

(様式 3)

## 論文内容の要旨

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The aim of this study was to investigate the possibility of combined lab-scale UASB/DHS processes for treatment of molasses-based wastewater in terms of COD removal rate, volatile fatty acid accumulation, biogas production and composition, profile of solid along the reactor etc. Secondly, we aim to evaluate the feasibility and the stability of a pilot-scale UASB/DHS/ASB following by microfiltration (MF), ultrafiltration (UF), and nanofiltration (NF) treating molasses bioethanol residue wastewater. And finally, we proposed the full-scale combined bio-physicochemical system for molasses-based wastewater treatment and investigated the microorganisms along with the evaluation of the system.

### **1. Development of Combined Anaerobic–Aerobic System for Treating Industrial Molasses Wastewater**

High-concentration industrial molasses wastewater treatment was examined using biological reactors coupled with physicochemical filtration membranes. The biological processes combined two mesophilic upflow anaerobic sludge blanket (UASB) reactors, a multi stage upflow anaerobic sludge blanket (MS)-UASB, and a regular UASB for primary anaerobic pre-organic removal, and a down flow hanging sponge (DHS) reactor, equipped with polyurethane sponge media for post-aerobic treatment. Concentrated blackstrap molasses was diluted [12,000–1,500 mg of chemical oxygen demand (COD)/L] with organic loading rate (OLR) of 4.5–57.7 kg-COD/m<sup>3</sup>/d (MS-UASB), 2.3–34.7 kg-COD/m<sup>3</sup>/d (UASB), and 0.2–6.0 kg-COD/m<sup>3</sup>/d (DHS). A 1:1.3 recirculation ratio within the MS-UASB was evaluated at different influent concentrations for COD, biogas (CH<sub>4</sub>) production, and nitrogen, phosphate, and color removal. The average total organic COD removal was over 92% with and without recirculation. A total of 150 NL/ d of biogas with 64–75% methane content was collected at the maximum loading rate and influent concentration. Ammonia was reduced from 30 mg-N/L to 5 mg-N/L in the DHS reactor. The dark influent could not be reduced biologically; however, ultrafiltration and nanofiltration removed 98% of the color.

### **2. Pilot-scale Anaerobic–Aerobic–Membrane System for Molasses Fermentation Residue Wastewater Treatment**

This paper describes the development of an effective system suitable for trial in a full-scale plant. A series of pilot-scale units consisting of upflow anaerobic sludge blanket (UASB) reactors, downflow hanging sponge (DHS) reactors, and anaerobic sludge blanket (ASB)

reactor, followed by microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) membrane units was assembled to conduct concurrent organic matter–nutrient removal and decolorization of molasses fermentation residue wastewater. The bioprocess operated with a total hydraulic retention time of 35 h. The UASB reactors removed 70% of the organic matter at the maximum chemical oxygen demand (COD) volumetric loading of 11.6 kg COD/m<sup>3</sup>/d. More than half (52%) of the raw wastewater organic nitrogen was converted to ammonia by the UASB reactors. The DHS reactors removed 60% of the total nitrogen. Denitrification (64%) occurred in the ASB reactor. MF was applied to remove particulate substances, preventing membrane fouling in the following units. Phosphorus was mainly removed by NF, with 82% rejection. Remarkably, the UF and NF units could achieve 90% and 99% color removal, respectively. No membrane fouling was detected in any of the membrane units along the entire operation.

### **3. Full-scale bio-physicochemical system for molasses-based wastewater treatment Performance and microbial community analysis of a full-scale hybrid anaerobic–aerobic membrane system for treating molasses-based bioethanol wastewater**

We evaluated the efficacy of a full-scale combined bio-physicochemical system for treating molasses-based bioethanol wastewater in terms of organic substances, nutrient, and dark brown color removal. The main organic removal unit, i.e., the up-flow anaerobic sludge blanket (UASB) reactor, achieved 80.7% removal and 4.3 Nm<sup>3</sup> methane production per cubic meter of wastewater with a hydraulic retention time of 16.7 h. Down-flow hanging sponge (DHS) reactors were important in reducing the biochemical oxygen demand (BOD), and the lowest possible organic waste intake prevented excessive biomass formation. The BOD removal efficiency was 71.2–97.9%. The denitrification up-flow anaerobic fixed bed (UFB) reactor achieved 99.2% total nitrogen removal. Post-physicochemical membrane treatment reduced the total phosphate, color, and remaining organic matter by 90.4%, 99.1%, and 99.8%, respectively. We analyzed the microbial diversity of the sludge from the UASB reactors. *Methanosaeta* was the dominant archaeal genus in the system, followed by *Methanolinea*, *Methanomicrospillum*, *Caldiserica*, *Bacteroidetes*, and *Deltaproteobacteria*.

With all above studies, we could conclude that a series of combined biological and physicochemical treatments effective for treating medium to high concentration molasses-based wastewater. The biological UASB-DHS-UFB had an important role as the main organic treatment, especially for reducing COD, SS, BOD, and nitrogen species, while the membranes (MF, UF, and NF) were a functional alternative treatment that enhanced the removal of untreated nutrients and color. In addition, the biogas recovered from UASBs could be used to increase the treatment performance and reduce the operational energy costs.