A Preliminary Study on Application of MBR+NF/RO (Membrane Bio-Reactor + Nanofiltration/Reverse Osmosis) Combination Process for Landfill Leachate Treatment in China

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Abstract. Landfill leachate involves high concentrations of organic compounds, ammonia nitrogen, heavy metals and other complex components. Hence, it is very difficult to treat using conventional biological processes alone. This research is a preliminary literature review on using MBR+NF/RO combination process to achieve purification of landfill leachate in China. MBR+NF/RO process is a combination of biological and physico-chemical technology, including MBR system, nanofiltration and reverse osmosis devices. The study found that, the benefit of a complementary combination of MBR and NF/RO process with respect to removal of ammonia nitrogen, trace organic and heavy metal ions appears quite intuitive. And this promising treatment has developed rapidly in China in last few decades. In this paper, representative project examples in China were selected for review. Mechanism and influencing factors of MBR+NF/RO process in China in the past decades will be reviewed and recommendations will be given for the future development of Chinese MBR+NF/RO process.

1. Introduction
In the past decades, the municipal solid waste (MSW) production has been increased rapidly throughout the whole world as an accompaniment of the increasingly wealthy lifestyles, continuing industrial and commercial growth in many countries [1]. Currently, landfill is the treatment which has been widely applied in most Chinese garbage disposal plant to treat MSW, compared to other treatments such as waste incineration, composting…, etc. In China, over 150 million tons of MSW are produced and keep a stable increase rate of 8%–10% every year. And about 80% MSW are disposed by local sanitary landfills [2, 3]. However, landfill leachate, a complex, hazardous wastewater, is formed during the biochemical, chemical and physical reaction process in the waste cells with the percolation of rainwater and the moisture contained in wastes and its composition fluctuates with the landfill age. The satisfactory purification of landfill leachate is hard to achieve by conventional processes alone, because it often contains a large quantity of organic matter, ammonia, heavy metals, and inorganic salts and varies of hazardous materials such as pharmaceuticals and personal care products (PPCPs) [3-6]. Therefore, a lot of physico-chemical and biological combination treatments process are studied and experimented by scientists domestic and abroad for effective leachate treatment [3, 6]. On the one hand, among all the biological treatment process, MBR has been expected to be a superior and effective way to treat landfill leachate due to its smaller footprint, higher effluent quality, increased biomass, process stability and low sludge production [4, 7-9]. Therefore, MBR process were widely
applied in China for wastewater treatment with the average annual growth rate of nearly 100% and the capacity becoming larger and larger in the past decades [10, 11]. On the other hand, a lot of physico-chemical processes for leachate treatment are intensively studied because of its resistance for toxic and hazardous materials. And NF/RO membrane process shows outstanding removal efficiency of colour, most organic contaminants and inorganic salt as a post-treatment of MBR for further desalination and purification [3]. In [12], Drewes et al. studied the efficacy of NF, RO and some conventional processes at removing organic carbon. It showed that TOC rejections by RO and NF were 94%-96.4% and 91.3%-94.5%, respectively, much higher than the conventional ones.

In order to meet those stricter environment quality standards (GB16889-2008), MBR and NF/RO process are combined together as a novel combination process for landfill leachate treatment [8]. A large amount of MBR+NF/RO combination process are put in use to treat landfill leachate both lab-scale and industrial scale in China. However, there is not such a detailed review on MBR+NF/RO combination process in China recently. In this paper, the mechanism and application situation of MBR+NF/RO combination process for landfill leachate treatment and purification in China were reviewed, and the perspective analysis of representative project examples was carried out as well to provide recommendations for future development.

2. Literature Review

2.1. Characterization and Mechanism

2.1.1. MBR Step

MBR is a combination of conventional activated sludge process and membrane separation technology. With efficient membrane filtration that holds small pore sizes of less than 0.1 mm inside, MBR can control over both solids retention times (SRTs) and hydraulic retention times (HRTs) separately and reduce sludge production, which makes MBR more advanced and efficient than other biological treatment [13, 14]. In [13], the performance of MBR and sequencing batch reactor (SBR) are compared in high-strength landfill leachate treatment. Result shows the removal efficiencies of MBR for BOD₅, TN and NH₃ surpass 95% with an improvement of SBR removal efficiencies ranging from 21 to 34%.

With the popularity of MBR in China, various kinds of MBR appear including anaerobic MBR, aerobic MBR and etc. However, the two-stage anoxic/oxic MBR process (AO/AO-MBR) is applied in most landfill leachate treatment project in China, due to its superior removal of nitrogen through nitrification and denitrification [15]. This MBR process contains of a two-stage anoxic and aeration tank followed by an ultrafiltration (UF) process to achieve further clarification. With the biodegradation reaction in the two-stage anoxic and aeration tank and the filtration of the UF, most of chemical oxygen demand (COD), ammonia (NH₄⁺-N) and total nitrogen (TN) can be removed effectively. In [15, 16], Liu et al. observed a two-stage AO/AO-MBR for landfill leachate treatment for a long period (81 d, 113 d). The best performance showed that the removal efficiencies of COD, ammonia and TN reached 85.6%, 99.1%, and 77.6%, respectively. Thus, the two-stage AO/AO-MBR can serve as an effective secondary treatment in landfill leachate treatment and the primary step in the MBR+NF/RO combination process.

2.1.2. NF/RO Step

Despite of high removal rates of MBR process on COD, ammonia and TN, the effluent still can not meet the effluent discharge standards (GB16889-2008) as shown in Table 1 [17]. And MBR also has its limitation in a lot of trace organic pollutants [18]. Hence, NF/RO process is adopted as the tertiary treatment for MBR effluent treatment due to its acceptably high removal efficiency on inorganic molecules, total solid and residual trace organic such as persistent organic pollutants (POPs), pharmaceutical active compounds (PhACs) and hormones [18, 19].

The key mechanisms of NF and RO membrane process are similar, mainly including steric hindrance, electrostatic interactions (repulsion), and hydrophobic interactions (adsorption) between the influent and the membrane [20]. The distinction between NF and RO is the level of rejection on varies of
pollutants. The effects of NF are between UF and RO. However, on the one hand, the rejection of divalent ions and the flux of NF are relatively higher than those of RO [19]. On the other hand, RO can retain certain trace organic contaminants which NF, UF and MBR can not remove [18]. Therefore, these two membrane technologies are combined together and scientists always put NF before RO to prevent membrane fouling and maintain membrane lifespan in the real landfill leachate treatment projects in China.

2.2. Application of MBR+NF/RO Combination Process in China

2.2.1. Structure and Scale

In order to obtain a further purification for landfill leachate and meet the discharge standards of the National Municipal Waste Sanitary Landfill Pollution Control Standard, MBR and NF/RO processes are combined together in most landfill leachate treatment projects in China [3, 21-23]. In [3], Wang et al. studied the treatment of landfill leachate using MBR+NF/RO combination process with granular active carbon (GAC) as pre-treatment in Taizhou, Jiangsu. The system exhibited excellent removal of those main pollutants and low rate of membrane fouling. Zhang et al. reported the situation of a landfill leachate treatment plant in Baoding, Hebei in [21]. In this project, an anaerobic biological denitrification MBR+NF/RO was adopted for a better removal of ammonia and TN (exceeding 99%) and it is reported that the Methane produced in the anaerobic process can be used to generate electricity. In [22], a project for leachate treatment by using up-flow blanket filter (UBF) and MBR+NF/RO combination process in the south of China was studied. And the paper showed that the final effluent can meet the standards of “the reuse of urban recycling water-Water quality standard for industrial uses” (GB/T 19923—2005) and be reused to replenish circulating cooling water.

There are various structures and scales of this combination process in practical project and some of the representative project examples in China were selected to show in Table 2 [3, 5, 17, 21-33]. Despite of all kinds of treatment projects, the core steps are still the MBR+NF/RO process which undertakes the main task of removing COD, trace organic, ammonia, TN, heavy metal, inorganic salts, color and so many other pollutants in landfill leachate [29]. The flow chart of the combination process is showed in Fig. 1. And more physico-chemical and biological process can be added as pre-treatment or post-treatment for water quality stability and membrane fouling remission.

2.2.2. Removal Efficiency

Standard for pollution control on the landfill site of municipal solid waste (GB16889-2008) strictly required the discharge standards for basic pollutants in landfill leachate and the specific standards can be found in Table 3. All the treatments and structures are designed for meeting the standard so that the effluent can be recycled and reused harmlessly. As the main body of the combination process, MBR can improve its removal efficiency of hydrophobic and biodegradable trace pollutants through the adsorption between the pollutants and the mixed liquor suspended solids (MLSS) which extends the retention time in the biological tank. Then the MBR effluent with some unqualified pollutants will flow into NF/RO system and most of them will be rejected by the membrane reaction [7]. From Table 4, the removal of the main pollutants in each step can be seen clearly. In general, the removal efficiency of each step is quite high and compatible. The content of various pollutants is weakened step by step, and finally meets the emission standard.

Some data about pollutants removal efficiency of several projects in various parts of China are collected in Table 5 [3, 5, 17, 21-33]. It can be seen from the table that the leachate compositions are extremely unstable and will fluctuate with the actual situation. After comprehensive treatment of MBR+NF/RO combination process, the removal efficiency of COD and ammonia nitrogen is generally high, which can reach more than 99%. And the removal rate of TS is relatively high, even reaching 100% in [25]. This combination process can not only guarantee excellent removal effect, but also extend the efficient time through the self-optimization inside the system. Firstly, the MBR prior can reduce the membrane fouling problem of NF/RO system after precipitation and adsorption [7, 34]. Secondly, the application of NF/RO membrane system can increase the volume of water reclaimed and reduce the infiltration problem in some reusing area such as agricultural irrigation and etc. [35]
3. Future Prospects in MBR+NF/RO Combination Process in China

From various papers and data, it can be known that the MBR+NF/RO combination process has been widely applied throughout China. Nowadays, China’s land resources are becoming increasingly tense, and regional development is extremely uneven. As the main technology of landfill leachate treatment in China, it has the advantage of smaller footprint, higher effluent quality and lower operating costs, which is very suitable for China's national conditions.

MBR technology has matured in engineering practice. The future research is mainly focused on the optimization of its processing effects, such as the following points. Firstly, membrane fouling is still the main problem affecting the treatment effect and membrane lifespan. Thus, how to eliminate membrane fouling more effectively should be studied further. Secondly, research on energy conservation and cost reduction should be encouraged. For example, methane produced during the anaerobic stage of MBR can be used to generate electricity to provide the pressure needed for RO step. Furthermore, the treatment of leachate concentrates formed during treatment should draw people’s attention. The current treatment methods mainly include recharging advanced oxidation, etc., which are highly cost and harmful to the environment. Therefore, harmless treatment and recycling of leachate concentrate will be the focus of future research.

![Flow chart of MBR+NF/RO combination process](image)

**Table 1.** Characteristics of the Raw Leachate and MBR Effluent. [17]

|                         | Color  | COD \(_{c}\) (mg/L) | \(\text{NH}_4^+\)-N (mg/L) | Conductivity (mS/cm) |
|------------------------|--------|---------------------|--------------------------|----------------------|
| Raw Leachate           | Black  | 4670-6700           | 820-960                  | 14.9-15.3            |
| MBR Effluent           | Brown  | 568-850             | 0-2                      | 0.78-11.4            |
| GB16889-2008\(^a\)     | Brown  | 100 (60)            | 25 (8)                   | -                    |

\(^a\) Standard for Pollution Control on the Landfill Site of Municipal Solid Waste. The data shown in the bracket is the first-class standard.

**Table 2.** Basic situation of landfill leachate treatment project examples in China.

| Start year | Landfill site | Scale (m\(^3/d\)) | Process                  | Cost (yuan/m\(^3\)) | Ref. |
|------------|---------------|--------------------|--------------------------|----------------------|------|
| -          | Taizhou       | 1000               | A/O-GAC-MBR+NF/RO        | -                    | [3]  |
| 2005       | Shanghai      | 950                | A/O-MBR                  | -                    | [5]  |
| 2005       | Tianjin       | -                  | MBR+NF                   | -                    | [17] |
| 2001       | Baoding       | 150                | A/O-MBR+NF/RO            | 23.24                | [21] |
| 2013       | Suzhou        | 700                | MBR+NF/RO                | 30                   | [22] |
| -          | -             | 200-300            | UBF\(^a\)+MBR+NF/RO      | -                    | [23] |
| 2003       | Chengdu       | 1300               | MBR+RO                   | 25                   | [24] |
| -          | Qingdao       | 150                | MBR+NF/RO                | -                    | [25] |
| 2009       | Tengzhou      | 80                 | MBR+NF/RO                | 20-28                | [26] |
| -          | Changzhou     | 247                | MBR+NF                   | 19.55                | [27] |
| -          | Penglai       | 120                | MBR+NF/RO                | -                    | [28] |
| 2001       | Tianjin       | 200                | MBR+NF/RO                | 36.5                 | [29] |
| 2014       | Yichang       | 200                | TMBR\(^b\)+NF/RO         | 29.5                 | [30] |

\(^a\) and \(^b\) stand for treatment methods.
Table 3. Standard for pollution control on the landfill site of municipal solid waste, China, GB16889-2008.

| Paramate        | Emission limit value |
|-----------------|----------------------|
| Chromaticity (times) | 40                   |
| COD$_c$ (mg/L)   | 100                  |
| BOD$_3$ (mg/L)   | 30                   |
| SS (mg/L)        | 30                   |
| NH$_3$-N (mg/L)  | 25                   |
| TN (mg/L)        | 40                   |
| TP (mg/L)        | 3                    |
| Hg (mg/L)        | 0.001                |
| Cd (mg/L)        | 0.01                 |
| Cr (mg/L)        | 0.1                  |

Table 4. Performance of different membrane process in treating effluent. [3]

| Parameter        | MBR  | NF$^a$ | RO$^b$ | Std$^c$ | Std$^d$ |
|------------------|------|--------|--------|---------|---------|
| COD (mg/L)       | 579.7| 276.25 | 12.39  | 100     | 60      |
| NH$_3$-N (mg/L)  | 33   | 26.55  | 2.6    | 25      | 10      |
| Chromaticity (times) | 128 | 8      | 1      | 40      | 30      |
| Conductivity (μS/cm) | 4510| 3740  | 134.22 | -       | -       |
| Cd (mg/L)        | 0.047| ND     | ND     | 0.01    | -       |
| Cr (mg/L)        | 0.17 | ND     | ND     | 0.1     | -       |
| Pb (mg/L)        | 0.12 | ND     | ND     | 0.1     | -       |
| Cl$^-$ (mg/L)    | 1233.9| 685.0 | 78.6   | -       | 250     |
| SO$_4^{2-}$ (mg/L) | 2334.6| 166.6 | 55.3   | -       | 250     |

ND: non-detectable.
$^a$ Operated under 1.4 MPa.
$^b$ Operated under 2.8 MPa.
$^c$ Standard for pollution control on the landfill site of municipal solid waste, China, GB8978-1996
$^d$ Reuse of recycling water for urban, water quality standard for industrial water consumpt-ion, China, GB/T 19923-2005

Table 5. Treatment effectiveness of landfill leachate treatment projects in China.

| Landfill site | COD (mg/L) | BOD (mg/L) | pH  | NH$_3$-N (mg/L) | SS (mg/L) | Removal (%) | Ref. |
|---------------|------------|------------|-----|----------------|-----------|-------------|------|
| Taizhou       | 3134.88    | 450        | 7.85| 434.76         | -         | 99COD 99NH$_3$-N | [3]  |
| Shanghai      | 15000-20000| 7000-8000  | 7-8.5| 2800-3300     | 1000-2000 | 94.2COD 95NH$_3$-N | [5]  |
| Tianjin       | 4670-6700  | -          | -   | 820-960       | -         | 99.1COD     | [17] |
| Baoding       | 19854      | 6254       | -   | 2894          | -         | 99.7COD 99.7NH$_3$-N | [21] |

$^a$ Up-flow blanket filter.
$^b$ Tubular Membrane Bioreactor.
4. Conclusion
The objective of this research was to have a primary study on the application of MBR+NF/RO combination process in China. A large number of domestic and foreign papers have been reviewed and analysed. Several of China's standards and specific projects have also been referenced. MBR+NF/RO combination process has excellent adaptability, integrating the physico-chemical and biological treatment technology. Therefore, this treatment of all sizes can function well across China. In addition, MBR and NF/RO system can complement each other very well, reducing membrane fouling and achieving the discharge standards. And the efficiency of MBR+NF/RO process to remove COD and ammonia nitrogen is quite high and can exceed 99% in most engineering objects in China. Furthermore, the combination process in some special areas, such as the removal of antibiotics and hormones in landfill leachate, also has a good effect, which has important significance for the ecosystem. Finally, solving membrane fouling problems, energy conservation issues, and concentrate problem should be the direction of future research. Expectantly, this paper is comprehensive to determine the sufficient information and as a reference for further data collection to study landfill leachate treatment situation in China.

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