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# The development of aerobic granules from conventional activated sludge under anaerobic-aerobic cycles and their adaptation for treatment of dyeing wastewater

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## HIGHLIGHTS

- Aerobic granules formed from conventional activated sludge in synthetic media.
- Protein content of tightly bound EPS had greatest influence on granule properties.
- 73% color and 68% COD removal of dyeing wastewater achieved with adapted granules.
- Dye removal occurred during aerobic phase when %GR of aerobic granules was above 50%.
- Exposure of granules to dyeing wastewater enhanced EPS production but reduced %GR.

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ABSTRACT

In the present study aerobic granules were formed from conventional activated sludge under anaerobic-aerobic cycles in synthetic media and subsequently used for treatment of real dyeing wastewater. Mature aerobic granules, characterised by their consumption of loosely bound extracellular polymeric substances (LB-EPS), and with $\%GR_{0.3}$ (percentage of granules with size $\geqslant$0.3 mm) that exceeded 80%, were formed after 94 days of operation. Scanning electron microscope (SEM) structural analysis of the aerobic granules pointed to the possibility of the occurrence of azo dye decolorisation inside the aerobic granules during the aerobic phase of sequencing batch reactor (SBR) cycle. The presence of statistically meaningful correlations between total EPS, tightly bound EPS (TB-EPS) – including its protein (PN) and carbohydrate (PS) fraction and their ratio (PN/PS) – with $\%GR_{0.5}$ (percentage of granules with size $\geqslant$0.5 mm) and $SVI_{30}$ were identified, with the protein component of TB-EPS having the greatest influence. A hitherto unreported trend of change of EPS during both the anaerobic and aerobic phase and COD during the anaerobic phase was observed. The aerobic granules were successfully adapted to real dyeing wastewater with 73% color removal and 68% COD removal being achieved with a cycle time of 24 h and ratio of anaerobic to aerobic period of 3. Measurement of anaerobic and aerobic color removal revealed aerobic dye decolorisation in the anaerobic core of the aerobic granules up to the point where the $\%GR_{0.3}$ was above 50%. However, during the 94 days adaptation period, despite the increase in both total EPS and PN/PS ratio in the greater part of SBR operation, $\%GR_{0.3}$ showed a continuous drop from 81% to 31%.


## 1. Introduction

The textile industry is one of the most polluting industrial sectors and consumes large amounts of water. Therefore, treatment of the effluents originating from this sector is extremely challenging and important. Textile wastewater generally consists of dyes, minerals, surfactants, heavy metal ions, electrolytes, detergents, solvents and recalcitrant compounds [1]. Dyes, which are the dominating source of contamination in the textile wastewater, affect the effluent's color and by extension reduce the light transmission, the dissolved oxygen content and the general biodegradability of the water body receiving the effluent.

Bacterial treatment of dye containing wastewaters have many advantages over other biological [2] as well as physicochemical treatment methods [3]. In the bacterial treatment process, the azo bond in azo dyes –which represent the majority of the dyes

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utilized in textile industry – are reduced anaerobically whereas the resulting aromatic amines are potentially oxidized aerobically. The former leads to the decolorization of the wastewater whereas the latter leads to dye mineralization and reduction of the toxicity of the wastewater. A combined anaerobic-aerobic bacterial process is, therefore, a potentially attractive method for treatment of dye containing wastewaters [4]. This process is conventionally performed sequentially; however, this two processes can also be performed simultaneously through the use of aerobic granule technology [5]. Another potential advantage of the use of aerobic granules for the treatment of textile dyeing waste waters is the reportedly higher resistance of aerobic granules to toxic factors [6].

Although aerobic granule technology has been widely studied as a more advanced method for treatment of municipal and industrial wastewaters compared to activated sludge, there are only a few studies on the application of this technology for the treatment of textile dyeing wastewaters, and most of these have been carried out with synthetic wastewaters. The first report of the use of aerobic granules for such application was that by Muda et al. [5]. These authors successfully developed aerobic granules in a sequencing batch reactor (SBR) operated under anaerobic-aerobic cycles with dye containing synthetic wastewater using a mixture of activated and anaerobic sludge as inoculum. The treatment of the synthetic wastewater with the developed granules resulted in good COD removal but only 63% of the color was removed. In a further study [7], through optimisation of the HRT and the length of the anaerobic phase of the SBR cycle, these authors managed to increase the color removal efficiency to 95%. In this study, the authors noted that both HRT and the length of anaerobic phase affected the properties of the aerobic granules. In other studies, aerobic granules – preformed in the presence or absence of dye – were used for treatment of synthetic wastewaters containing the azo dyes Reactive Blue (RB) 59 [8], Acid Red (AR) 18 [9] and AR 14 [10].

There are several questions to be answered when considering the application of aerobic granules for treatment of real textile dyeing wastewater. The first is whether the granules should be pre-formed in non-dye containing synthetic media or in dye-containing synthetic (or real) wastewater? The former has the advantage that the complex granule formation process would not be adversely affected by dye or other components of the real wastewater and the granules would develop in a shorter period; the latter, on the other hand, has the advantage that the bacterial population in granules would become adapted to the dye and other wastewater constituents during the granule formation phase. Successful development of aerobic granules in synthetic media containing dyes has been reported [5,10]. To the knowledge of the authors, there are only two reports on the use of aerobic granules for the treatment of real textile wastewaters [11,12], and in both aerobic granules were formed in real textile wastewater. However, the period required for formation of mature granules was indeed much higher than that reported for synthetic media. Also, in both these studies the granules were formed in sterilised textile wastewater, presumably to eliminate the interference of the wastewater microbial flora on the aerobic granule formation process.

In the two aerobic granule studies conducted with real dyeing wastewater the granules were developed from a mixture of bacterial species isolated from textile sludge or soil contaminated with textile wastewater and sterilised activated sludge. Another strategy, not employed in previous studies, would be to develop the granules from readily available municipal activated sludge, which would simplify the granulation process. An uncertainty with this strategy is whether the aerobic granules formed in this way would perform satisfactorily in the treatment of real textile wastewater.

Another important point regarding the use of aerobic granules for treatment of real dye containing wastewaters is their stability during long term exposure to these wastewaters. This has not been considered in the two studies with real wastewater. Some previous studies with synthetic wastewaters suggest that under certain conditions some dyes can destabilize aerobic granules. For example, Sadri Moghadam and Alavi Moghadam [9] reported that although the presence of AR 18 at a concentration of 50 mg/l had little effect on aerobic granule structure, when the concentration of the dye was increased to 100 mg/l the granules disintegrated within a short time.

Since bacterial treatment of dye containing wastewaters is best performed under combined anaerobic-aerobic process, in previous reports aerobic granules for treatment of dye-containing wastewaters have been both formed and used under anaerobic-aerobic cycles. This provides both an opportunity and a challenge.

The opportunity is the possibility of the occurrence of azo dye decolourisation in the anaerobic inner core of the granules during the aerobic phase of the SBR cycle. This means that, unlike activated sludge processes in which the reduction of dyes only happens in the anaerobic phase of this combined process, with aerobic granules dye reduction can occur throughout the combined process. For this to happen the azo dye and organic compounds have to diffuse into the inner anaerobic core whereas simultaneously the products of the azo reduction process have to diffuse out. Significant aerobic decolourisation was reported during treatment of both AR 18 [9] and AR 14 [10] containing synthetic wastewaters with aerobic granules. However, the extent of aerobic decolourisation has not been reported in the two previous studies in which aerobic granules were used for the treatment of real textile wastewater.

The challenge concerning the development and use of aerobic granules under anaerobic-aerobic cycles is insufficient knowledge concerning the process of granule formation and uncertainties as to their stability under such cycles. This is partly because in previous studies of the use of aerobic granules for treatment of other wastewaters either aerobic only or anoxic-aerobic cycles have been employed. The recent study by Sadri Moghadam and Alavi Moghadam [13] suggests that the presence of an anaerobic phase in a SBR cycle has a profound effect on the properties of the formed aerobic granules. More studies is therefore needed in this area.

The aim of the present study was the development of aerobic granules from conventional activated sludge under anaerobic-aerobic cycles in synthetic media and their subsequent adaptation for treatment of real dyeing wastewater using a specific protocol. In order to shed more light on the development of aerobic granules under anaerobic-aerobic cycles and their stability when exposed to real dyeing wastewater, the trend of change of tightly-bound (TB-) and loosely-bound (LB-) extracellular polymeric substances (EPS) and their carbohydrate (PS) and protein (PN) fractions, as well as other parameters pertaining to the characteristics of the granules, were monitored during both the granule formation and adaptation periods of SBR operation, including during single cycles. The treatment performance of the aerobic granules during the adaptation period was assessed through the monitoring of COD and color removal during both the anaerobic and aerobic phases of operation as well as during a single cycle.

## 2. Materials and methods

### 2.1. Wastewaters and seed sludge

Synthetic wastewater with following composition was used for granule development and for dilution of real dyeing wastewater