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A step-feed hybrid membrane bioreactor process for advanced wastewater treatment

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ABSTRACT

A step-feed anaerobic-(oxic/anoxic)^n-membrane bioreactor [An-(O/A)^n-MBR] process was developed to treat synthetic domestic wastewater. Characteristics of nutrient removal and membrane fouling at three hydraulic retention time (HRT) settings were investigated on a lab-scale system. Results showed that COD removal in the An-(O/A)^n-MBR process was high and stable (up to 98%) throughout the operation. At the constant sludge retention time condition, MLSS concentration increased with the decrease of HRT. Removal efficiencies of the total nitrogen (TN) and the total phosphorus (TP) demonstrated the same trend. When the HRT was at 8.70, 6.96 and 4.97 h, the average removal efficiencies of TN and the TP were 73.15%, 79.76% , 81.98% and 67.79%, 80.99%, 92.16%, respectively. However, analysis of membrane fouling showed that the short HRT obtained by high flux operation resulted in the acceleration of membrane fouling.

Keywords: Hydraulic retention time; Membrane bioreactor; Membrane fouling; Nitrogen and phosphorus removal; Step-feed

1. Introduction

Nitrogen and phosphorus are considered as the main sources of eutrophication and give rise to severe effects on receiving water [1]. In the past decades, a number of biological nutrient removal (BNR) processes had been developed [2–6]. Technically, the developed BNR processes, including anaerobic, anoxic and oxic phase, could be divided into two types according to the implementing approaches of the three phases: the temporal BNR process and the spatial BNR process. The temporal BNR process, such as the sequenced batch bioreactors, achieved the anaerobic, anoxic and oxide conditions by arranging them temporarily in a single reactor. As compared to the conventional activated sludge systems, the temporal BNR system had many advantages such as smaller footprint, improved nitrogen and phosphorus removal, less bulking, and flexible operation mode [7]. However, since all the phases occurred in a single reactor, this type of BNR process always operated in a discontinuous flow system. In a continuous flow system, the spatial BNR process, such as A^2/O process, was widely used throughout the world. This type of BNR process assigned different zones for each period (with sludge recycling serving as inoculum) to obtain the three conditions spatially. However, most of the spatial BNR process required additional energy for internal nitrified liquor circulation or addition of external carbon substrate for denitrification in anoxic zones, which led to the increase of the operational cost.

Step-feed anoxic/oxic activated sludge process (SAOASP) was one of the most practical methods to

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solve these problems because of its elimination of internal recycling and optimizing organic carbon utilization for denitrifcation [8–10]. The SAOASP processes were widely studied for nitrogen removal. To improve the phosphorus removal, SAOASP system was improved by adding an anaerobic zone before the multiple stages of aerobic-anoxic zones [11].

For decentralized, sensitive and yet unsewered areas, membrane bioreactor (MBR) technology could provide an elegant, robust and cost-effective treatment solution to achieve high effluent standard. When combined with enhanced biological phosphorous removal and/or phosphorous co-precipitation, high and stable phosphorous removal can be expected [12]. However, and in contrast to conventional activated sludge plants, process optimization still has to be done.

On the basis of above knowledge, the aim of the current work, therefore, was to develop a step-feed anaerobic-(oxic/anoxic)^n-membrane bioreactor [An-(O/A)^n-MBR] process for nutrient removal. This process, combining SAOASP with membrane separation, was composed of an anaerobic reactor, a multiple phases of aerobic and anoxic zones in sequence followed by a continuous aerated MBR. Three total hydraulic retention time (HRT) settings consisting of 8.70, 6.96 and 4.97 h were investigated on a lab-scale system. Nutrient removal performance, as well as membrane fouling, was compared at the different conditions imposed. The relationship between nutrient removal and membrane fouling was also analyzed.

2. Materials and methods

2.1. An-(O/A)^n-MBR system

The experimental set-up was shown in Fig. 1. The lab-scale An-(O/A)^n-MBR system was composed of an anaerobic reactor, a multiple phases of aerobic and anoxic zones in sequence followed by a continuous aerated MBR. The working volume for individual reactors was 14.6, 34.6 and 23.3 L respectively. The multiple phases of aerobic and anoxic zones consisted of eight compartments, the aerobic and the anoxic zones were arranged alternately, and the volume ratio of aerobic zone to anoxic zone was 2:1.

The first synthetic wastewater flow (Q_1) supplying nutrients for microorganisms growth and carbon for phosphorus release was fed into the anaerobic reactor while the second flow (Q_2) combined with the third flow (Q_3), the forth flow(Q_4) and the fifth flow (Q_5), was fed into the anoxic zone by stepwise feeding (Q_1:Q_2:Q_3:Q_4:Q_5 = 7:0.75:0.75:0.75:0.75). For the duration of the experimental period, the dissolved oxygen (DO) concentrations in the aerobic zones were controlled at 0.8–1.2 mg/L by adjusting the valves of aeration.

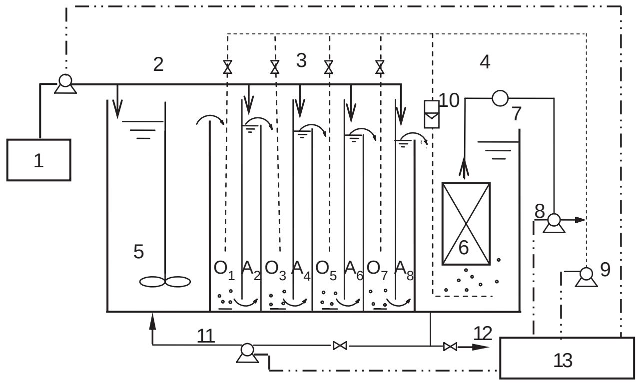

Fig. 1. Schematic diagram of the An-(O/A)^n-MBR system. 1. Wastewater reservoir, 2. Anaerobic reactor, 3. Multiple phases of aerobic and anoxic zones, 4. MBR tank, 5. Agitator, 6. Membrane module, 7. Pressure gauge, 8. Peristaltic pump, 9. Air blower, 10. Air flow meter, 11. Return sludge, 12. Excess sludge, 13. PLC system.

A MF hollow fiber membrane module (MOTIMO, China) was immersed in the MBR tank. The membrane module had an effective filtration area of 1.0 m^2 and its nominal pore size is 0.22 μm. An air diffuser was installed underneath the membrane module to provide air at 0.5 m^3/h. The bubbling air was used to serve for three purposes, providing oxygen for the microorganisms in aerobic tank, mixing the aerobic tank and removing of cake deposition on the membrane surface. The mixed liquor at the bottom of MBR tank was recycled to the anaerobic reactor continuously at a rate of 0.75. To alleviate membrane fouling, the membrane was operated in an intermittent mode, 10 min suction and 2 min rest.

2.2. Influent quality and sludge seeding

Synthetic wastewater, composed of glucose, starch, NH_4Cl, KH_2PO_4, $NaHCO_3$ and mineral solution (Mg, Ca and Fe), was fed to the lab-scale system throughout the operation. The initial influent contained 342.8–424.1 mg COD/L, 29.7–36.0 mg TN/L, 23.76–34.32 mg NH_3-N/L and 3.47–5.67 mg TP/L. The pH value was about 7.3.

The seeding sludge was supplied from the aeration tank at a municipal wastewater treatment plant in Xuzhou city (China). After seeding, the sludge was incubated with the synthetic wastewater in batch culture for a week. Then they were put into the An-(O/A)^n-MBR system for acclimation of the microorganisms. Three weeks later, it was judged that the process of incubation and acclimation was completed.

2.3. Operation

In this study, three different HRTs were implemented by the adjustment of membrane flux. The HRT was 8.70,

Table 1
Specifications of the experimental conditions.

Conditions	Run 1	Run 2	Run 3
Experimental period, d	45	45	28
Influent flow rate, m^3/d	0.21	0.26	0.36
Flux, $L/(m^2\ h)$	10	12.5	17.5
HRT, h	8.70	6.96	4.97
Membrane cleaning	Off-line chemical cleaning after each experiment		
SRT, d	26		
Temperature, °C.	25.3–32.5		

6.96 and 4.97 h when the membrane flux was maintained at 10, 12.5 and 17.5 $L/(m^2\ h)$. The synthetic wastewater was fed into the system with a flow rate of 0.21, 0.26 and 0.36 m^3/d for 45 days, 45 and 28 days in Run 1, Run 2 and Run 3, respectively. Table 1 showed the operation conditions of these experiments. The excess sludge was withdrawn to keep the sludge retention time (SRT) of 26 days throughout the operation.

In addition, membrane cleaning was conducted after Run 1 and when the trans-membrane pressure (TMP) reached at about 30 kPa in Run 2 and Run 3. When cleaning, membrane was taken out of the MBR tank and rinsed with 0.5% (v/v) sodium hypochlorite solution for 24 h followed by 2% hydrochloric acid solution for 12 h. The other membrane was placed to the MBR tank to operate the An-$(O/A)^n$-MBR system during the cleaning period.

2.4. Analytical methods

All the results presented were obtained from the An-$(O/A)^n$-MBR system at its steady state. The samples taken from bioreactors were filtered using 0.45 µm filter paper. Dissolved oxygen concentration was measured using the DO meter (WTW Oxi 340, Germany). pH using the pH meter (PHSJ-4A, China). Measurement of COD, MLSS, total nitrogen (TN), oxidized nitrogen (NO_3^--N and NO_2^--N), ammonium nitrogen (NH_4^+-N), orthophosphate concentration (*ortho*-P) and total phosphorus (TP) followed standard methods [13].

3. Results and discussion

3.1. Nutrients removal

3.1.1. COD removal

Daily variation of the influent and effluent qualities was studied during the operation (as shown in Table 2). It seemed that HRT had little influence on the organic pollutants removal in the An-$(O/A)^n$-MBR system. As presented in Fig. 2(a), although the influent COD fluctuated from 342.8 to 424.1 mg/L, the COD removal efficiency was high and stable, and was 97.30–99.54%, 95.70–99.51% and 97.09–99.06% in Run 1, Run 2 and Run 3, corresponding to 1.53–9.63, 1.98–11.32 and 3.42–10.95 mg/L in the effluent.

The effluent COD level in the An-$(O/A)^n$-MBR process was sufficient to meet the standard of water reclamation in China. The high COD removal was due to the growth of high biomass in the system and the efficient utilization of organic compounds in the anaerobic reactor for phosphorus release and in the anoxic zones

Table 2
Characteristics of nutrient removal in each experiment (with standard deviations in parentheses).

Items	Run 1			Run 2			Run 3		
	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Influent (mg/L)	Effluent (mg/L)	Removal (%)	Influent (mg/L)	Effluent (mg/L)	Removal (%)
COD	386.25 (19.97)	4.06 (2.80)	98.94 (0.75)	385.33 (21.67)	6.91 (4.32)	98.17 (1.22)	384.80 (23.84)	7.74 (2.46)	97.99 (0.63)
TN	33.38 (1.23)	8.97 (1.43)	73.15 (3.97)	33.67 (1.17)	6.81 (0.79)	79.76 (2.32)	33.11 (1.50)	5.95 (0.71)	81.98 (2.30)
TP	4.36 (0.32)	1.40 (0.40)	67.79 (9.21)	4.39 (0.50)	0.82 (0.27)	80.99 (6.59)	4.21 (0.24)	0.32 (0.14)	92.16 (3.77)

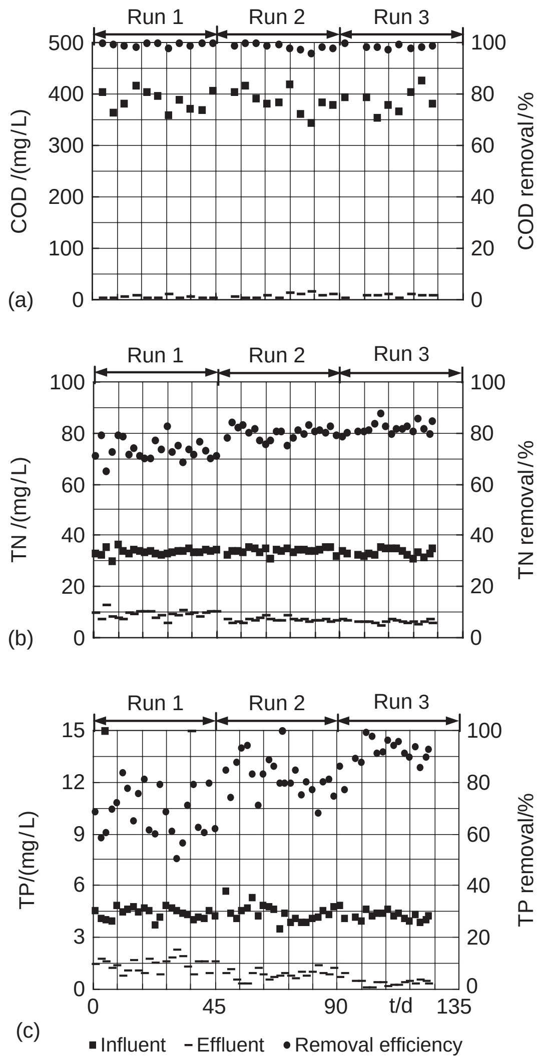

Fig. 2. Influent and effluent qualities and removal efficiency: (a) COD, (b) TN and (c) TP.

for denitrification. In addition, the high removal efficiency could also be attributed to complete particulate retention of suspended COD and BOD_5, high molecular weight organics, and biomass [14].

3.1.2. TN removal

As shown in Table 2 and Fig. 2(b), the An-(O/A)^n-MBR system achieved high performance on TN removal throughout the operation and TN removal efficiency increased with the decrease of HRT. The average TN removal was 73.15%, 79.76% and 81.98% in Run 1, Run 2 and Run 3, corresponding to 8.97, 6.81 and 5.95 mg/L in the effluent.

The high removal of TN was attributed to the high performance of nitrification as well as the denitrification in the system. NH_4Cl was used as the sole nitrogen source in the synthetic wastewater, little ammonium nitrogen, however, was detected and it even became undetectable in the effluent, which implied that full nitrification was achieved in the An-(O/A)^n-MBR process. When the step feeding strategy was adopted, the HRT in the front stages was longer due to the less influent flowrate and the microbes would grow in quantities in the aerobic reactors [15]. The growth rate of microbes increased with the decrease of HRT. At the constant SRT condition, the MLSS concentration in the aerobic compartments increased by about 1.23%, 1.39%, and 2.03%, respectively in run 1, run 2 and run 3 (data not shown). Consequently, the biomass density in the An-(O/A)^n-MBR system in Run 3 was higher than that in Run 1 or Run 2 and MLSS concentration in the MBR tank increased from 9062 mg/L at HRT of 8.70 h to 11,729 mg/L at that of 4.97 h correspondingly. High MLSS concentration and low COD level in the aerobic compartments and in the MBR tank ensured the predominance of autotrophic bacteria. Furthermore, the high ability for nitrification could also be attributed to the excellent retainability of the membrane to biomass which caused the nitrobacteria flourish.

The special configuration adopted in the An-(O/A)^n-MBR process enhanced the high denitrification ability in the An-(O/A)^n-MBR process. Firstly, besides receiving influent at the anaerobic zone, the process also received influent at plural oxic/anoxic tanks, which could be used as an external carbon source and thus improved the denitrification ability. Secondly, more combinations of oxic and anoxic tanks in series enabled nitrification stream flow from oxic zone of one pass to anoxic zone of next pass straightly, which could decrease the accumulation of nitrate. It should be noted that denitrification in the An-(O/A)^n-MBR system was accomplished not only by normal denitrifying bacteria but also by denitrifying phosphate-accumulating bacteria (DPB), resulting in the increase of denitrification capacity (discussed later). In addition, low DO concentration (about 1 mg/L) in the oxic tanks and high MLSS concentration in the MBR reactor allowed nitrification and denitrification to occur simultaneously.

However, in an ideal SAOASP system, complete denitrification and full nitrification could be achieved sequentially in the alternating anoxic and oxic periods, so the effluent TN concentration was determined by the influent fraction flowed into the last stage. In our study, the highest TN removal efficiency obtained in the An-(O/A)^n-MBR R system was 81.98%, lower than that calculated theoretically (95.71%). Since full nitrification was achieved in the An-(O/A)^n-MBR process, denitrification would be the limiting factor of TN removal in the system.

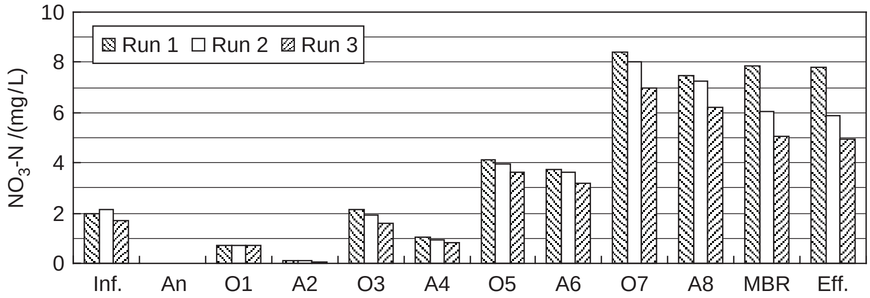

Fig. 3. Nitrate variation in each period at different HRTs.

To evaluate the denitrification performance of the An-(O/A)^n-MBR system, Nitrate variation in each period at different HRTs settings were studied. As presented in Fig. 3, the variation of nitrate concentration in the multiple phases of aerobic and anoxic zones showed the same trend at different runs. With the external addition of the carbon source and the residual organic substances from the anaerobic reactor, a higher denitrification rate and lower nitrate concentration were achieved in the former anoxic compartments (A_2 and A_4). In the following anoxic compartments (A_6 and A_8), to which the less flow (0.075Q) was fed and little residual carbon source from previous compartment flowed, less organic substrate was available for utilization, giving rise to the accumulation of nitrate concentration. Compare to long HRT condition, short HRT condition introduced more organic substrates to the system. This was the reason why the nitrate concentration in Run 3 was lower than that in Run 1 or Run 2. Furthermore, simultaneous nitrification and denitrification could be enhanced in the aerobic compartments due to higher MLSS concentration in Run 3.

3.1.3. TP removal

As shown in Fig. 2(c), the An-(O/A)^n-MBR process was also effective in achieving high performance on phosphorus removal. A similar trend in TP removal was observed as for TN reduction, but the difference of TP among three HRT settings was more greatly found as compared to that of TN. Total phosphorus removal increased with the decrease of HRT. When the HRT was 8.70 h, the TP removal was low and unstable, which fluctuated from 50.1% to 83.4%, the average TP concentration in the effluent was 1.40 mg/L. When the HRT was 6.96 h, the TP removal varied from 74.5% to 94.1%, and effluent TP from 0.27 to1.24 mg/L. The effluent TP concentration was as low as 0.5 mg/L when HRT was changed to 4.97 h. This could meet the water quality standard for discharge in China.

It was observed that phosphorus was up-taken in the anoxic compartments as well as in the aerobic compartments in this system. Studies show that the presence of a small amount of nitrate may stimulate the growth of DPB. Denitrifying phosphate-accumulating bacteria, a fraction of PAOs, can also take up phosphorus under anoxic conditions using nitrate as the electron acceptor instead of oxygen [16]. Therefore, the decrease of phosphorus concentrations in the anoxic compartments might attribute to the simultaneous denitrification and phosphorus uptake by DPB.

Short HRT introduced more organic substrates to the anaerobic reactor as well as to the anoxic compartments. So the phosphorus release and the simultaneous denitrification and phosphorus uptake were enhanced. As a result, the phosphorus removal was improved in Run 3. In addition, the particulate phosphorus matters retained by membrane filtration were responsible for the low level of phosphorus concentration in the effluent.

3.2. Membrane fouling

Trans-membrane pressure variation at different HRTs was illustrated in Fig. 4. In run 1, the TMP value increased slowly and maintained at a low level (<11 kPa) for 45 days. It took 26 days in Run 2 and only 6 days in

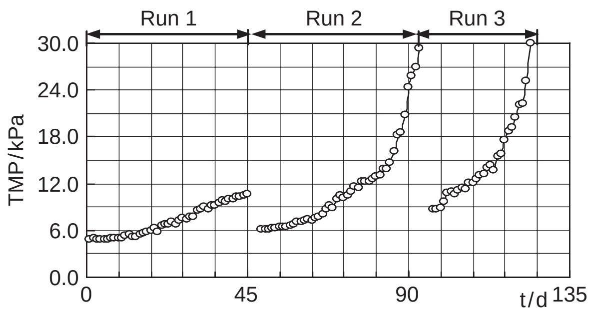

Fig. 4. Variation of the TMP at different HRTs.

Run 3 to increase to the same level. In Run 2, a characteristic three-stage TMP profile was observed with an initially extended period of slow and fast TMP rise followed by a sudden transition to a rapid rise and the rate of TMP rise was 0.11, 0.31 and 1.92 kPa/d correspondingly. It took about 45 days for the TMP to reach about 30 kPa in Run 2 and only 28 days in Run 3.

The lower flux enabled the sub-critical flux condition in which the TMP was maintained at a low level for a long time. Thus, it was clear that a flux of 10 L/(m^2·h) was most desirable for retarding membrane fouling compared with 12.5 and 17.5 L/(m^2·h). It seemed that flux of 17.5 L/(m^2·h) was too high to maintain the membrane operate below the critical flux condition because the TMP increased right after the beginning of membrane filtration.

In addition, off-line chemical cleaning seemed to be an effective cleaning method because it ensured perfect regeneration of the membrane. Moreover, this method avoided the possible negative effect of the chemicals on the microorganisms in the bioreactor when on-line chemical cleaning was conducted.

3.3. Effect of HRT on nutrient removal and membrane fouling

In this study, the influent quality was controlled at a steady state (as shown in Table 2), so the carbon to nitrogen ratio, as well as the carbon to phosphorus ratio, was kept at a relatively stable level. Therefore, it could be concluded that influent quality exhibited no great effect on nutrient removal in this study.

Short HRT brought more organic substances to the system, resulting in the rapid growth of microorganisms and the variation of the organic load rate (OLR). The OLR was 0.190 kg COD/kg MLSS•d, 0.231 kg COD/kg MLSS•d and 0.276 kg COD/kg MLSS•d in Run 1, Run 2 and Run 3, respectively. Therefore, the difference of nitrogen and phosphorus removal in Run 1, Run 2 and Run 3 could be caused by the variation of OLR due to the different HRT in the An-(O/A)^n-MBR system. Short HRT introduced higher OLR to the system, resulting in the enhancement of the biological capacity of denitrification and phosphorus release. Eventually, under the conditions imposed, nitrogen and phosphorus removal were improved [17]. Therefore, the performance of nitrogen and phosphorus removal was better in Run 3 than that in Run 1 or Run 2. It was also shown that not only the removal efficiency but the removal stability was improved under the conditions of short HRT (illustrated in Table 2 and Fig. 2).

Long HRT resulted in low flux of the membrane operation, which was desirable in terms of retarding membrane fouling. However, Long HRT introduced low OLR in the system, resulting in the decrease of nitrogen and phosphorus removal due to the decrease of bioactivity. From this point, it seemed difficult to obtain high nutrient removal and slow membrane fouling simultaneously. In order to solve this problem, more membrane modules should be installed to achieve a higher HRT condition without increasing the membrane flux.

4. Conclusions

An innovative step-feed An-(O/A)^n-MBR process was developed to treat synthetic domestic wastewater and its performance was investigated on a lab-scale system. Major findings from this study are summarized as follows:

1. Combining SAOASP with membrane separation, the step-feed An-(O/A)^n-MBR process might be a promising process alternative for wastewater treatment because of its elimination of internal recycling and optimizing organic carbon utilization as well as its high effluent quality.
2. At the constant SRT condition, COD removal was high and stable (up to 98%) throughout the operation. MLSS concentration increased with the decrease of HRT. Removal efficiencies of TN and TP demonstrated the same trend. When the HRT was at 8.70, 6.96 and 4.97 h, the average removal efficiencies of TN and TP were 73.15%, 79.76%, 81.98% and 67.79%, 80.99%, 92.16%, respectively.
3. Short HRT was desirable in terms of improving the treatment capacity as well as the nutrients removal. However, short HRT resulted in the high flux operation, which led to the acceleration of membrane fouling. To obtain high nutrient removal and slow membrane fouling simultaneously, more membrane modules should be installed to achieve a higher HRT condition without increasing the membrane flux.

Further investigations should be conducted on the optimization of influent distribution according to different influent quality and on the microbial kinetic analysis of the step-feed An-(O/A)^n-MBR process.

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