Ref. Ares(2022)1487048 - 28/02/2022

intelWATT

D3.2 – Lab scale development, integration and optimization of the RED/MD treatment process

Project Information

Grant Agreement Number	958454
Project Full Title	Intelligent Water Treatment for water preservation combined with simultaneous energy production and material recovery in energy intensive industries
Project Acronym	intelWATT
Funding scheme	IA
Start date of the project	1st October 2020
Duration	42 months
Project Coordinator	Andreas Sapalidis (NCSR)
Project Website	https://www.intelwatt.eu

Deliverable Information

Deliverable n°	3.2
Deliverable title	Lab scale development, integration and optimization of the RED/ MD treatment process
WP no.	3
WP Leader	PoliTo
Contributing Partners	CNR-ITM; REDstack; CUT; NCSR
Nature	Demonstrator
Authors	Enrica Fontananova, Gianluca Di Profio (CNR-ITM)
Contributors	
Reviewers	Andreas Sapalidis (NCSR); Jordi Moreno, Kristan Goeting (REDstack); Ines Bettermann (CUT).
Contractual Deadline	28/02/2022 (extended deadline)
Delivery date to EC	28/02/2022

Dissemination Level

PU	Public	✓
PP	Restricted to other programme participants (incl. Commission Services)	

RE	Restricted to a group specified by the consortium (incl. Commission Services)	
CO	Confidential, only for the members of the consortium (incl. Commission Services)	

Document Log

Version	Date	Author	Description of Change
V1.0	07/02/2022	E. Fontananova	1st draft
V1.1	16/02/2022	G. Di Profio	2nd draft
V.1.2	23/02/2022	E. Fontananova	Final Draft
V2.1	27/02/2022	E. Fontananova, G. Di Profio	1st Review of the final draft
V2.2	28/02/2022	E. Fontananova	Final Review of the final draft
V2.3	28/02/2022	A. Sapalidis	Final document

Table of Contents

1 Executive Summary.. 4
2 Introduction.. 5
2.1 Ultrafiltration (UF) pre-treatment unit... 7
2.2 Reverse electrodialysis (RED) unit 8
2.3 Membrane distillation (MD) unit 12
3 References...21

1 Executive Summary

This document describes the deliverable *No.13 (D3.2) "Lab scale development, integration and optimization of the RED/ MD treatment process"* concerning the design and assembling of a lab-scale demonstrator for the evaluation and optimization of the intelWATT's case study 2 approach: valorization of a brine collector stream from mining activities in order to harvest salinity gradient power (SGP) by reverse electrodialysis (RED) and to recover deionized water by membrane distillation (MD), in an integrated system. In addition to the RED and MD unit, the setup includes a cross flow ultrafiltration (UF) lab unit for the pre-treatment of the feed streams.

Besides the mechanical components and equipment, a large number of sensors have also been integrated in the lab devices (in agreement with *D2.3 – Report on sensor requirements*), which are necessary for the determination of the Key Performance Indexes of the proposed processes (in agreement with *D2.5 – – Report on the performance indexes of the intelWATT's prototypes*).

The lab-scale demonstrator will be also upgraded with new sensors, including specialty sensors, as well as, innovative control solutions and technologies that will be progressively developed and provided by Partners during the implementation period of intelWATT's project.

2 Introduction

The intelWATT's Case Studies 2 aims to valorise waste brines from mining activities in order to recover energy and water, exploiting renewable energy resources by an integrated membrane process (Figure 1).

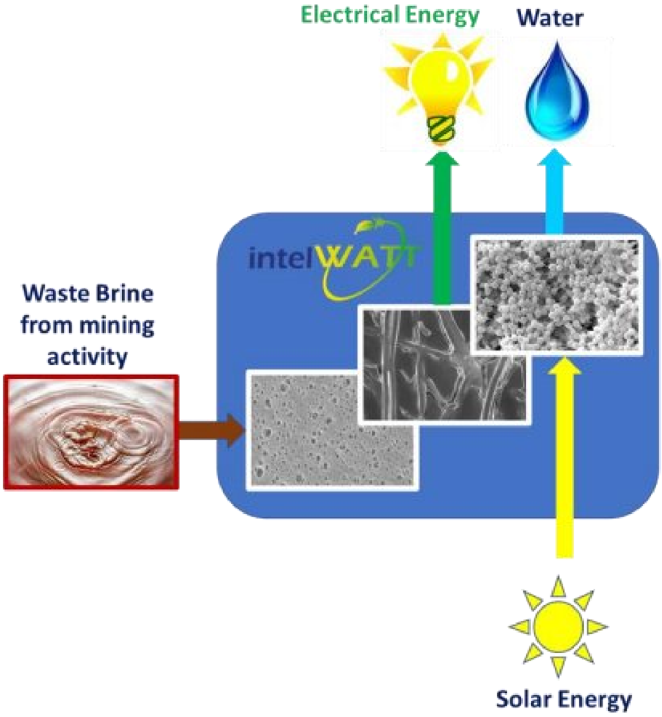


Figure 1 Approach proposed in the Case study 2

The pilot plant integrating RED and solar assisted MD processes, will be located in Castellgalí (near Barcelona, Spain), which hosts the intersection point of a brine collector which picks up the brines from different salt mining industries. ACSA has the management since 2017 of this collector. The preliminary conceptual design of the pilot plant is depicted in Figure 2.

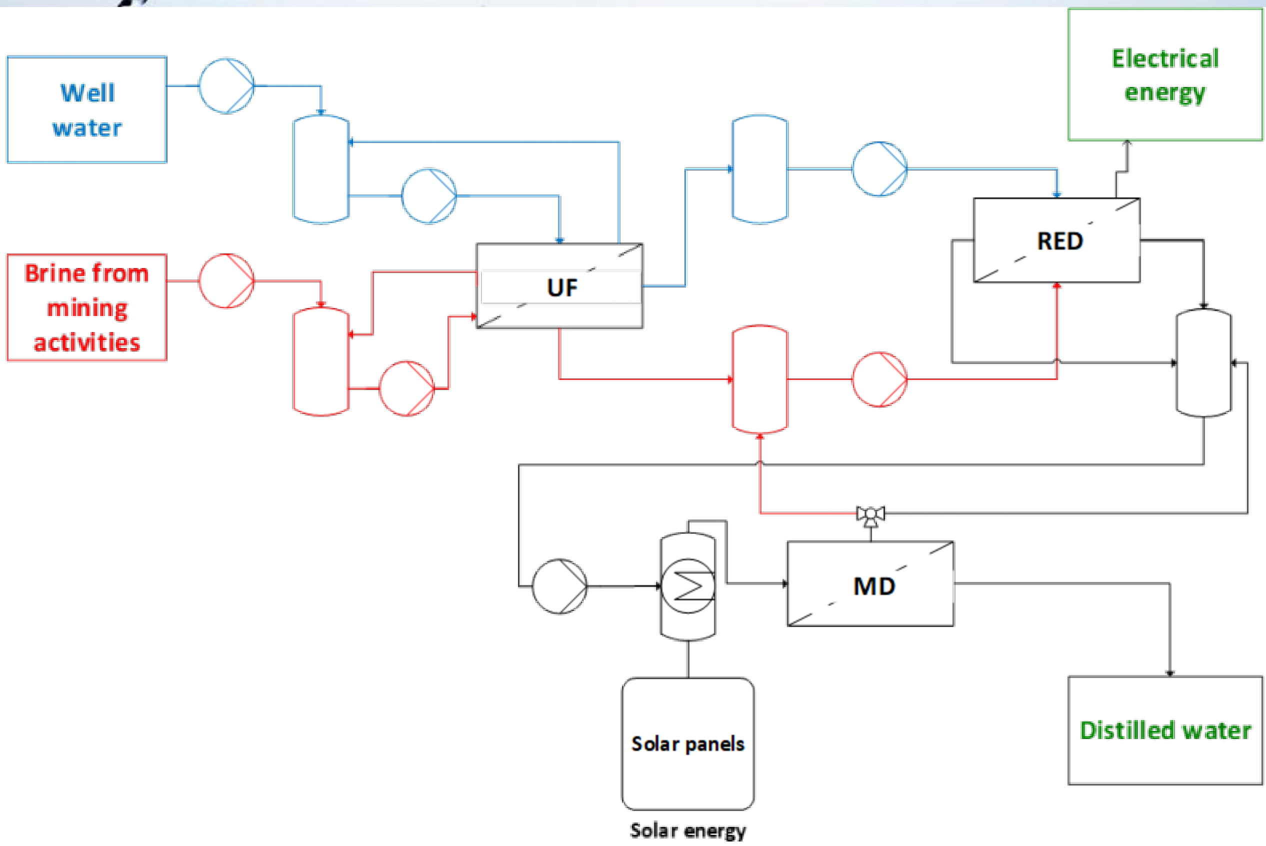


Figure 2 Preliminary conceptual design of the UF/RED/MD integrated system

The pilot plant will comprise the following sections: a) a pre-filtration unit for the reduction of the suspended solids and turbidity of the feed streams, b) a RED unit for SGP harvesting by converting the chemical potential difference between two streams with different salinity (brine from mining activity and well water) into electrical energy [1] c) a solar assisted MD unit [2] that will operate on the mixed solutions outgoing from RED in order to recover distillate water. Moreover, the concentrated from MD unit operating at high recovery factor could be potentially re-used as high salinity solution for RED in a closed loop. The integrated system will have buffer tanks between the three sections (UF/RED/MD) to comply with the different operative flow rates of each process.

In accordance with this concept, a lab scale UF/RED/MD system with buffer tanks between the three sections, was designed and assembled at CNR-ITM for evaluation and optimization of CS2 in-laboratory environment by testing membranes and modules using artificial and real solutions and investigating the effect of operative conditions on process performance. In the following sections are reported the details of each unit of the laboratory scale demonstrator.

2.1 Ultrafiltration (UF) pre-treatment unit

Ultrafiltration is a pressure driven membrane operation in which the membrane separates components of a solution predominantly according to their size and shape. CUT tubular UF membranes and membrane modules will be used for pre-treatment of the feed solution in all the three intelWATT's case studies. Moreover, a cross flow UF lab test facility was provided by CUT for each of the 3 Case Studies. For CS2 it was agreed that the unit was delivered to ACSA in the future pilot location (Castellgalí) for testing in continuous with large volumes of real solutions. Technical details about this type of lab-scale facility are reported in *D3.1 – Lab scale CTBD treatment unit*.

CNR-ITM has received from CUT tubular membranes and housing modules for lab scale testing. The material includes:

a) Two set of housing modules:
- N. 2 modules for 8mm membranes
- N. 2 modules for 5 mm membranes

b) Two different tubolar membrane-types (both 8 mm):
- N. 10 *Standard CUT membranes*
- N. 10 *IntelWATT opt.1 membranes*

The cross-flow ultrafiltration set-up was equipped with a CUT tubular membrane modules containing a single 8mm tubular membrane (Figure 3).

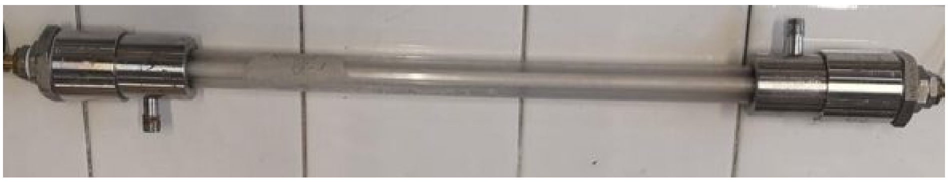


Figure 3 CUT single membrane (8mm) tubular module

The feed solution (water, artificial or real solutions) is pumped by a digital peristaltic pump (Masterflex™ digital pump drive L/S equipped with a pump head Easy-Load II Mod. 77200-50) from a tank (~20 L) to the membrane module (lumen side membrane, surface area 0.0121 m^2) with the possibility to operate in concentration mode or single passage (Figure 4)

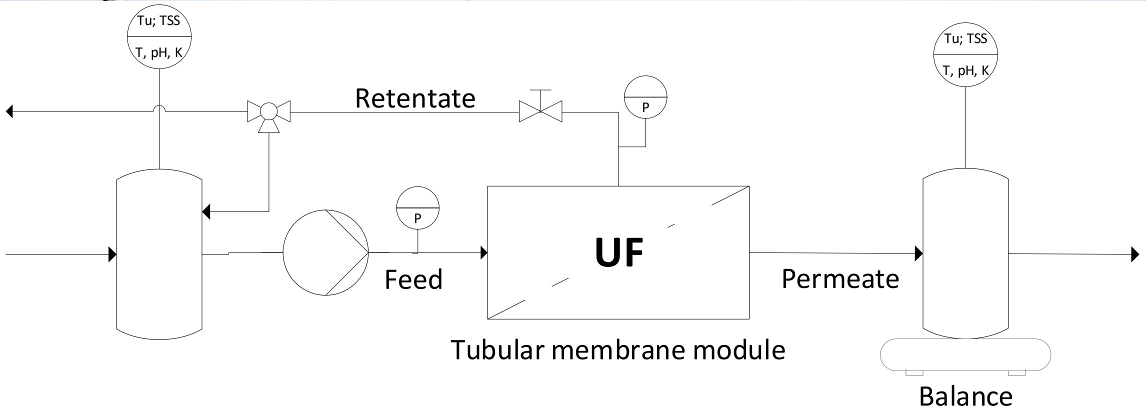


Figure 4 Scheme of the cross-flow ultrafiltration unit

Permeate is collected in a separate tank (~20 L) monitoring the mass variation in the time with a balance (ABCbilance, mod. BL/TRB30D or BL/GAM3200) while transmembrane pressure and pressure drop were monitored by digital pressure meters at the inlet and outlet of the module (PCE-910; range: ±2000 mbar; accuracy ±2% full range; reproducibility ±1%; interface RS-232) to quantify permeate flux, recovery factor, as well as, to estimate the pumping specific energy consumption (D2.5).

Turbidity (Tu) and total suspended solids (TSS) of the feed solutions have a key role on membrane fouling and membrane modules channels clogging. These parameters need to be monitored in order to have a smart management of the pre-treatment. A combined turbidity and suspended solids analyser is used to monitor the permeate quality (Hach LXV322.99.00001). The instrument uses a combined multiple beam alternating light method with infrared diode system and beam focusing. Measuring principle of the turbidity: 2-channel 90° scattered light measurement in accordance with DIN EN 27027 / ISO 7027. Measuring principle for TSS: six-channel multiple angle measurement. Measuring Range TSS: 0.001 - 400 g/L; measuring range turbidity:0.001 - 9999 FNU; Interface: RS485. The conductivity (K) and pH of the solutions are measured by a benchtop pH/conductimeter (Orion Star™ A215 pH/Conductivity Benchtop Multiparameter with ROSS Ultra Triode epoxy-body pH/ATC electrode, DuraProbe 4-cell conductivity sensor). The solutions temperature (T) is measured by a portable thermometer with microprocessor for Pt100probes (Temp 7RDT, XS Instruments).

2.2 Reverse electrodialysis (RED) unit

In a RED process, cation exchange membranes (CEMs) and anion exchange membranes (AEMs) are alternatively aligned to form a series of adjacent compartments: high concentration compartments and low concentration compartments. When the compartments are filled with the respective feed solutions, low salinity solution (LSS) and high salinity solution (HSS), an electrochemical potential is generated due to the difference in salinity gradient. When an external load/electrical consumer device is connected to the electrodes, an ion flux across membranes is initiated which is converted in electrical current mediated by redox reactions at the electrodes, resulting in controlled of the HSS and LSS (Figure 5).

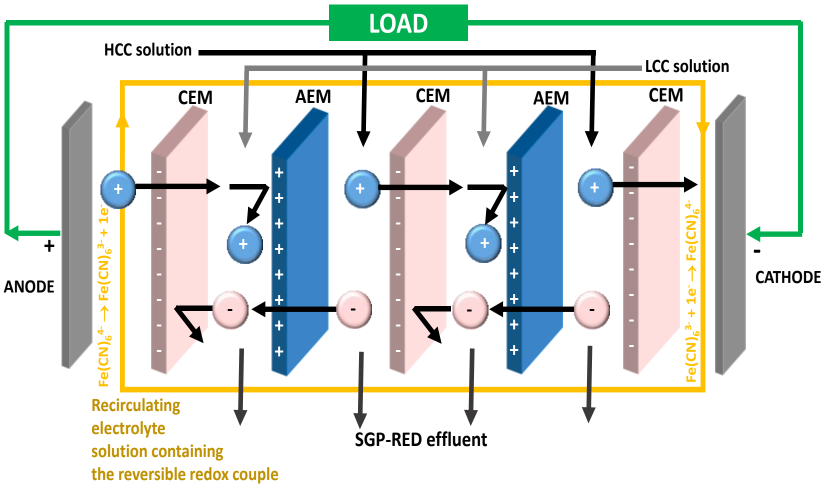


Figure 5 Operative principle of a RED process [3]

The lab-scale RED demonstrator includes a cross-flow 10x10 cm^2 RED stack manufactured and supplied by REDstack BV. This stack can host from 3 to 20 cell pairs. Currently it is assembled with 10 cell pairs (yielding a total area 0.2 m^2).

The stack is fed by cross-flow single passage of the HSS and LSS by two digital gear pumps (Digital dispensing gear pump drive mod. 75211-47 used with micropump head mod. GJ-N21.JF1S.A, Cole Parmer). An An electrode rinse solution containing a reversible redox couple (e.g. $K_4Fe(CN)_6$ /$K_3Fe(CN)_6$) is recirculated by a peristaltic pump (Masterflex™ digital pump drive L/S equipped with a pump head Easy-Load II Mod. 77200-50) from the anode to the cathode in order to compensate the electroneutrality at the electrode compartments due to the ion transport.

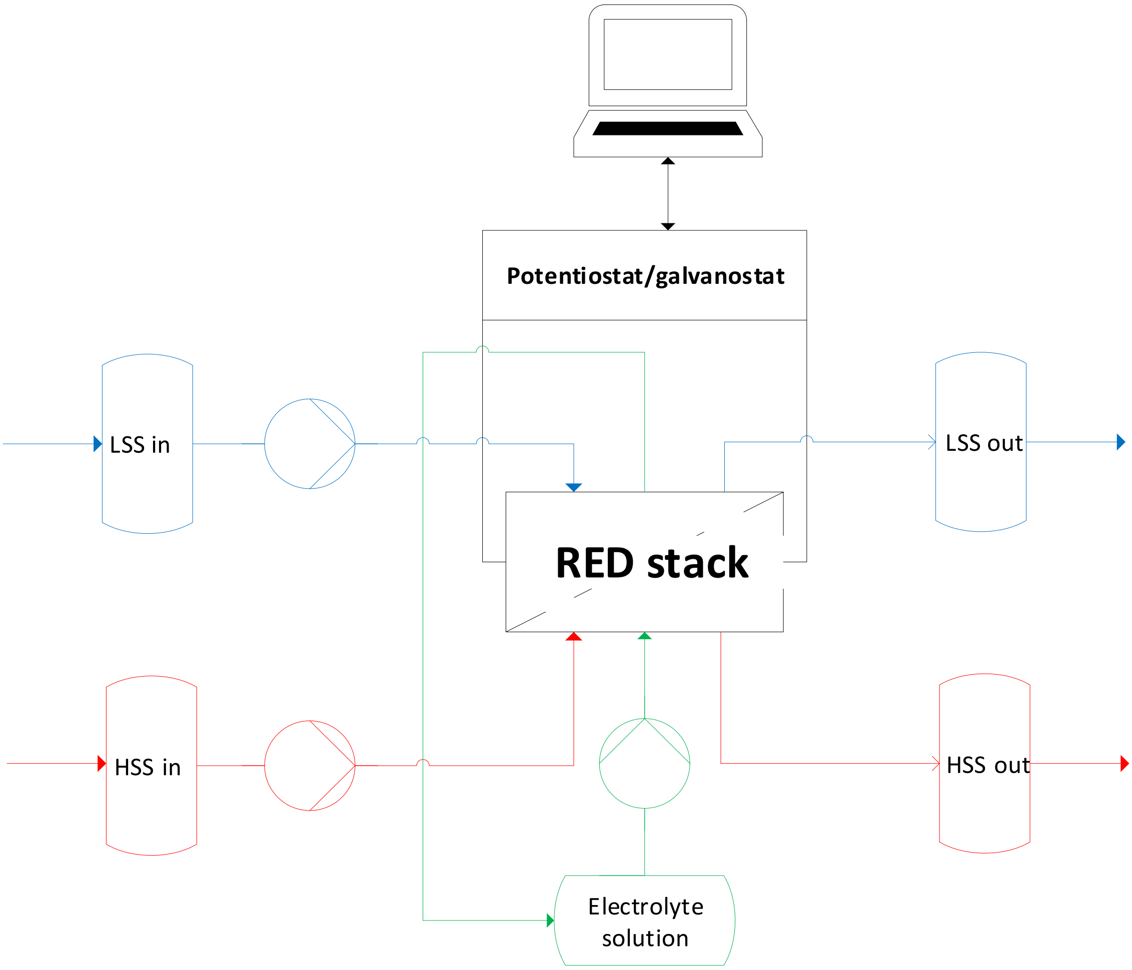


Figure 6 Scheme of the reverse electrodialysis unit

Four tanks (~20 L) are used for containing the LSS and HSS inlet and outlet solutions (Figure 6). A 2-channel peristaltic pump (Watson Marlow mod 232, 10-1400 mL/min) is used to recirculate each inlet solution inside the tank in order to guarantee a homogeneous concentration of the feed solution during lab scale investigation. The RED demonstrator is equipped with a mobile chemical hood (SYSTEM NFX, Human Arredi) for aspiration of any vapour or gas that could be released form the electrolyte solution [4][3]. Figure 7 show a picture of the lab-scale RED unit of the demonstrator.

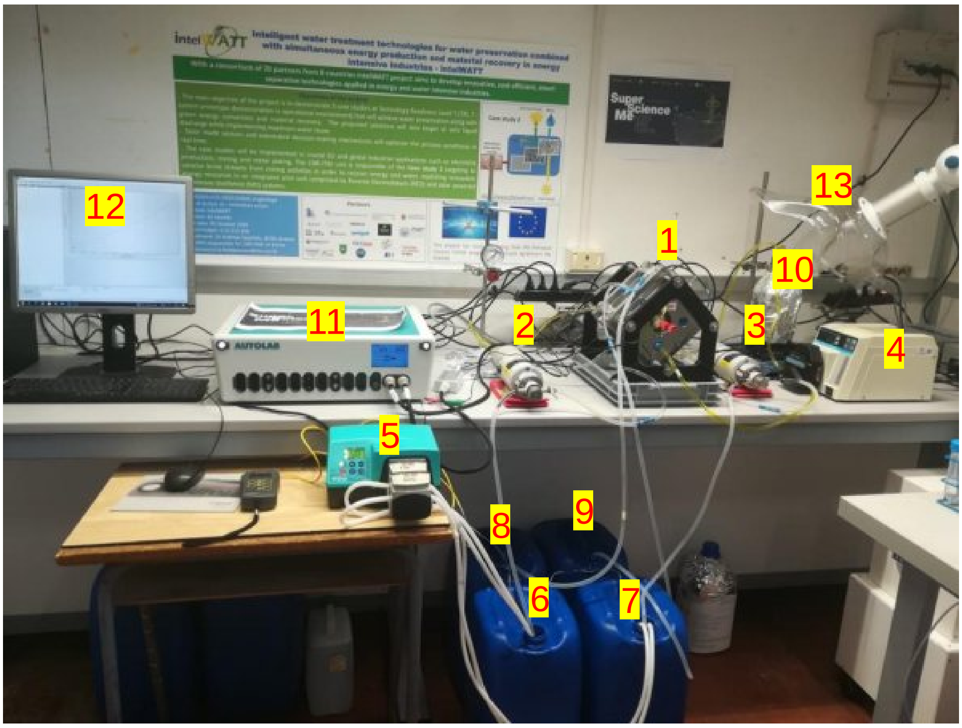


Figure 7 Picture of the RED lab unit of the demonstrator. Main components: 1. RED stack; 2. Gear pump (LSS); 3. Gear pump (HSS); 4. Peristaltic pump (electrolyte solution containing reversible redox couple); 5. 2-channel peristaltic pump (HSS and LSS recirculation in the corresponding tank); 6. LSS brine IN tank; 7. HSS brine IN tank; 8. LSS brine OUT tank; 9. HSS brine OUT tank; 10. Electrolyte rinse solution containing a reversible redox couple; 11. Potentiostat/galvanostat; 12. Computer for test management and data acquisition; 13. Chemical hood.

Performance indexes measured in the RED lab unit are the gross and neat power density (D2.5). The main parameters monitored are the following: voltage and current at the end electrodes; pressure drop over the HSS and LSS compartments (hydraulic losses), feed solutions flow rates of the streams. Moreover, pH, conductivity and temperature of the solutions, are also monitored.

Voltage and current at the end electrodes of the RED stack are monitored and registered by a potentiostat/galvanostat (Autolab/PGSTAT302N; maximum current ±2 A; potential range+/- 10 V; I and V accuracy +/- 0.2%). Two digital multimeters (Fluke-289 True-rms Industrial Logging Multimeter; current up to 10 A, accuracy: DC 0.05%, AC 0.6%; voltage up to 1000V, accuracy: DC 0.025%, AC 0.4%) with wireless data transfer connection (FLUKE-IR3000FC) is also available for voltage and current measuring. Pressure drops over inlet and outlet of the LSS and HSS streams are measured by two digital differential pressure meters (PCE-910). Conductivity and pH of the solutions are measured by a benchtop pH/conductimeter (Orion Star™ A215). The solutions temperature is measured by a portable thermometer with microprocessor for Pt100probes (Temp 7RDT, XS Instruments).

2.3 Membrane distillation (MD) unit

In a MD process a porous hydrophobic membrane is in contact with a heated aqueous solution on the feed side. The hydrophobic nature of the membrane prevents the mass transfer in liquid phase and creates a vapor-liquid interface at the entrance of each pore. Here, water evaporate, diffuse through the stagnant air in the membrane pores and it is condensed and/or removed on the permeate or distillate side. The specific method used to activate the vapour pressure gradient across the membrane (the driving force for this membrane operation) characterizes four main different MD configurations. In the most common arrangement, known as Direct Contact Membrane Distillation (DCMD), the permeate side of the membrane consists of a condensing fluid (generally water) that is directly in contact with the membrane. Alternatively, the vaporized solvent can be recovered on a condensing surface separated from the membrane by an air gap (AGMD), vacuum (VMD), or removed by a sweep gas (SGMD) [5]. The energy efficiency of MD is limited by the thermal separation of water and dissolved solutes. However, in recent years novel MD membrane designs and process configurations have reduced transmembrane heat loss and increased the heat recovered from the permeate stream, leading to reductions in energy consumption [6]. Moreover, MD process can operate with low-grade or renewable heat sources, including waste-heat from industrial sources and solar energy.

Currently the MD unit of the demonstrator operates in DCMD mode. The unit is designed to operate with both flat sheet and hollow fibres membrane modules and includes peristaltic pumps for fluids recirculation, heat exchangers/thermostatic baths for the generation of the driving force for the process (gradient of vapour pressure across the hydrophobic porous membrane), feed/distillate vessels with refill tank, sensors probe system connected to a programmable logic controller (PLC, DEltaE) to monitor process parameters and performances (transmembrane flux, fluids flow rates, pressure, inlet/outlet feed and inlet distillate temperature, conductivity) during operation (Figure 8 and Figure 9).

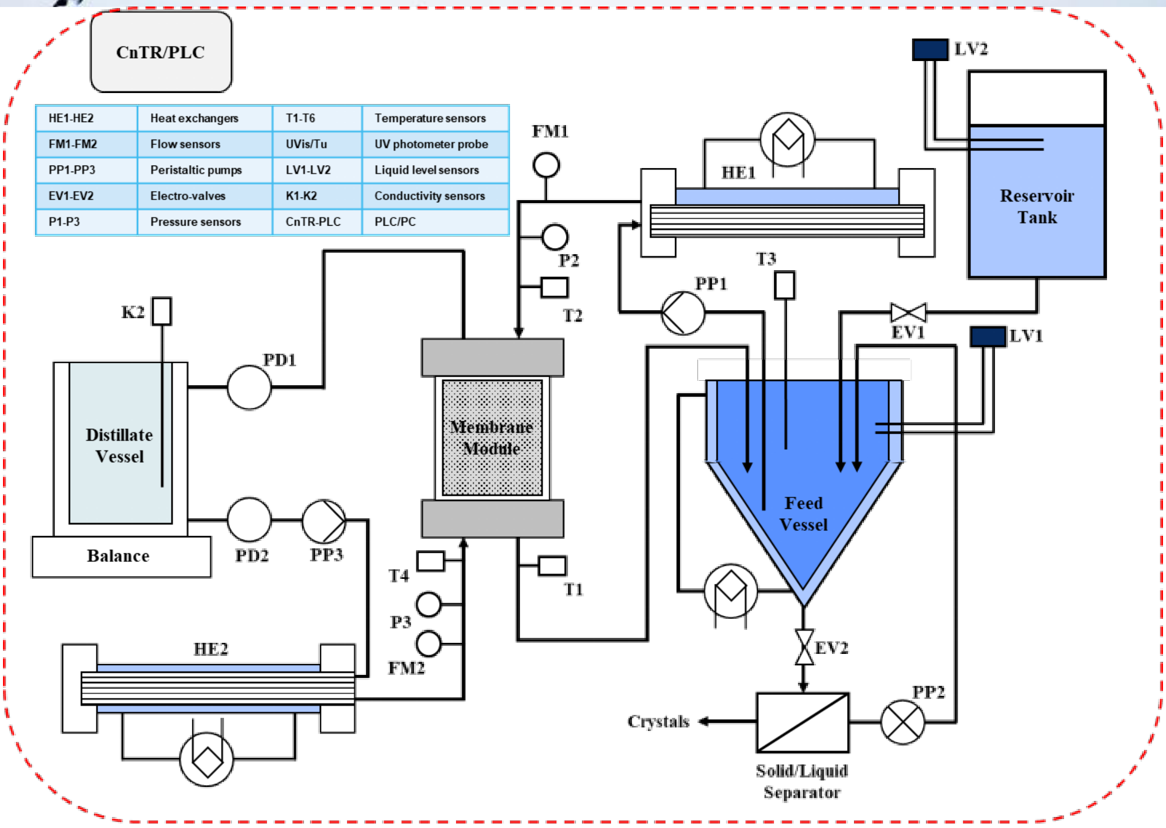


Figure 8 Scheme of the DCMD unit currently working in the demonstrator

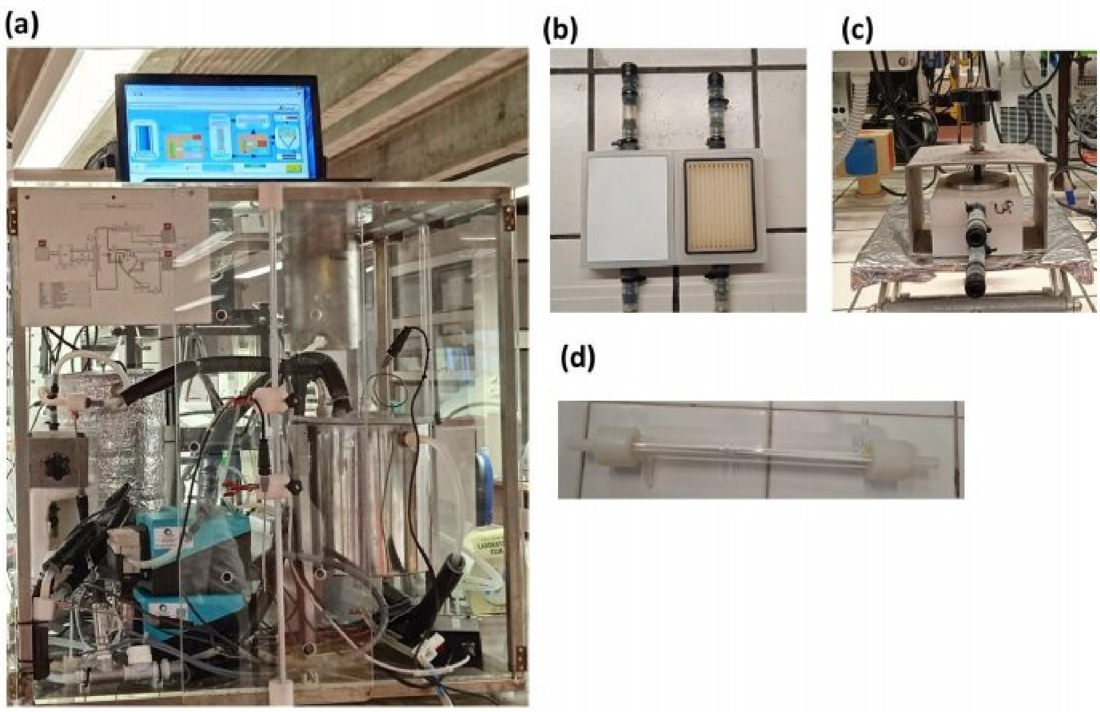


Figure 9 (a) Picture of the MD lab unit of the demonstrator. (b, c) flat sheet and (d) hollow fibers membrane modules.

This plant is currently used to perform DCMD tests using different commercial porous hydrophobic membranes using several feed solution compositions (e.g. pure HSS and LSS and mixed solutions outgoing from the RED unit) and operational conditions. Figure 10 provides a screenshot of the main process information delivered by the current monitoring system implemented with a PLC.

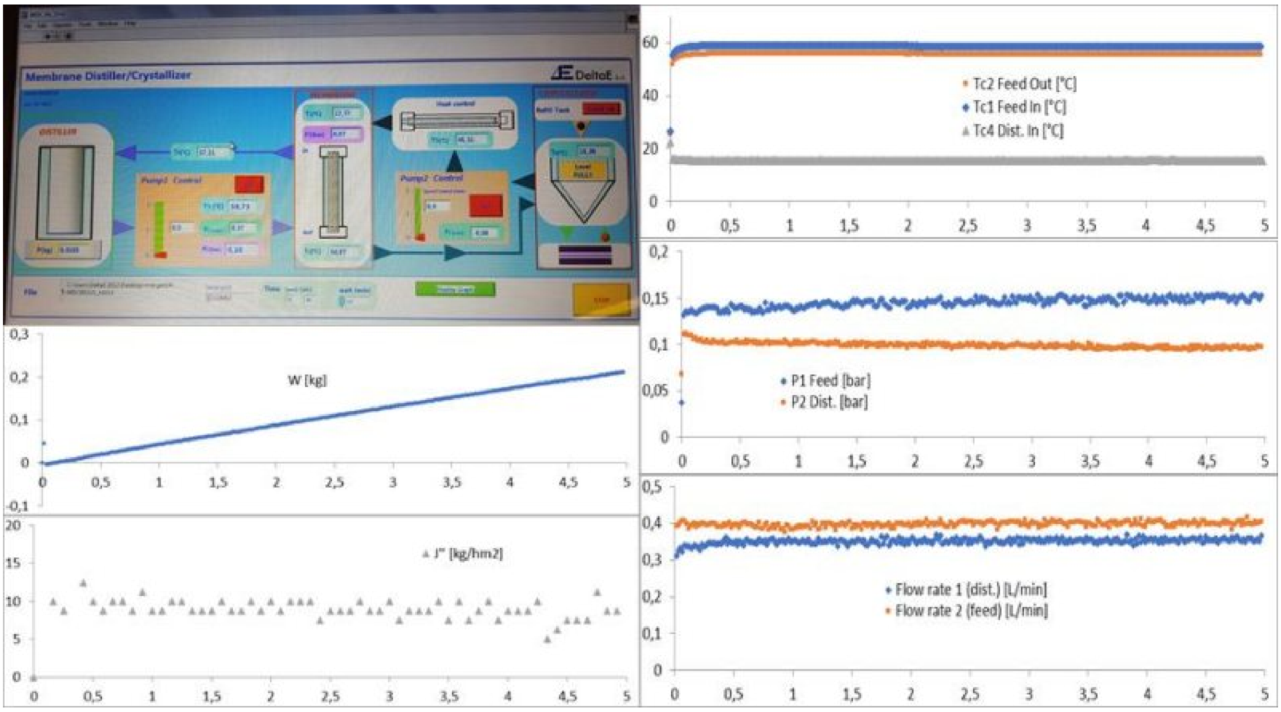


Figure 10 Example of output of the monitoring system for the currently used DCMD plant.

The performance of the membranes is estimated in term of flux, rejection, gain-output ratio (GOR) and the specific heat consumption (SHCv) as reported in D2.5.

Specifications of the main components of the MD lab unit operating in direct contact mode are reported in Table 1.

Table 1 Specifications of the main components of the MD unit operating in direct contact mode

Control system & membrane module		
CnTR/PLC	**Programmable logic controller** (PLC) (DeltaE)	 
Membrane module	**Membrane module** (DeltaE).	

Feed stream		
PP1	**Peristaltic pump** flow rate 15-2000 mL/min (drive 323DU/D with 313DW pump head 3 rollers, Watson Marlow)	
FM1	**Flow meter**, liquid acetal copolymer Sensor/Switch flow rate 0.25-6.5 L/m (257-149, RS)	
T1-T3	**Type K thermocouple probes** (RS)	
P2	**Pressure Sensor**, RS PRO 6 bar max pressure reading analogue (461-379, RS)	

Distillate stream		
PP3	**Peristaltic pump** flow rate 15-2000 mL/min (drive 323DU/D with 313DW pump head 3 rollers, Watson Marlow)	
FM2	**Flow meter**, Liquid Acetal Copolymer 4.5-16 V DC Sensor/Switch, flow rate 0.25-6.5 L/m (257-149, RS)	
T4	**Type K thermocouple probes** (RS)	
P3	**Pressure Sensor**, RS PRO 6 bar max pressure reading analogue (461-379, RS)	
K2	**Bench Combined Conductivity/pH Meter** (3540, Jenway)	
Balance	**Load cell technical balance**, capacity 15000±0.5 g (EU-C 15000PT USB, Gibertini)	

CNR-ITM will also receive from NCSR a vacuum multi effect membrane distillation (VMEMD) module (estimated delivery at M19). The MD unit of the demonstrator will be modified according to Figure 11 to operate with this type of module.

The analytical specifications of the main components of the MD unit that will operate with the VMEMD module are reported in Table2.

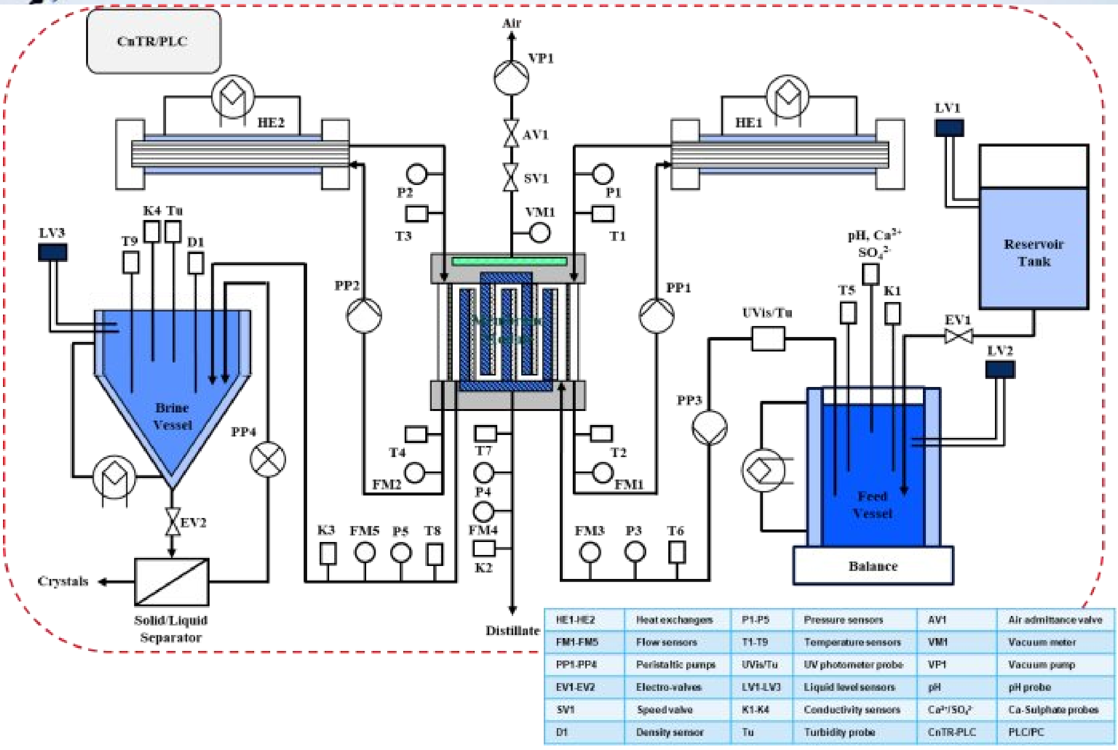
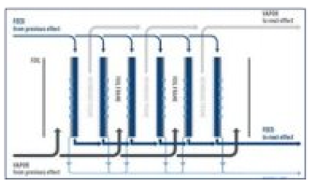


Figure 11 Design of the final MD unit of the demonstrator operating with a VMEMD module

Table 2 Specifications of the main components of the MD unit operating with a VMEMD module.

Control system & membrane module		
CnTR/PLC	**Programmable logic controller** (PLC) (DeltaE)	 
VMEMD module	**Vacuum multi effect membrane distillation module by NCSR**	

MD feed tank		
T5	**Type K thermocouple probes** (RS)	
K1, pH	**Bench Combined Conductivity/pH Meter** (3540, Jenway)	
Balance	**Load cell technical balance**, capacity 15000 g ± 0.5 g (EU-C 15000PT USB, Gibertini)	

Feed stream		
UVis-Tu	**UV Photometer w/280-880 nm light sources**, benchtop (SPEC-L-2-280-880 PHOTO, PendoTECH)	
	UV Flow Cell, 0.5 cm path length, polysulphone, ¼'' hose barb (SPECPS-N-025, PendoTECH)	
	Fiber Optics Cables (SPEC-OC-FIBER, PendoTECH)	
T6	**Type K thermocouple probes** (RS)	
P3	**Pressure Sensor**, RS PRO 6 bar max pressure reading analogue (461-379, RS)	
FM3	**Flow meter**, Liquid Acetal Copolymer 4.5-16 V DC Sensor/Switch, flow rate 0.25-6.5 L/m (257-149, RS)	
PP3	**Peristaltic pump** flow rate 15-2000 mL/min (drive 323DU/D with 313DW pump head 3 rollers, Watson Marlow)	

Hot stream steam riser		
P1	**PressureMAT PLUS monitor**, transmitter, alarm for 2 pressure sensors, 2 analogue inputs (PMAT2A, PendoTECH)	   
	Pressure Sensor, polysulfone, 0.25'' hose barb (PREPS-N-025, PendoTECH)	
T1-T2	**Monitor and transmitter for sensors** (4 analog outputs, 2 temp, 2 conduct) (CMONT, PendoTECH)	
	Temperature sensor, polysulfone & stainless steel, ¼'' hose barb (TEMPS-N-025, PendoTECH)	
PP1	**Peristaltic pump** flow rate 15-2000 mL/min (drive 323DU/D with 313DW pump head 3 rollers, Watson Marlow)	
FM1	**Flow meter**, Liquid Acetal Copolymer 4.5-16 V DC Sensor/Switch, flow rate 0.25-6.5 L/m (257-149, RS)	

Distillate stream		
T7, K2	**Monitor and transmitter for 2 conductivity sensors** (4 analog outputs, 2 temp, 2 conduct) (CMONT, PendoTECH)	  
	Conductivity/temperature Sensor, polysulphone ¼'' hose barb (CONDS-N-025, PendoTECH)	
FM4	**Flow meter**, liquid acetal copolymer 4.5-16 V DC Sensor/Switch flow rate 0.25-6.5 L/m (257-149, RS)	
P4	**Pressure Sensor**, RS PRO 6 bar max pressure reading analogue (461-379, RS)	

Vacuum line		
VP1	**Scroll vacuum pump** 43 m^3/h, ultimate vacuum 0.01 mbar (XDS35i, Edwards)	
AV1	**Manual vent/air-admit valve** leak rate <1x10^{-6} mbar L/s (AV10K, Edwards)	
SV1	**Speedivalves with Viton diaphragm** (SP40K, Edwards)	
VM1	**TIC instrument controller** 3 head RS232/RS485 (Edwards)	
	Active Strain Gauge, model ASG2 NW16 1000 mbar (Edwards)	
Cold stream condenser		
P2	**PressureMAT PLUS monitor**, transmitter, alarm for 2 pressure sensors, 2 analogue inputs (PMAT2A, PendoTECH)	
	Pressure Sensor, polysulfone, 0.25'' hose barb (PREPS-N-025, PendoTECH)	
T3-T4	**Type K thermocouple probes** (RS)	
PP2	**Peristaltic pump** flow rate 15-2000 mL/min (drive 323DU/D with 313DW pump head 3 rollers, Watson Marlow)	
FM2	**Flow meter**, Liquid Acetal Copolymer 4.5-16 V DC Sensor/Switch, flow rate 0.25-6.5 L/m (257-149, RS)	

Concentrate stream		
T8, K3	**Monitor and transmitter for 2 conductivity sensors** (4 analog outputs, 2 temp, 2 conduct) (CMONT, PendoTECH)	
	Conductivity/temperature Sensor, polysulphone ¼'' hose barb (CONDS-N-025, PendoTECH)	
FM5	**Flow meter**, liquid acetal copolymer 4.5-16 V DC Sensor/Switch flow rate 0.25-6.5 L/m (257-149, RS)	
P5	**Pressure Sensor**, RS PRO 6 bar max pressure reading analogue (461-379, RS)	

3 References

[1] R. A. Tufa, S. Pawlowski, J. Veerman, K. Bouzek, E. Fontananova, G. Di Profio, S. Velizarov, J. G. Crespo, K. Nijmeijer, E. Curcio, Progress and prospects in reverse electrodialysis for salinity gradient energy conversion and storage, Applied Energy 225 (2018) 290–331. DOI: 10.1016/j.apenergy.2018.04.111

[2] J.A. Andrés-Mañas, A. Ruiz-Aguirre, F.G. Acién, G. Zaragoza, Assessment of a pilot system for seawater desalination based on vacuum multi-effect membrane distillation with enhanced heat recovery, Desalination 443 (2018) 110–121. DOI: 10.1016/j.desal.2018.05.025

[3] E. Fontananova, D. Messana, R. A. Tufa, I. Nicotera, V. Kosma, E. Curcio, W. van Baak, E. Drioli, G. Di Profio, Effect of solution concentration and composition on the electrochemical properties of ion exchange membranes for energy conversion, Journal of Power Sources 340 (2017) 282-293. DOI: 10.1016/j.jpowsour.2016.11.075

[4] O. Scialdone, A. Albanese, A. D'Angelo, A. Galia, C. Guarisco, Investigation of electrode material – Redox couple systems for reverse electrodialysis processes. Part II: experiments in a stack with 10-50 cell pairs, Journal of Electroanalytical Chemistry 704 (2013) 1-9. DOI: 10.1016/j.jelechem.2013.06.001.

[5] E. Curcio, G. Di Profio, E. Drioli, Membrane distillation and osmotic distillation, In: Comprehensive Membrane Science and Engineering, Volume 4, Chapter 4.01, E. Drioli & L. Giorno Eds., Elsevier, Amsterdam, The Netherlands, 2010, p. 1-20. ISBN: 978-008093250-7; 978-044453204-6. DOI: 10.1016/B978-0-08-093250-7.00015-3

[6] A. Deshmukh, C. Boo, V. Karanikola, S. Lin, A. P. Straub, T. Tong, D. M. Warsingerab, M. Elimelech Membrane distillation at the water-energy nexus: limits, opportunities, and challenges, Energy Environ. Sci., 11 (2018) 1177-1196. DOI: 10.1039/c8ee00291f