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Improvement of the degradation of sulfate rich wastewater using sweetmeat waste (SMW) as nutrient supplement

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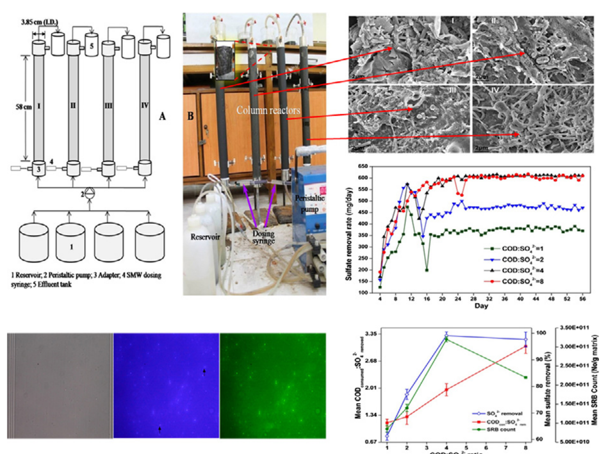
HIGHLIGHTS

- Sweetmeat waste (SMW) as a nutrient supplement in the SO_4^{2-} reduction system.
- COD/SO_4^{2-} ratio of 4 was found suitable for SO_4^{2-} removal.
- Sulfate reducing and acidogenic bacteria were dominant microbes in the system.
- Microbial diversities were almost remained unaltered at different COD/SO_4^{2-} ratios.

GRAPHICAL ABSTRACT



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ABSTRACT

External dosing of sweetmeat waste (SMW) dosing into exhausted upflow packed bed bioreactor (PBR) resulted in prompt reactivation of SO_4^{2-} removal. Different SMW concentrations in terms of chemical oxygen demand (COD)/SO_4^{2-} ratios (1, 2, 4 and 8) were introduced into four identical PBR where process stability was found within 3 weeks of operation. SO_4^{2-} removal was proportional to COD/SO_4^{2-} ratios up to 4 at which maximum sulfate removal (99%) was achieved at a rate of 607 mg/d. The value of COD$_{\text{consumption}}$:$SO_4^{2-}{}_{\text{removal}}$ was much higher at ratio 4 than 8 whereas, ratio 2 was preferred over all. Net effluent acetate concentration profile and total microbial population attached to the reactor matrices were corresponding to COD/SO_4^{2-} ratio as 4 > 8 > 2 » 1. Sulfate reducing bacteria (SRB) population was found to be inversely proportional to COD/SO_4^{2-} ratio in which acetate oxidizing SRB and fermentative bacteria were the dominant.

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1. Introduction

Organic carbon plays an important role in the microbial sulfate reduction in wastewater. Sulfate rich wastewater from mines and engineering excavations rapidly exhaust organic carbon [1]. Constructed wetlands, permeable reactive barrier, successive alkalinity producing systems are used to treat sulfate rich wastewater including acid mine drainage (AMD). An organic layer is common to all the substrates used for such treatment models. Different solid organic matrices such as bone char, diatomaceous earth pellets [2], vegetal carbon [3] oyster shells [4] were tested in the reactor system targeting field application. Exhaustion of organic carbon and its supply from this layer deactivate sulfate reduction [5,6].

External organic carbon addition is a choice to reactivate the process. Liquid or semisolid organic waste materials from food industries, municipal or domestic sewages are being tested currently for readily available and cheap bulk organic materials for microbial sulfate reduction. Application of potato processing plant waste [7], sugar beet waste [8], wine waste [9], sweetmeat waste [10], raw sewage [11], domestic sewage [12] as organic carbon and/or electron donor for sulfate reducing bacteria (SRB) in the laboratories or field scale applications is reported in several literature. Uses of waste material instead of a pure and synthetic substance such as lactate or ethanol can reduce the cost of the treatment. From an engineering point of view, these materials can also be stored in tanks and be fed continuously into the reactors [13]. Choice of these materials, however, would depend on the abundance in different geographical regions.

Sweetmeat waste (SMW) is abundantly available in the Indian subcontinent. It comprises of high sucrose (40%, w/w) along with diverse organic acids [10]. Reactivation of sulfate removal in an exhausted spent mushroom compost (SMC) packed reactor with fractionated SMW dosing was reported [14,15]. Before application in any field scale, optimization of the required SMW concentration was found necessary.

Reduction of 1 g SO_4^{2-} by SRB theoretically consumes 0.67 g chemical oxygen demand (COD) in standard condition [16,17]. This signifies all influent electron donors should be degraded via sulfate reduction, or all electrons would be finally donated to SO_4^{2-} if the COD/SO_4^{2-} ratio is kept below 0.67 [16]. In a mixed culture reactor, COD/SO_4^{2-} over 0.67 thus can intensify the competition of SRB with other anaerobic bacteria namely fermentative, syntrophs, homoacetogens and methanogens for the same electron donor [18,19].

Studies were conducted to understand the effect of different COD/SO_4^{2-} ratios on performance parameters [17,20–22]. The COD/SO_4^{2-} ratio in the range of 1–8 was reported to result in maximum sulfate removal using sugar (predominantly sucrose) in different reactors [23–26]. When lactate was used, the ratio was found to be optimum around 1.5–2.25 [27,28]. However, single substrate was tested in most studies. Therefore, the optimum COD concentration would be different in the case of SMW that contains diverse substrates.

Tests of reactivation of sulfate reduction in an organic carbon exhausted reactor by external carbon addition are relatively new and rarely addressed. In the earlier study, bactotryptone was used as a nitrogen source to study the effectiveness of SMW, primarily as a carbon source for sulfate reducing bacteria [10]. Because of its high cost, bactotryptone cannot be used as a nitrogen source for SRB in the large scale application. Subsequently, various nitrogen sources were used in combination with SMW. The result indicated that the relatively inexpensive NH_4HCO_3 was the preferable nitrogen source because of its ability to support sulfate reduction [15]. The present study is the continuation of the work.

The present study investigates the effect of different COD/SO_4^{2-} ratios on the process performance in an exhausted SMC packed reactor for reactivation. Four different SMW concentrations in terms of COD were applied keeping influent SO_4^{2-} concentration constant. The composition of the microbial community in the bioreactors was monitored. The novelty of the present study was the aim to find out the optimum dosing of SMW in terms of COD/SO_4^{2-} ratio that can support maximum sulfate reduction.

2. Materials and methods

2.1. Reactor and experiment design

The four identical tubular bioreactors (58 × 3.85 cm) were used in this study. These reactors were made of an acrylic sheet with steel adaptors at both ends [15]. Bioreactors were packed with 80 g of autoclaved SMC collected from a household farm after mushroom cultivation. The packed beds in bioreactors had porosities ranging from 0.63 to 0.64, and void volumes ranging from 425 to 435 mL. The beds had an equal mass density around 0.12 g/cm. Bioreactors were designated as I, II, III and IV. Initially, they were purged with N_2 (99.99% purity). Reducing agent supplements (pH 7.2) were passed through (with 24 h HRT) up to when the effluent ORP reached a consistent value around −150 mV. The SMC matrix of sulfate reducing bioreactor of the previous study conducted (fed with SMW along with NH_4HCO_3 after organic carbon exhaustion) was used as inoculum [15]. The inoculum was maintained at 37 °C in anaerobic water [15] with 1 mM bromoethane sulfonic acid (BESA) prior to the use in the reactor. The total cell count in inoculums was 2.26×10^{11} ($\pm 8.256 \times 10^9$/mL). It was introduced into the bioreactors with recirculation mode for 4 days to let the biomass attach to the matrix material. The bioreactors were run for another 80 days with the corresponding media without SMW until the effluent DOC concentration reached below 50 mg/L, at this stage sulfate removal from all reactors dropped below 50%. The temperature ranged from 25 to 28 °C at this period.

After cessation of sulfate reduction, external nutrient supplementation was started to revive the process by continuous addition of SMW rich nutrients. Another round of inoculation was done for 2 days in recirculation mode just before external nutrient addition.

The operational parameters of bioreactors tested for reactivation with different COD/SO_4^{2-} ratios are presented in Table 1. The reactors were operated in environmentally uncontrolled condition, as in a tropical country like India, to mimic the real situation where control of temperature and atmospheric pressure would cause additive cost. However, the flow rate was maintained as mentioned in Table 1. Reservoir solutions were prepared by: Na_2SO_4 2100 mg/L; $MgCl_2 \cdot 6H_2O$ 125 mg/L; $FeSO_4 \cdot 7H_2O$ 25 mg/L, trace elemental solution 10 mL/L, vitamin solution 100 μL/L, bromoethanesulfonic acid (BESA) 1 mM. Filtered SMW solution (1:10, w/v) was added accordingly to maintain respective COD/SO_4^{2-} ratios along with NH_4HCO_3 and K_2HPO_4 as N and P source. Final COD/N/P values were set 224.5:15.39:1 [23]. pH of the solutions were maintained around 7.2 by adding $NaHCO_3$. Trace elemental solution was prepared [29] with some modifications (mg/L): $FeCl_2 \cdot 4H_2O$, 1500; $CoCl_2 \cdot 6H_2O$, 190; $MnCl_2 \cdot 4H_2O$, 100; $ZnCl_2$, 70; H_3BO_3, 62; $Na_2MoO_4 \cdot 2H_2O$, 75; $NiCl_2 \cdot 6H_2O$, 30; $CuCl_2 \cdot 2H_2O$, 17 and 25% HCl, 10 mL/L. Vitamin solution was prepared as biotin 100 mg/L; Vitamin B12, p-aminobenzoic acid, and calcium D (+) pantothenate 500 mg/L; thiamine, pyridoxine-HCl, and nicotinic acid, 1000 mg/L [19]. All the ingredients except SMW were added into autoclaved and cooled with deoxygenated water, sterilized through 0.2 μm cellulose nitrate membrane (Whatman GmbH, Dassel, Germany). Autoclaved and cooled SMW solutions were added later separately. The SMW samples used in this report