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Synergizing MBR and MCDI systems as a sustainable solution for decentralized wastewater reclamation and reuse

Wang-Sheng Yu[1], Huei-Cih Liu[1], Hsin-Chieh Lin[1], Mengshan Lee[2] and Chia-Hung Hou[1*]

Abstract

Decentralized wastewater reclamation and reuse systems have drawn much attention due to their capability for reducing the energy demand for water conveyance and reclaiming wastewater for local re-use. While membrane bioreactor (MBR) stands as a mature technology offering comprehensive solid and liquid separation, membrane capacitive deionization (MCDI) presents a promising avenue for ion separation. Unfortunately, MCDI has seldom been incorporated into decentralized wastewater reclamation and reuse systems. This study aims to exemplify the design and the operation of the synergistic integration of MBR and MCDI system with a practical capacity of 1 m^3 d^{-1}, showcasing its efficacy in reclaiming and reusing water at regional level. The integrated system demonstrated significant high removal of total organic carbon (from 97 to 2 mg L^{-1}) and chemical oxygen demand (COD, from 218 to <3 mg L^{-1}). Meantime, nearly complete transformation (approximately 91%) of NH_3 to NO_3^- within the MBR effluent was observed with a hydraulic retention time of 4.3–4.8 h and a food-to-microorganism of 0.15–0.20 kg COD kg^{-1} MLSS d^{-1} which can be further removed through the MCDI system (>92% TN removal). A significant milestone of MCDI unit was reached with the remarkable removal efficiency of total ions (93%) and water recovery (80%) using a stop-flow regeneration approach coupled with an optimized voltage of 2.0 V. The MCDI unit not only proved its high stability but also featured low energy consumption (0.44 kWh m^{-3}). Overall, synergizing MBR and MCDI systems emerges as a sustainable and effective solution for decentralized wastewater reclamation and reuse, contributing to a more environmentally friendly and resource-efficient water management paradigm.

Keywords Decentralized wastewater reclamation, Membrane bioreactor, Membrane capacitive deionization, Energy efficiency, Ion removal, Nitrogen removal

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1 Introduction

A well-functioning wastewater system is the core of water resource management, which is regarded as a promising strategy to alleviate the pressure on water supply through water reclamation and reuse [1]. Water reclamation and reuse have been demonstrated to be feasible for agricultural, industrial, and environmental applications [2], but they still suffer from intensive energy requirements during the treatment processes. As the interdependence between water and energy (i.e., water-energy nexus) is well recognized for the wastewater system [3], research efforts are being made to explore sustainable strategies that provide energy and environmental benefits for wastewater reclamation in order to address both energy and water challenges.

Decentralized wastewater reclamation and reuse systems are generally considered to be a sustainable solution from both economic and environmental perspectives [4]. One main advantage of decentralized system is the possibility of reusing treated water with relatively low resource requirements for the water supply network [5]. For instance, Kavvada et al. [6] reported that decentralized non-portable water reuse could have relatively lower energy requirements and associated greenhouse gas emissions compared to centralized ones. Risch et al. [7] also declared that the environmental impacts of decentralized systems are positively correlated with their service size (number of household or person-equivalents), but decentralized systems still offer preferable environmental performance compared to centralized ones, particularly in terms of human health and resource impacts resulting from reduced resource use in infrastructure and energy consumption. That being said, a major challenge to the implementation of a decentralized wastewater treatment system remains in its applicability and selection of adopted technologies [8]. Previous studies also highlighted that the required treatment level for intended users is the most important factor for the selection of adopted technologies, that is, the adopted technologies may vary among different reuse applications [9, 10].

Membrane-based treatment technologies, including membrane bioreactor (MBR) and membrane capacitive deionization (MCDI), have emerged as efficient technologies for wastewater treatment and reclamation for decentralized systems. MBR is advantageous in removing organic substances and suspended solids (SS) through the principle of membrane filtration, which provides nearly complete solid and liquid separating during treatment [11, 12]. MCDI, on the other hand, has been demonstrated to be practical for wastewater reclamation with high efficient desalination through electrosorption and ion exchange mechanisms [13, 14]. Previous studies have shown that the MBR was commonly integrated with advanced oxidation processes (AOPs) or reverse osmosis [15], in order to improve their treatment efficiency. In fact, MBR offers unique opportunities in terms of lower chemical oxygen demand (COD) and SS in effluents and thus could enhance the desalination performance of MCDI. Our previous study has verified the high feasibility of reclaiming MBR effluent using scaled-up MCDI (40 pairs of electrodes with effective surface area of 3.2 m^2), contributing to nearly 83–94% of total ion removal [13]. Nevertheless, to the authors' knowledge, little work has been done to integrate MBR with MCDI systems at full scale for decentralized purpose, indicating the need to understand their capability and stability in enhancing wastewater reclamation performance for further reuse.

This study aims to understand synergic effects for integrating MCDI and MBR systems as a sustainable solution for decentralized wastewater reclamation and reuse. The study's primary focus is on investigating the technical performance for the systems, specifically for improving water quality and associated energy use efficiency, for practical implementation at decentralized level. Attention is also paid to water characteristics at points of influent and effluent and their influence on the long-term operation. The results presented in this study is expected to support planning for decentralized water reclamation infrastructure with practical solution for adopting technology.

2 Methods

2.1 System design and set-up

This study proposes the integration of MBR-MCDI system with a capacity of 1 $m^3\ d^{-1}$ for decentralized wastewater treatment and reclamation. The system consisted of an influent tank, two magnetic drive pumps (MD-70RZ, Iwaki Co. Japan), a MBR module, a MCDI module, a UV module, and an effluent tank (Fig. 1a). The design capacity for the system can be categorized as a small-sized system, which should not exceed service for 50 person-equivalents [16, 17], based on the estimation of per capita wastewater production for Taiwan (East Asia) at approximately 0.14 $m^3\ d^{-1}$ $capita^{-1}$ (51.5 $m^3\ yr^{-1}$ $capita^{-1}$) [18, 19]. It is worth noting that future practical applications of the system can involve utilizing one to several units for individual onsite system or to a serious of larger clusters [20, 21].

The MBR had a working volume ranging from 180 to 200 L and housed 5 membrane modules within the reactor. The membrane modules were arranged in a parallel configuration and each module utilized flat-sheet microfiltration membranes (FS, KUBOTA, Japan) with a pore size of 0.2 μm. The hydrostatic pressure during the operational term was maintained at 20 kPa, and the effective membrane surface area was 4 m^2. Additionally, the reactor was inoculated with activated sludge from a municipal wastewater treatment plant in Tamsui, Taiwan, and

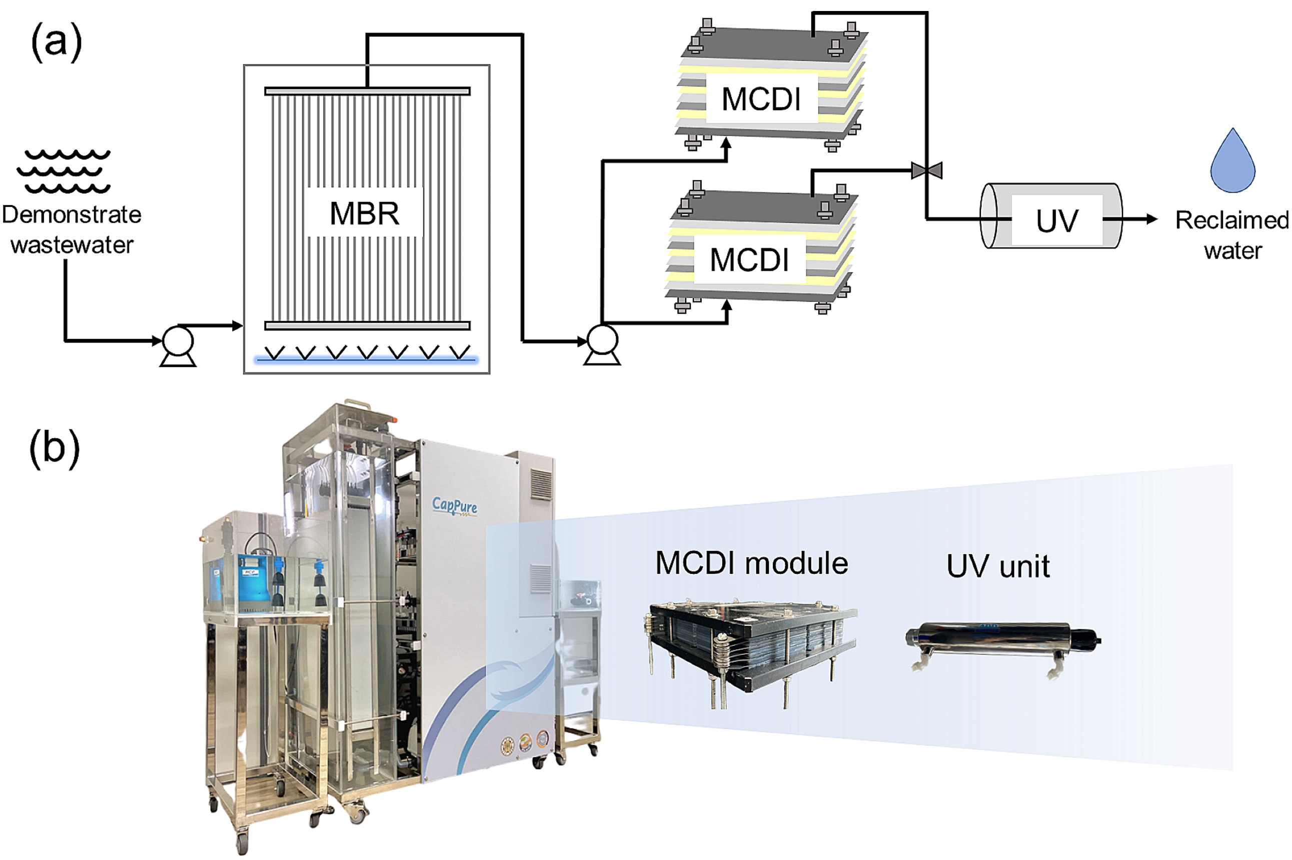

Fig. 1 **(a)** Schematic diagram of the pilot-scale integrated system, **(b)** On-site photos of the integrated water treatment system used in field

Table 1 Operation parameters of MF-MBR

Parameters	Value
MLSS (mg L^{-1})	6000–8000
HRT (h)	4.32–4.80
F/M (kg COD kg^{-1} MLSS d^{-1})	0.15–0.2
DO (mg L^{-1})	2–4
pH	6–8

was continuously fed with synthetic wastewater. To prevent overflow, a water level sensor was attached to the system. Aeration in the reactor was meticulously controlled by managing the airflow at a rate of 75 LPM. The hydraulic retention time (HRT), dissolved oxygen (DO), and pH were kept at 4.32–4.80 h, 2–4 ppm, and 6–8, respectively. Additionally, the food-to-microorganism (F/M) ratio is maintained at a moderate level of 0.15–0.20 kg COD kg^{-1} MLSS d^{-1}, corresponding to the typical range for biological treatment facilities for household sewage [22, 23]. The operating parameters of the MBRs was summarized in Table 1.

The MCDI module comprised two parallel MCDI units, each containing 6 pairs of electrodes stacked in series. Each electrode consisted of a titanium body sheet coated with activated carbon. Both sides of electrode were then covered with ion-exchange membranes giving an effective area of 20×40 cm^2. A power supply was used for controlling voltage and monitoring current through the MCDI operation. The operation of MCDI was divided in to three steps: charging step, regeneration step, and discharging step. To achieve a high water recovery of 80% and ensure complete desorption of ions from the electrodes, the regeneration step involved open-circuit discharge during the first half and reverse-voltage discharge during the second half under stop-flow conditions. Simultaneous detection of electrical conductivity (EC) and pH value were conducted by EC meter (EC-410, Suntex, USA) and pH meter (PC-310 A, Suntex, USA), respectively.

2.2 Characterization of water quality

This study utilized synthetic wastewater, as detailed in Table 2, for system performance measurement, and the wastewater primary includes nutrients of CH_3COONa, NH_4Cl and KH_2PO_4, as well as some trace nutrients such as $CaCl_2$ and $MgSO_4{\cdot}7H_2O$ [24]. To ensure that the pH remained within the target range of 6 to 8, $NaHCO_3$ was introduced at a concentration of 25 mg L^{-1}. However, it is worth noting that slight fluctuations in COD and NH_4^+ concentration occurred due to the degradation of organic matter in the influent tank.

Table 2 Characteristics of synthetic wastewater used in the study

Constituents	Concentration (mg L^{-1})
CH_3COONa	315
NH_4Cl	118
KH_2PO_4	36
$NaHCO_3$	41
$MgSO_4{\cdot}7H_2O$	63
$CaCl_2$	49

Total organic carbon (TOC) was analyzed using the TOC-L series analyzer (Shimadzu, USA). For the assessment of COD, a colorimetric method was employed, using a spectrophotometer (DR 3900, HACH, USA). Nessler method (Hach method 8038) was used to measure the concentration of NH_4^+. The concentrations of cations and anions were quantified using inductively coupled plasma-optical emission (700 series, Agilent Technologies, USA) and ion chromatography (ICS-1100, Thermo Fisher, USA), respectively, for most of the studied ions in Table 3. In addition, the determination of mixed liquor SS (MLSS) adhered to the standard set forth by the Taiwan EPA NIEA W210.58 A.

2.3 Performance metrics

The performance of the MCDI system was assessed using a comprehensive set of metrics [13, 14], including mean deionization capacity (MDC), mean deionization rate (MDR), water recovery (WR), productivity (P), charge efficiency (η), and energy consumption (E_v). These metrics were calculated using following equations:

$$MDC\ (\text{mg g}^{-1}) = \frac{M \times \varphi \times \int_0^{t_c}(C_0 - C_{t_c})\text{d}t}{m} \tag{1}$$

$$MDR\ (\text{mg g}^{-1}\ \text{min}^{-1}) = \frac{MDC}{t_c} \tag{2}$$

$$WR\ (\%\ ) = \frac{t_c}{t_c + t_d} \tag{3}$$

$$\text{P}\ (\text{L h}^{-1}\ \text{m}^{-2}) = \frac{t_c \times \varphi}{t_t \times n \times \text{A}} \tag{4}$$

$$\eta\ (\%\ ) = \frac{\text{F} \times \varphi \times \int_0^{t_c}(C_0 - C_{t_c})\text{d}t}{\int_0^{t_c} I\text{d}t} \tag{5}$$

$$E_\text{v}(\text{kWh m}^{-3}) = \frac{V \times \int_0^{t} I\text{d}t}{\varphi \times t} \tag{6}$$

$$t_\text{t} = t_c + t_r + t_d \tag{7}$$

where M is the molecular weight of salt (g mol^{-1}); ϕ is the volumetric flow rate (mL min^{-1}); C_0 and C_{tc} represent the influent and effluent concentrations (mM), respectively; m is the weight of the activated carbon electrodes (g); t_c is the duration of charging stage (min); t_r is the duration of regeneration stage (min); t_d is the duration of discharging stage (min); t_t is the total time of a cycle (min); n is the number of electrode pairs; A is the effective surface area of a pair of electrode (cm^2); F is the Faraday's constant (96,485 C mol^{-1}); V is the applied voltage (V); and I

Table 3 Water quality of effluent for MBR and MCDI in comparison to standards of NEWater and Tainan wastewater reclamation plan

Parameters	Unit	MBR influent	MBR effluent	MCDI effluent	NEWater [a]	Tainan reclaimed water standard
Physical parameters						
Conductivity	μS cm^{-1}	1021	988	122	< 250	< 250
pH value	-	6.5	6.4	8.2	7.0–8.5	6.0–8.5
Chemical Parameters						
Ammonia-N	mg L^{-1}	40	0.9	0.1	< 1.0	< 0.5
Nitrate-N	mg L^{-1}	0.3	37.5	3.0	< 5	N.A.
TOC	mg L^{-1}	97	1.8	2.0	< 0.5	< 1.0
COD	mg L^{-1}	218	< 3.0	< 3.0	N.A.	< 4
Total Hardness (as $CaCO_3$)	mg L^{-1}	62	60	8.0	< 50	< 50
Calcium	mg L^{-1}	14	14	2	< 20	N.A.
Magnesium	mg L^{-1}	6.2	5.8	0.6	N.A.	N.A.
Potassium	mg L^{-1}	10	10	0.8	N.A.	N.A.
Sodium	mg L^{-1}	99	96	16	< 20	N.A.
Chloride	mg L^{-1}	127	120	11	< 20	N.A.
P	mg L^{-1}	17	15	2.2	N.A.	N.A.
Sulphate	mg L^{-1}	21	20	4	< 5	< 10

N.A.: not applicable

[a] Source: PUB (2023)

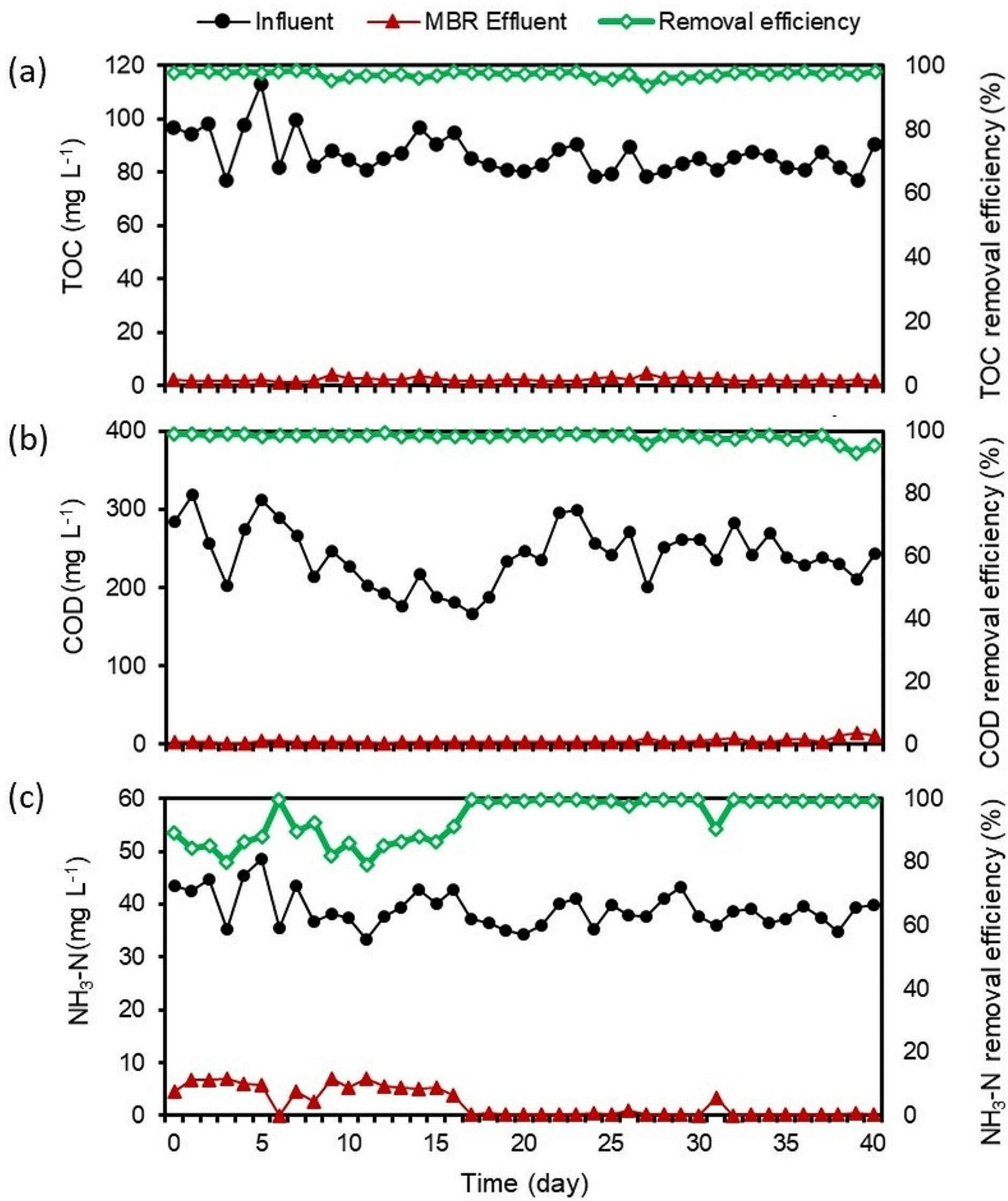

Fig. 2 Long-term stability performance of MBR system as recorded in influent and effluent concentration and removal efficiency: **(a)** TOC, **(b)** COD, and **(c)** NH_3-N

is the corresponding current (A) measured during MCDI operation.

3 Results and discussion

3.1 Long-term performance of the MBR unit

The concentration profiles of TOC and COD in the MBR are presented in Fig. 2a and b. The influent

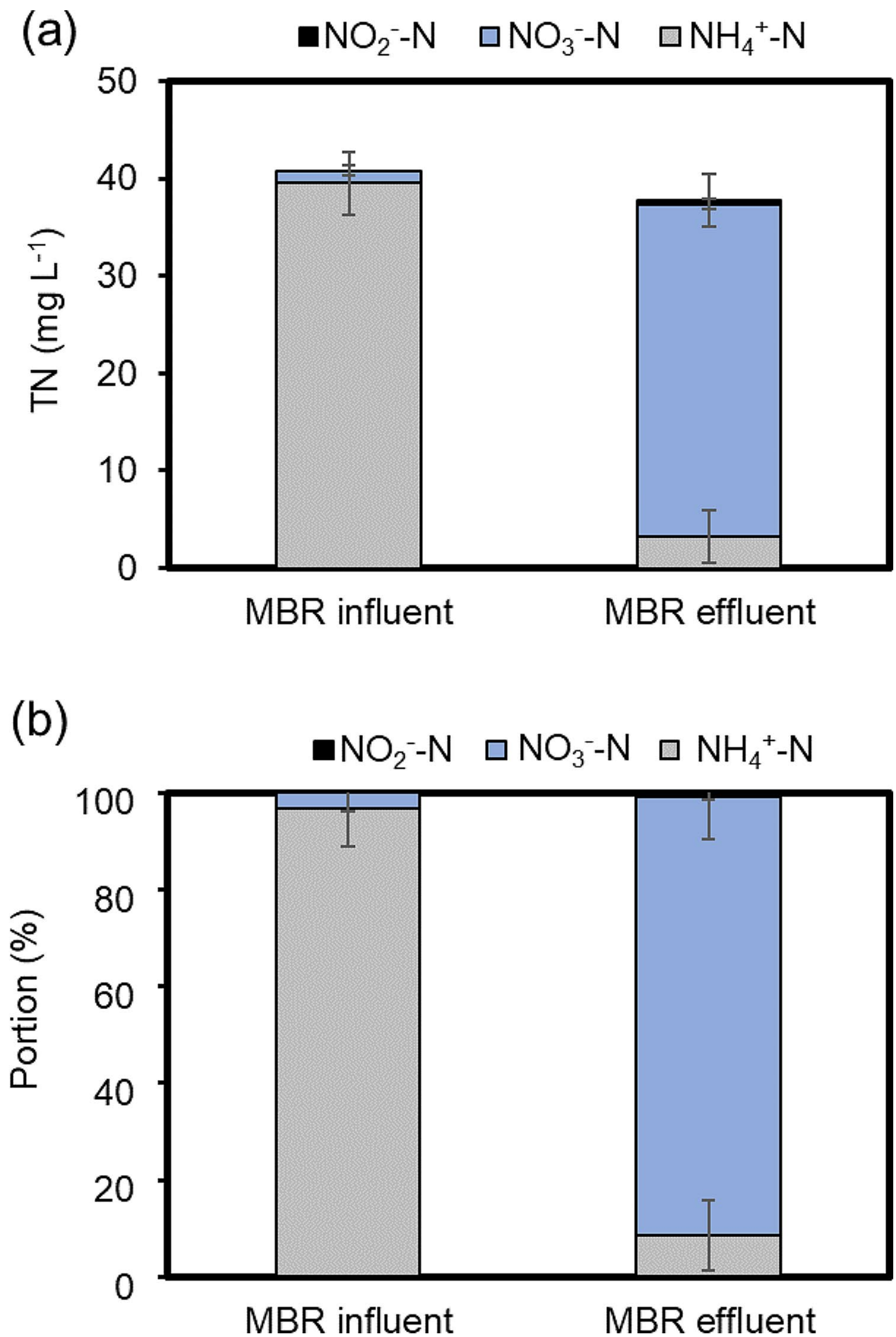

Fig. 3 **(a)** Distribution and **(b)** portion of N species in both the influent and effluent of the MBR

concentrations of TOC and COD were around 86.7±7.5 mg L^{-1} and 241.1±37.2 mg L^{-1}, respectively. The concentration of COD was approximately 2.8 times higher than that of TOC. This could be attributed to the presence of CH_3COONa as primary carbon source in the synthetic wastewater, which has a theoretical COD/TOC ratio of 2.7. After the biological treatment within the MBR, the concentrations of COD in the permeate exhibited a significant decrease, falling below the detection limit of 3 mg L^{-1} (Table 3). Moreover, the concentration of TOC experienced a similar decreasing trend as that of COD, reaching a level of 2.3±0.7 mg L^{-1} with a consistently high removal efficiency of above 93%. These remarkable removal efficiencies can be attributed to the

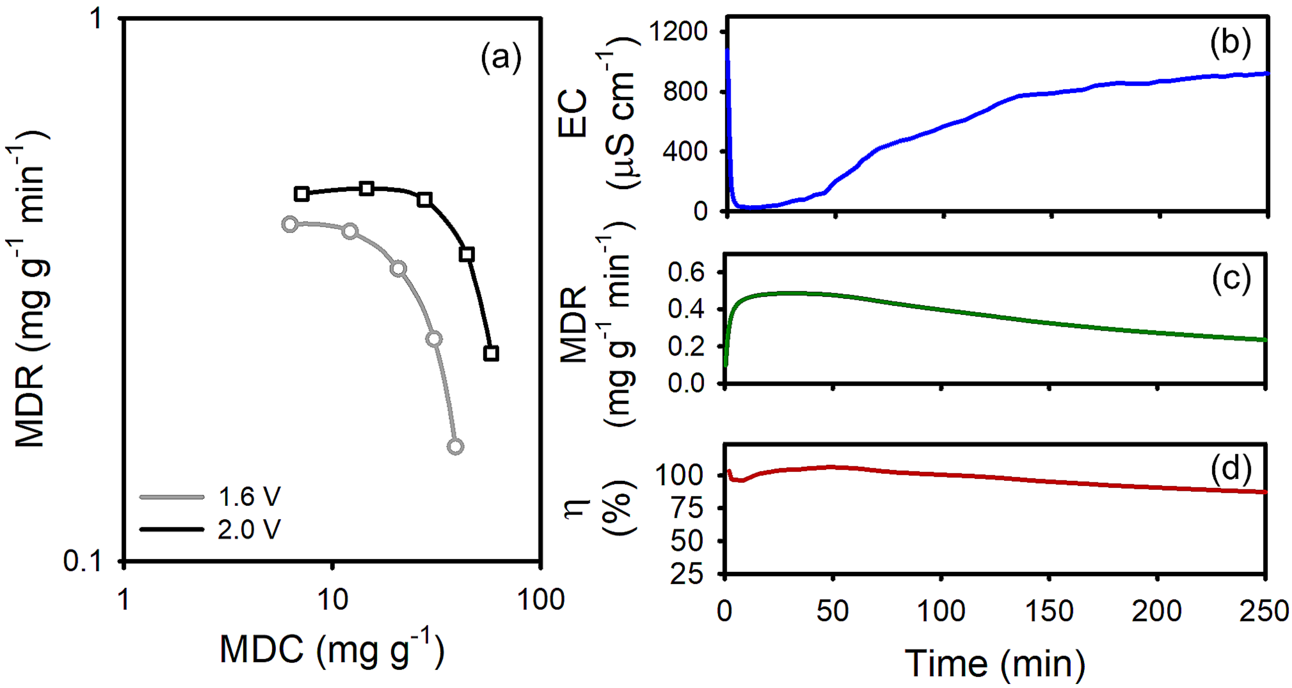

Fig. 4 **(a)** Variation of CDI Ragone plots according to the applied voltage and performances of MCDI system using MBR effluent at 2.0 V with flow rate of 350 mL min^{-1}: **(b)** EC, **(c)** MDR, and **(d)** η

advantages of MBR, which promote aerobic organic oxidation, facilitated by a high MLSS concentration in the range of 6000–8000 mg L^{-1}. Following the initial 15 days of operation, an improvement in the nitrification performance of the MBR unit was observed. Starting on day 16, favorable NH_3-N removal was attained, with an average of 0.5±0.9 mg L^{-1} in the effluent with a removal efficiency exceeding 91% for the subsequent 25 days (Fig. 2c). This reduction can be ascribed to the efficient nitrification performance via controlling DO in the range of 4–6 mg L^{-1}, coupled with the preservation of suspended microbial biomass.

Changes in the distribution of N species in the effluent is shown in Fig. 3. It's noteworthy that while the NH_3-N concentration exhibited a significant decrease, the MBR process resulted in only a marginal reduction in total nitrogen (TN). A significant share of NH_3-N was transformed into NO_3^- and NO_2^-, as shown in Fig. 3a. In the MBR effluent, the NO_3^- concentration is measured at 34±3 mg L^{-1}, constituting 91% of all nitrogen-containing species, as shown in Fig. 3b. Hence, the integration of MBR with MCDI is deemed imperative for effective removal of ionic species, thus ensuring compliance with water reclamation and reuse standards.

3.2 Operation optimization of the MCDI unit

Different applied voltages (1.6 and 2.0 V) were employed to investigate their effect on the desalination performance while treating MBR effluent at a flow rate of 350 mL min^{-1}. As shown in Fig. 4(a), CDI-Ragone plot shifted towards the top-right corner as the applied voltage increased from 1.6 to 2.0 V, illustrating a significant augmentation in both MDC and MDR. Hence, 2.0 V was employed as the optimum voltage condition for the MCDI modules in this study. Figure 4b depicted the variation of EC within the MCDI modules for 250 min of charging. The operation of MCDI at 2.0 V achieved a desirable product water quality which met the regulations (EC $<$ 250 μS cm^{-1}), under the designed treatment capacity of 1 m^3 d^{-1} for decentralized applications, as shown in Table 3.

To dive deeper into the MCDI performance, MDR and time relation curves shown in Fig. 4c could be useful to understand the kinetics during charging process. It was found that the MDR achieved the highest value of 0.49 mg g^{-1} min^{-1} at 2 V within the first 30 min. After 30 min, the MDR began to gradually decreased, indicating that it was no longer kinetically favorable for the ion removal at this stage of charging process. The charge efficiency in Fig. 4d consistently reached at least 87% during charging. However, similar to the MDR, the charge efficiency displayed a decline after initially surpassing 99% in the early charging stage. Based on the information regarding EC of effluent, MDR, and charge efficiency, a charging time of 30 min was chosen for all subsequent MCDI tests.

To minimize the brine volume and attain 80% water recovery, stop-flow regeneration approach was implemented during the optimized MCDI operation [25]. In

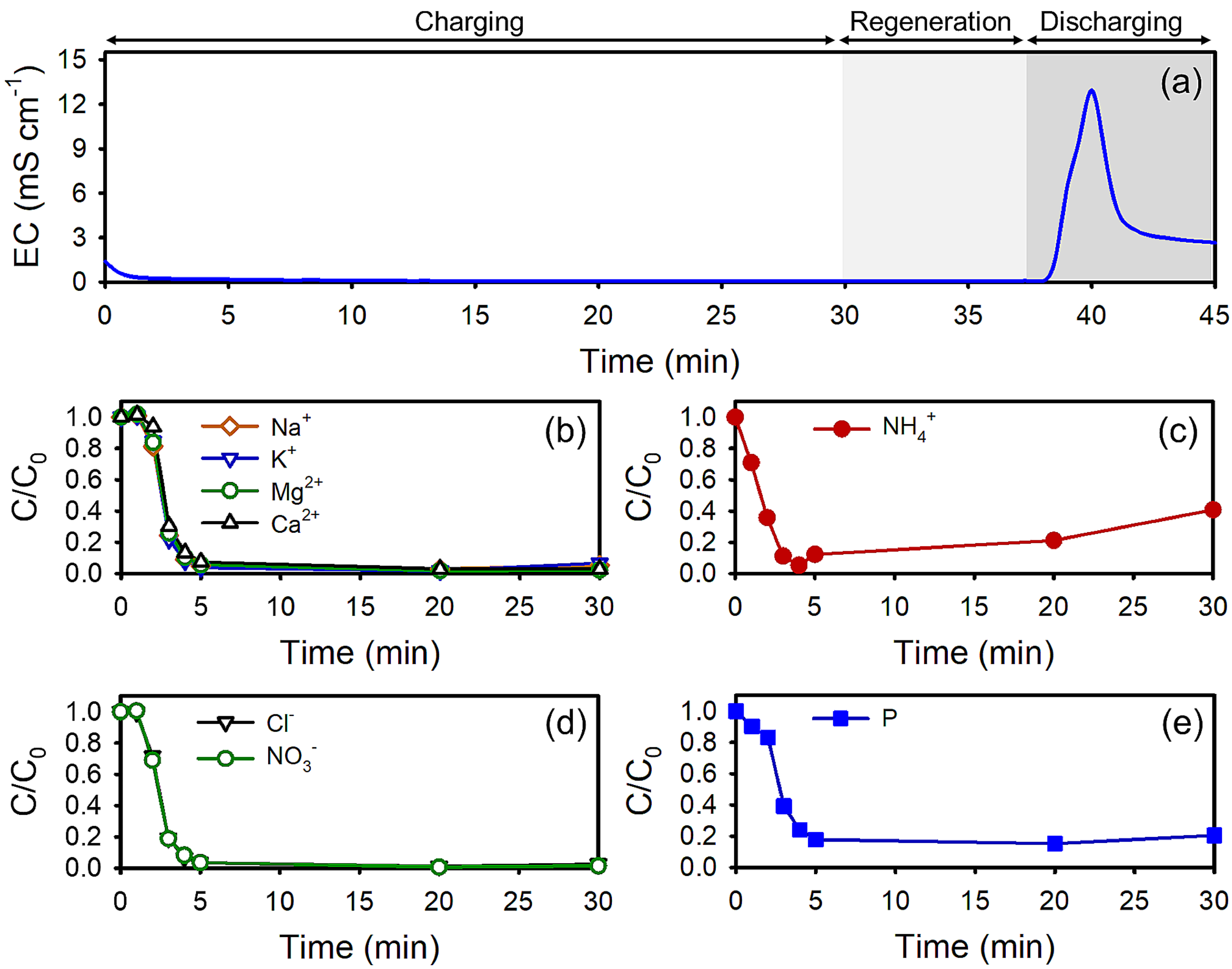

Fig. 5 MCDI system performance under optimal operation condition (2.0 V for 30 min in charging step, 0.0 V for 3.75 min followed by −2.0 V for 3.75 min in stop-flow regeneration step, and −2.0 V for 7.5 min in discharging step). Variations in **(a)** EC, **(b)** Na^+, K^+, Ca^{2+}, and Mg^{2+}, **(c)** NH_4^+, **(d)** Cl^- and NO_3^-, and **(e)** total P measured as phosphate ions

this approach, the 7.5-min regeneration step was divided into two stages: a 3.75 min duration at 0 V followed by an additional 3.75 min at −2.0 V. Regarding the discharging step, note that the longer duration can better regenerate the electrodes at the expense of productivity [26]. Thus, a potential of −2.0 V was applied for 7.5 min with a constant flow rate of 350 mL min^{-1}, yielding the productivity of 29.2 L h^{-1} m^{-2}.

The EC profile of optimized MCDI operation is illustrated in Fig. 5a. The EC decreased from an initial value of 1.07 mS cm^{-1} to the lowest point at 0.07 mS cm^{-1}, which can be attributed to the electrosorption of ions during the charging step. Subsequently, ions adsorbed within the electrodes were then released back into the solution during the stop-flow regeneration step. This release was driven by the concentration difference without voltage and further driven by the electric potential difference under −2.0 V. Following the regeneration step, the released ions were flushed out of the MCDI module in the discharging step, resulting in the highest EC value of 13 mS cm^{-1}.

The ion compositions of MCDI effluent during charging step were further investigated in details. The C/C_0 of major ions bottomed out at a mere 0.07 after a 5-min charging period, indicating an optimal removal efficiency of at least 93%, as shown in Fig. 5b and d. In the case of NH_4^+, the C/C_0 reached its nadir after a 4-min charging period and then gradually rose from 0.05 to 0.41 at the end of charging, as depicted in Fig. 5c. As for P (representing total phosphate species, specifically phosphate ions including $H_2PO_4^-$ and HPO_4^{2-}), the C/C_0 decreased from 1 to the lowest point of 0.15 at 20 min and experienced a slight increase to 0.2 by the conclusion of the charging step, as illustrated in Fig. 5e. Overall, the MCDI module demonstrated outstanding ion removal performance under the optimized operation.

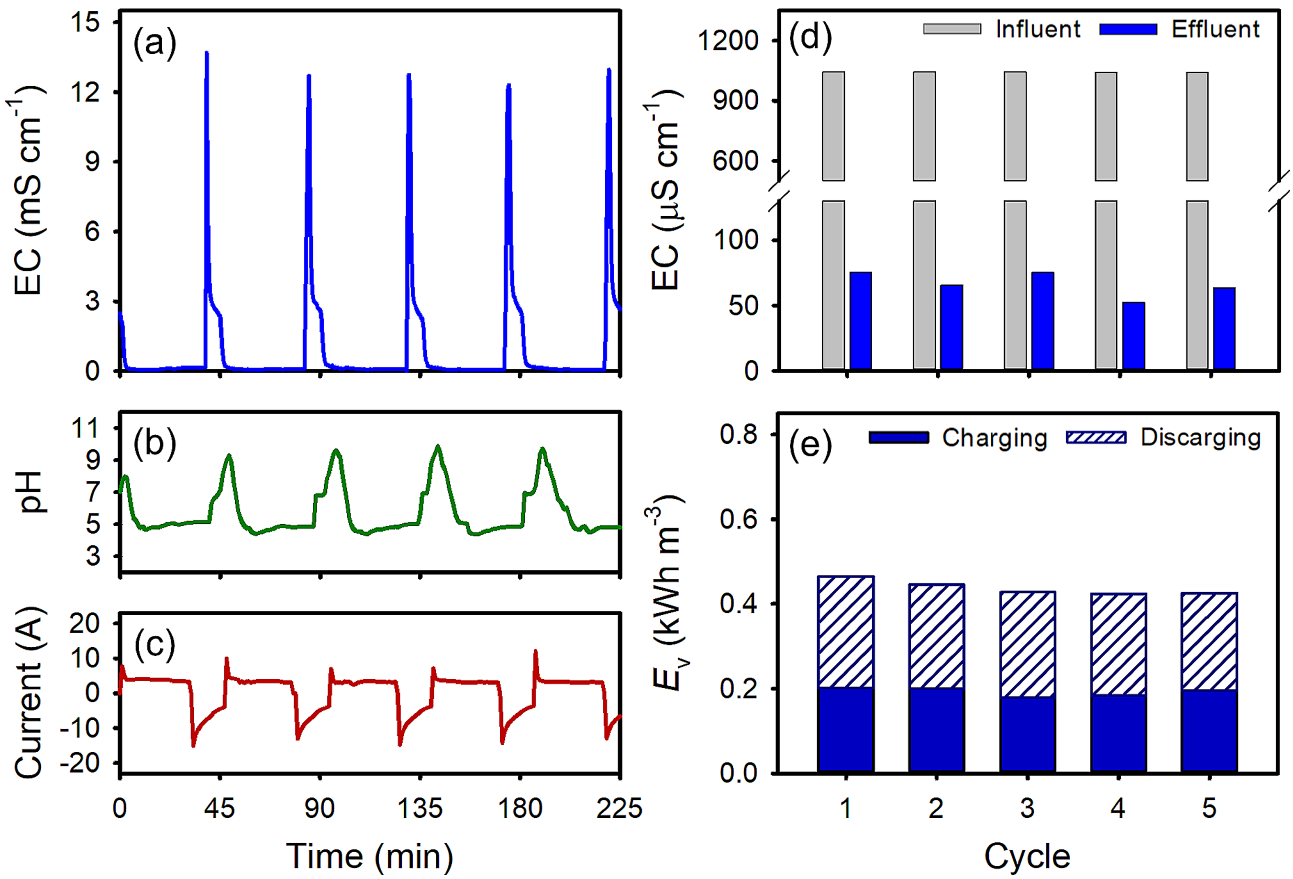

Fig. 6 System performance of MCDI experiment over 5 repeated cycles under optimal operation condition. **(a)** EC, **(b)** pH, and **(c)** current profile for 5-cycle operation. **(d)** EC of influent and effluent, and **(e)** E_v of charging and discharging step for MCDI system

3.3 Stability evaluation of the MCDI unit

The cyclic long-term operation was employed to validate the performance stability of the MCDI modules, as shown in Fig. 6. The result of EC from 5 consecutive cycles revealed a notable reduction in EC for the MCDI effluent during charging process. Conversely, the highest EC of the MCDI effluent during discharging averaged at 12.88 mS cm^{-1}, which was 12-fold higher than that of the influent. This highlighted the superior desalination and concentration performance during MCDI operation (Fig. 6a). While the pH value of the collected effluent stood at 8.2, it notably surged to 9.4 during the discharging phase (Fig. 6b). The heightened pH value, coupled with increased ion concentration, raises concerns regarding the potential precipitation of ions, posing challenges for sustained long-term operation [27]. In Fig. 6c, it is evident that the corresponding current experienced an instantaneous increase at the initial stage and then gradually decreased in both charging and discharging step.

In comparison of EC of influent and effluent after each charging-discharge cycle, the product water exhibited an average EC ranging from 53 to 75 µS cm^{-1}, demonstrating a notable 93% removal efficiency from an average EC of 1044 µS cm^{-1} in influents (Fig. 6d). Additionally, the average energy consumption was estimated at 0.44 kWh m^{-3} for the complete MCDI operation, including 0.19 kWh m^{-3} during charging and 0.25 kWh m^{-3} during discharging (Fig. 6e). The increased energy consumption during the discharging step, compared to the charging step, can be attributed to the higher current observed during the discharging process. Overall, the proposed MCDI operation demonstrated an outstanding desalination/concentration performance, achieving high water recovery (80%) while maintaining low energy consumption.

3.4 Water quality from the integrated MCDI and MBR systems

The comprehensive water quality characterization for each unit of the integrated system were detailed in Table 3, which incorporated the NEWater standard [28] and the standards for reclaimed water in Taiwan for comparison.

It was evident that the water quality for the proposed wastewater reclamation system was significantly enhanced coupling the advantages of the MBR and the MCDI, which can be effectively employed in removing organics, nutrients, and ions. It is noteworthy that, upon integration the MBR unit with the MCDI unit, the system demonstrated a relatively high TN removal efficiency exceeding 92%, thereby overcoming the limitation of the MBR unit, which primarily facilitates nitrogen species transformation without TN reduction [29]. Overall, the water quality results revealed that, from the system influent to the effluent, the NH_4^+-N decreasing form 39.6 to 0.1 mg L^{-1}, the TOC concentration declining from 97 to 2 mg L^{-1}, and the total dissolved solids concentration dropping from 350 to 40 mg L^{-1}. Despite a 98% TOC removal was obtained through the MBR process, a slight elevation of TOC was reported in the effluent after MCDI processing, which fails to meet the standards of both Newater (<0.5 mg L^{-1}) and Tainan reclaimed water (<1.0 mg L^{-1}). This result could be attributed to the presence of slowly/non-biodegradable organic pollutants, implying the requirement for implementing AOPs as post-treatment for further reduction of TOC [30].

4 Conclusions

This study presents a sustainable solution through the synergistic integration of MBR and MCDI systems for decentralized wastewater reclamation and reuse. The results indicated the efficacy of MBR in treating TOC and COD, with simultaneous transformation of the NH_4^+ into NO_3^-. Subsequent to MBR treatment, the MCDI unit demonstrated proficient removal of various ionic species, resulting in a substantial reduction in conductivity. The implementation of a stop-flow regeneration approach of MCDI unit resulted in an impressive 80% water recovery. Furthermore, the high stability performance of MCDI unit was proved through cyclic operation, featuring remarkable ion removal efficiency (93%) and low energy consumption (0.44 kWh m^{-3}). After the integrating MCDI and MBR systems, the water quality met most of the standards for Newater and reclaimed water in Taiwan, except for TOC. Consequently, an AOP process is recommended to address this residual TOC. In summary, our study validated the synergized MBR and MCDI systems as a sustainable and effective wastewater treatment solution for decentralized wastewater reclamation and reuse.

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Author contributions
Wang-Sheng Yu processed conceptualization, methodology, investigation, and writing- original draft, Huei-Cih Liu contributed to data curation and methodology, Hsin-Chieh Lin contributed to data curation and methodology, Mengshan Lee participated in conceptualization, writing- reviewing and editing, and Chia-Hung Hou led the conceptualization, project administration; supervision, writing- reviewing and editing, and funding acquisition. All authors read and approved the final manuscript.

Data availability
Upon reasonable request, the corresponding author can provide access to the data produced in this study.

Declarations

Competing interests
The authors declare they have no competing interests.

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