
AUSB	Aerobic up-flow sludge blanket reactor
COD	Chemical oxygen demand
EGSB	Expanded granular sludge bed reactor
EPS	Extracellular polymeric substances
HGs	Hydrogenic granules
HPB	Hydrogen producing bacteria
HRT	Hydraulic retention time
LCA	Life cycle assessment
MGs	Methanogenic granules
OLR	Organic loading rate
OPGs	Oxygenic photogranules
SBR	Sequencing batch reactor
SRB	Sulfate reducing bacteria
UASB	Up-flow anaerobic sludge blanket reactor

physical, chemical, and biological units. In particular, biological treatments play an important role, being responsible for removal of more than half of all total organic pollutants in wastewater. They are also considered environmentally friendly technologies owing to less chemical use, and energy input, as well as renewable energy generation under certain conditions [2].

In biological treatment units, it is better to retain dense biomass to ensure the effectiveness of treatment and increase economic feasibility. Especially, it can directly determine the success of operation when slow-growing microbial species such as methanogenic and Anammox archaea perform main function [3,4]. The separation of cell retention from hydraulic retention has traditionally been carried out by settling of biomass and recycling them to the reactor. However, to reduce the footprint, advanced technologies such as use of immobilizing matrix, centrifugal systems, membrane filtration units, or other external sources of materials have been developed. Instead of using extra equipments, microbial granulation can be applied as an alternative method to attain high treatment efficiency [5].

Biogranules are discrete well-defined cell aggregates formed by cell-to-cell attraction that usually occurs in up-flow type reactors. Compared to conventional microbial flocs, biogranules have regular, dense, and strong structure with excellent settleability, enabling high cell retention, and the ability to withstand a high OLR [6]. The first biogranules discovered in the environmental field were used to treat industrial wastewater under anaerobic condition converting organics to CH<sub>4</sub> [3]. Until the end of 1990s, the main research on microbial granules was conducted for these MGs; however, this has expanded to various microbial processes, such as aerobic/anaerobic wastewater treatment, bio-H<sub>2</sub> production, Anammox, and photosynthesis. Color images with main reaction of each biogranules are shown in Fig. 1 [7–10]. It seems that each biogranules have different shapes, colors, and sizes, which might have resulted from different microbial species involved and formation mechanisms. There have been a few review articles of each individual biogranule type [11–13];

however, to our knowledge, no reports have investigated various biogranules applied in environmental engineering at the same time.

Therefore, this review was to introduce various biogranule types with their history, theories on how they are formed, physico-chemical and morphological characteristics, and their effects on enhanced performance by applying them. Moreover, future research required for each biogranule type is briefly addressed. The information here will enable a deeper understanding on biogranules from both fundamental research and engineering point of views.

### Various granule types

### Methanogenic granules

#### History/theory

The first MGs were discovered in a novel high-rate reactor, known as an UASB, in 1976 by Gatze Lettinga's group at Wageningen University in the Netherland. This system was used to treat sugar beet wastewater with an OLR of 15–40 kg  $COD/m^3/d$  at HRT 3–8 h in a 6 m<sup>3</sup> pilot plant [3]. The advent of UASB with a core component of MGs has revolutionized the conventional wastewater treatment process and become the most popular high rate reactor configuration for anaerobic wastewater treatment.

The success of an UASB highly depends on the establishment of healthy and strong biogranules, and the long start-up period (2-8 months) for developing MGs is considered the major drawback. To enhance the understanding of granulation process, many mechanisms and models have been introduced based on various perspectives as summarized in Table 1 [3,14-31]. The first proposed MGs model was an Inert Nuclei Model developed by Lettinga et al. [3]. In the presence of microsize inert materials, anaerobic bacteria can attach onto inert particles to form an initial biofilm. Subsequently, granules mature through the growth of these attached bacteria. Hulshoff et al. [14] proposed the Selection Pressure Model, in which microbial granulation is the result of a continuous sludge selection through washing out light and dispersed particles and retaining heavier biomass in the UASB. As the surface of bacteria is negatively charged, reducing the electrostatic repulsion between negatively charged bacteria by introducing multi-valence positive ions, such as Al<sup>3+</sup>, Ca<sup>2+</sup>,  ${\rm Fe}^{2+},$  and  ${\rm Mg}^{2+}$  was proposed as a granulation promotion method [16,17]. In addition, based on the microstructure of MGs under scanning electron microscope, Wiegant [23] proposed a Spaghetti model, in which the development of MGs is initiated by attachment of filamentous Methanosaeta on small flocs, followed by the formation of a three dimensional network through a branched-growth process. However, some models are known to be only applicable under specific conditions, and opposite observations have been widely reported. Furthermore, the validity of several suggested models is reportedly confined to only the initial stage of granulation, not the entire process. To accommodate the entire formation process of MGs, the following 4-step general model derived from the aforementioned previous models has been developed [11].

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