



Determination of the Optimum Hydraulic Retention Time in Two-Stage Anaerobic Fluidized Bed Bioreactor for Landfill Leachate Treatment

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Abstract. Leachate in Indonesian landfill sites poses a high risk to the surrounding environment should there be leakage in the accumulating ponds. Anaerobic digestion is an attractive option to clean up leachate, mostly due to the affordability of its operational cost. To enhance the efficiency of leachate digestion, anaerobic microbes were immobilized on the surface of natural zeolite powder. The powder was fluidized in a mesophilic anaerobic fluidized bed reactor (AFBR) for more stable biofilm formation. The AFBR scheme was split into two stages, with the first stage dominated by the acidogenic process and the second stage dominated by the methanogenic process. The dominating microbes in each stage were provoked by pH control to maintain the first stage acidic at pH 5-5.5 and the second stage neutral at pH 7-7.5. The first stage was run at five different hydraulic retention times (HRTs), while the second stage was run at three different HRTs to determine the optimum HRT for each stage. For acidogenic AFBR with HRTs of 5 days and 2.5 days, the VFA concentration profile increased for a longer period compared to the other HRTs. The COD removal efficiency at steady state was almost identical for all HRTs. For methanogenic AFBR, all three HRTs showed an identical rate of biogas formation at steady state.

Keywords: *anaerobic fluidized bed reactor; biogas; hydraulic retention time; immobilization; landfill leachate; zeolite.*

1 Introduction

One of the major pollution problems caused by municipal solid waste (MSW) landfill sites is landfill leachate, which is generated as a consequence of precipitation, surface run-off, and infiltration of ground water percolating through the landfill and inherent moisture content of the wastes. Generally, leachate may contain a large number of organic matter (bio-degradable but also refractory to biodegradation), as well as ammonia-nitrogen and heavy metals

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(such as Cd, Cu, and Ni), which form a great threat to the surrounding environment including soil, groundwater, and even surface water [1].

Generally, the best way of controlling pollution of the environment by landfill leachate is treating the leachate to remove or convert the hazardous components into non-hazardous ones before it enters the water system [2,3]. To do so, fluidized anaerobic treatment promises both pollution reduction of leachate and biogas production as energy source since it is used the same way in other biotechnological processes such as the fermentation and production of enzymes [4,5]. This process is considered more efficient than the conventional method using a pond system [6]. Anaerobic digestion is a technologically simple process with low energy requirement for converting organic material from a wide range of wastewater types, solid wastes and biomass into methane [7].

In anaerobic digestion, there are two processes: acidogenesis and methanogenesis [8]. For leachate anaerobic digestion usually only one single bioreactor is needed to accommodate both processes [9-11]. However, in this work, fresh landfill leachate was treated sequentially by separating the acidogenic and methanogenic processes. Two bioreactors were used in series, where acidogenesis was conducted in the first bioreactor and methanogenesis in the second one. The purpose was to maximize the organic content removal and volatile fatty acid formation in the first bioreactor and to enhance the biogas production in the second bioreactor. The objective of this study was to investigate the most suitable conditions in terms of pH level and hydraulic retention time (HRT) for each bioreactor in executing the aforementioned scheme.

The anaerobic fluidized bed reactor (AFBR) used in this study was basically an up-flow reactor operated anaerobically with small particles inside as microbial immobilization medium. An AFBR was chosen because it has higher organic removal efficiency at shorter HRT and better energy efficiency compared to other anaerobic reactors.

2 Materials and Methods

2.1 Materials

Fresh leachate was obtained from Piyungan Sanitary Landfill, Yogyakarta, Indonesia. Starter in the form of active digester effluent was supplied by the cow-manure based biogas mini-plant located at Gadjah Mada University's Center for Agro-technology Innovation in Berbah, Sleman. Immobilization media was produced from Mojokerto natural zeolite supported by bentonite as binding agent. To produce the immobilization media, the raw natural zeolite

powder (undersize 100 mesh) was mixed with bentonite at a weight ratio of 1:1. Then, the mixture was molded to form Raschig rings with an inside diameter of 1 cm, 5 mm thickness and 2 cm length, using an extruder. Lastly, the molded mixture was heated at 110 °C for 12 hours in a furnace (Thermolyne Tube Heater F21100). To control the pH level inside the acidogenic bioreactor, acetic acid glacial 98% (Merck) was used. The solid rings were then crushed into a coarse powder (0.5-0.8 mm) to be fluidized in both bioreactors (acidogenic and methanogenic).

2.2 Anaerobic Digestion of Leachate

Continuous anaerobic digestion was operated in a set of anaerobic fluidized bed reactors (AFBRs) mainly comprising of two bioreactor columns made of acrylic equipped with a closed-loop recirculation system for fluidization (Figure 1). The first one (AFBR 1) was a 15-L acidogenic bioreactor, while the second one (AFBR 2) was a 10-L methanogenic bioreactor. The acidogenic bioreactor was larger so as to provide flexibility in accommodating the fresh feed with fluctuating concentrations. The methanogenic reactor was smaller as it received more ideal feed, i.e. the effluent of the well-controlled acidogenic reactor, so that the microorganisms were less likely to experience shock loading.

To ensure that AFBR 1 was dominated by the acidogenic process and AFBR 2 was dominated by the methanogenic process, the pH values were set at different values in each bioreactor. For the acidogenic bioreactor, an acidic condition (pH 5-5.5) was preferred so as to prevent the growth of methnogenic cells since methanogenic cells like neutral conditions (pH 7-7.5). The pH was adjusted by adding weak acid (acetic acid) to the fresh leachate as influent stream to the acidogenic bioreactor. Observation of the acidogenesis process in the acidogenic bioreactor was conducted based on the volatile fatty acid (VFA) concentration profile.

In this work, five different HRTs were tested for the acidogenic bioreactor, i.e. 20 days, 15 days, 10 days, 5 days, and 2.5 days. Meanwhile, for the methanogenic bioreactor three different HRTs were used, i.e. 20 days, 10 days, and 5 days. Only two bioreactors were used in this work so that each bioreactor would be run beginning from the highest HRT followed by a lower HRT. The purpose was to keep the cells from shock loading and to help the adaptation of the cells [12].

2.3 Analysis of sCOD, VFA, and CH₄

In this work, the variable used to represent the substrate concentration was soluble chemical oxygen demand (sCOD). The analysis of sCOD and volatile fatty acids (VFA) during the experiment followed the standard procedure by

APHA [4]. sCOD analysis was conducted following the closed reflux colorimetric method. VFA analysis was conducted using the titrimetric method. The gas volume was measured using the gasometer method outlined by Walker [13], while the methane content was analyzed using gas chromatography (Shimadzu GC 8A).

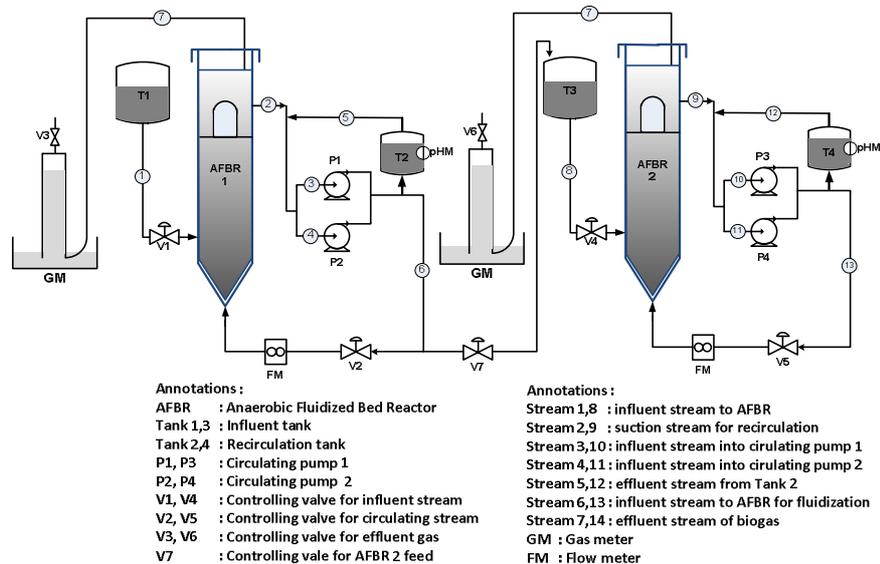


Figure 1 Experimental setup.

3 Results and Discussion

3.1 sCOD Profile in Acidogenic Bioreactor

Continuous operation of the acidogenic bioreactor was started from 20 days HRT with the initial concentration of leachate fluctuating between 2,500 and 4,000 mg/L, depending on the condition of the fresh leachate taken from the Piyungan landfill site. Since this was the first HRT to be applied in the continuously operating bioreactor, there was a startup period. In Figure 2, the startup period of the bioreactor was observed from day 0 to day 13. The percentage of sCOD removal reached a constant value (about 75%) after 20 days of HRT, which indicates that the bioreactor had achieved steady-state condition.

The experiments for lower HRTs than 20 days were conducted by maintaining the immobilized microorganisms in the AFBR and by only adjusting the input of the fresh leachate to set the HRT. This way, the microorganisms could adapt

to reach stable condition faster at other HRTs. As a consequence, the AFBR showed a very short startup period for the HRTs of 15 days, 10 days, 5 days, and 2.5 days.

The percentage of sCOD removal after reaching steady state was similar for all HRT values. After reaching steady state, the AFBR achieved a relatively similar percentage of sCOD removal (about 70-75%) at all HRTs.

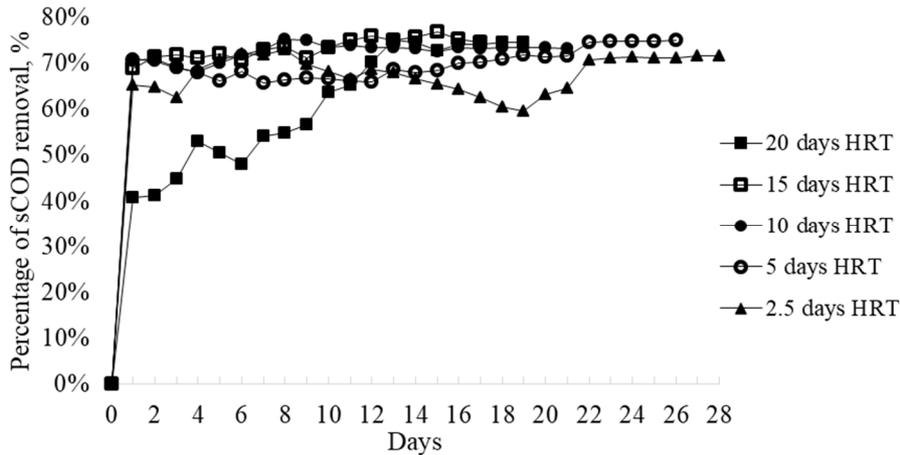


Figure 2 Percentage of sCOD removal of acidogenic bioreactor over five different HRTs.

3.2 VFA Concentration Profile in Acidogenic Bioreactor

From Figures 3 and 4, the VFA concentration in the effluent stream of the acidogenic bioreactor was lower than the VFA concentration in the influent stream. The removal of VFA was followed by biogas production in the acidogenic bioreactor at all five HRTs (Figure 5). The addition of acetic acid at low concentrations to adjust the pH level of the acidogenic bioreactor resulted in an increase of the VFA concentration in the influent stream. The VFA concentration of the fresh leachate was usually between 2,000 and 4,000 mg/L. Figure 3 highlights the fact that even the addition of acetic acid in the influent stream to lower pH below 6 did not enhance acidogenic activity. On the contrary, the methanogenic cells still survived in these acidic conditions.

Further experiments showed that the VFA effluent concentration tended to increase quite steadily at 5 days and 2.5 days of HRT. For both HRTs, the VFA concentration only started decreasing after 19 days of digestion, which was the longest period compared to the other HRT values. The decrease of the VFA concentration indicates that the methanogenic cells were able to consume a

significant amount of VFA. From these experimental data, it can be concluded that the most suitable HRT for the acidogenic bioreactor is a low HRT, possibly lower than 5 days. A trend of increasing VFA concentration is preferred because it confirms that the acidogenic process dominates the digestion process of leachate in the acidogenic AFBR.

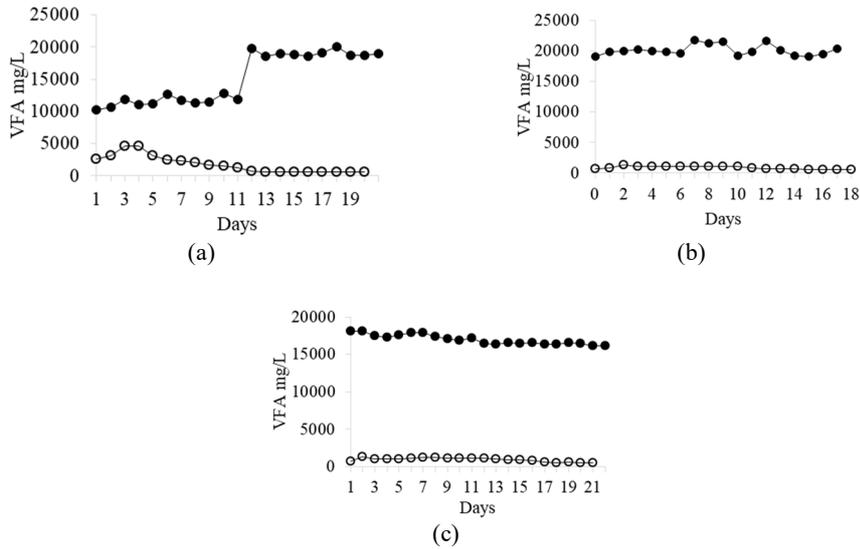


Figure 3 VFA concentration profile of the acidogenic bioreactor at the influent (●) and effluent (○) streams for (a) 20 days of HRT (b) 15 days of HRT (c) 10 days of HRT.

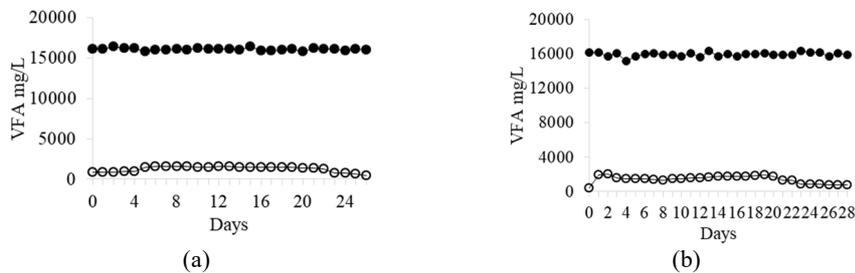


Figure 4 VFA concentration profile of the acidogenic bioreactor at the influent (●) and effluent (○) streams for (a) 5 days of HRT and (b) 2.5 days of HRT.

Unfortunately, even at low HRTs (5 days and 2.5 days), at which the VFA concentration profile kept increasing for a long time, there was still methane production (see Figure 5) at a small amount and with low methane purity. This indicates that a low HRT and acidic conditions (pH 5-5.5) cannot prevent both acidogenic and methanogenic processes occurring in the acidogenic bioreactor,

but at least it can support the domination of the acidogenic process over the methanogenic process.

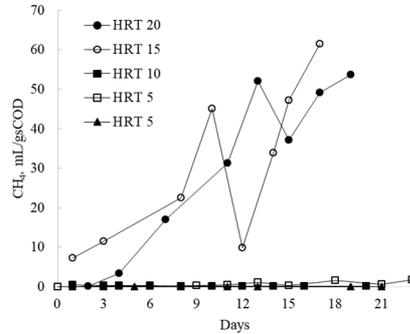


Figure 5 Profile of methane production in the acidogenic bioreactor over five different HRTs.

3.3 sCOD Profile in the Methanogenic Bioreactor

In this experiment, measurement of the VFA concentration was included in the sCOD measurement [4]. The decrease in sCOD concentration indicates that the

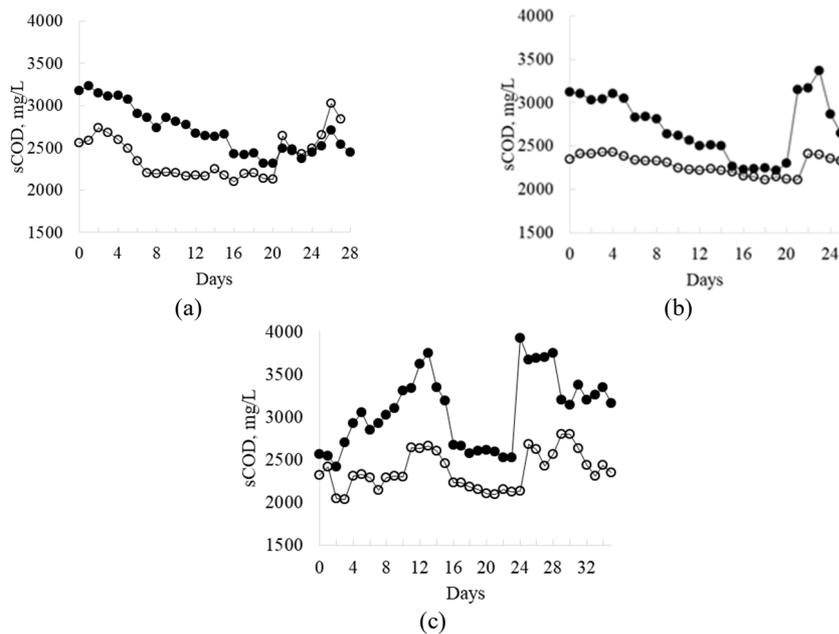


Figure 6 sCOD profile at influent (●) and effluent (○) streams of methanogenic bioreactor for: (a) 20 days of HRT, (b) 10 days of HRT, (c) 5 days of HRT.

VFA production by acidogenic microbes was smaller than the conversion of VFA into biogas by methanogenic microbes. In Figure 6(a), the sCOD in the effluent is higher than in the influent because the 10-day HRT was run only after the acidogenic reactor produced sufficient effluent. At this first run of the methanogenic reactor, it was initially inoculated and started up with a higher concentration of substrate so that the effluent had a higher sCOD concentration than the influent.

3.4 VFA Concentration Profile in the Methanogenic Bioreactor

Figure 7 confirms the effectivity of the methanogenic bioreactor. The VFA concentration in the effluent stream was always lower than in the influent stream. The interesting point of these experimental outcomes is the concentration of VFA in the effluent stream when it was in steady state. The

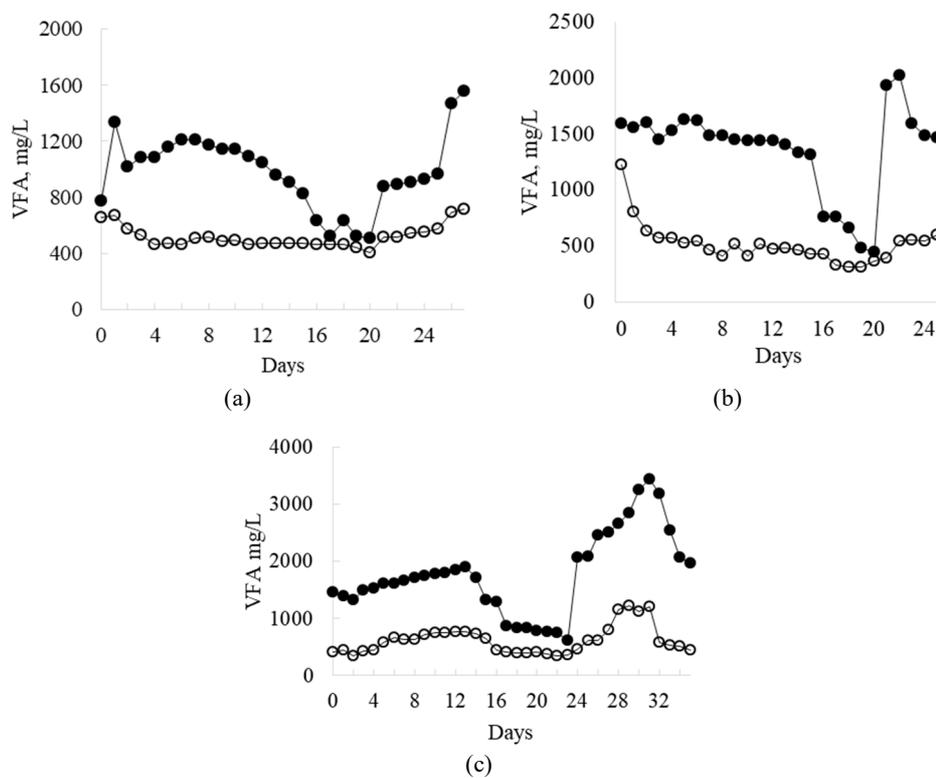


Figure 7 VFA concentration profile at influent (●) and effluent (○) stream of methanogenic bioreactor for: (a) 20 days of HRT, (b) 10 days of HRT, (c) 5 days of HRT.

VFA concentration in the effluent stream was always about 300-400 mg/L, which may be the minimum substrate concentration for methanogenic microbes to live in. Among the three HRTs tested, the methanogenic bioreactor was better at consuming VFA at 10 days of HRT and 5 days of HRT. However, the best HRT must be decided based on the biogas yielded as a product of VFA consumption.

3.5 Biogas Production in the Methanogenic Bioreactor

To decide the best HRT, the rate of biogas and methane purity was the concern. In Figure 8, the rates of biogas production of the three different HRTs have similar trends and also similar slopes. This indicates that all three HRTs were equally productive. The reason is because the VFA concentrations in the effluent streams of each bioreactor were also in the same range, between 300 and 400 mg/L.

The VFA concentration in the effluent stream represents the VFA concentration inside the bioreactor since the fluidization causes perfect mixing conditions inside the bioreactor. Insensitivity of methanogenic cells inside the methanogenic bioreactor (as explained in Sub-section 3.4) led to a constant VFA effluent concentration, meaning that the methanogenic cell could only produce methane at a minimum VFA concentration of around 300-400 mg/L. As the variation of VFA concentration in all HRTs was not significant, the biogas produced for all HRTs was almost identical.

In the case of bio-methane production, the methane purity fluctuated but in general was still in the same range (50-70%) for all three HRTs. The average methane purity produced by the methanogenic AFBR at a HRT of 20 days, 10 days, and 5 days was 50.6%, 57.9%, and 60.1% respectively. These values slightly differ but show that at lower HRT, the methanogenic microorganisms released more methane. However, based on Figure 8, the slight increase of biogas production and average methane purity from HRT 20 days to HRT 5 days shows that it is possible that the performance of the methanogenic AFBR can still be maintained with an HRT shorter than 5 days, which is interesting for further study.

The typical value of methane production from COD is 0.35 L methane/g COD [14]. The values after steady state was reached at day 30, as shown in Figure 7, ranged between 0.16 and 0.32 L/g COD. This indicates that the biogas productivity of the AFBR was quite good once the tricky start-up period was successfully overcome.

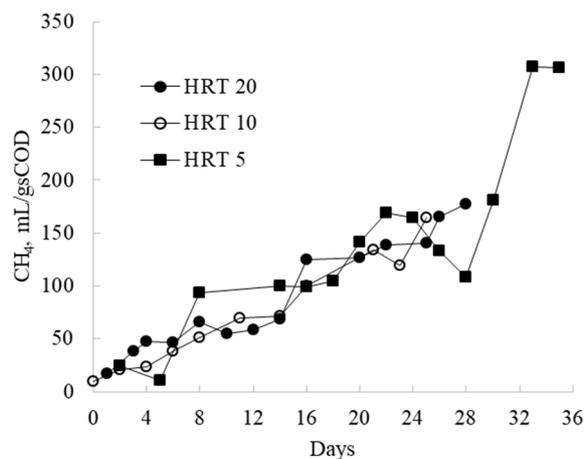


Figure 8 Profile of methane production in the methanogenic bioreactor for three different HRTs.

4 Conclusions

Lowering the pH level to acidic conditions did not guarantee that the acidogenic anaerobic fluidized bed bioreactor could totally dominate the process. However, an effort could be made to push the domination of the acidogenic process over the methanogenic process inside the bioreactor. By running the bioreactor at acidic conditions (pH 5-5.5) and at an HRT of 5 days or less, the sCOD decreased and the VFA effluent concentration kept increasing for a very long time, followed by a small amount of biogas production.

In neutral conditions (pH 7-7.5), the methanogenic bioreactor had a VFA effluent concentration that was stagnant in the range of 300-400 mg/L, even though the VFA influent concentration fluctuated. The different HRTs tested in this study did not significantly affect to the biogas production or the methane purity of the biogas from the methanogenic bioreactor. However, the possible increase of biogas production and average methane purity when the methanogenic AFBR is run at HRT shorter than 5 days could be the subject of a future study.

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