

## Appendices

### Annex 1 List of Abbreviations and Symbols

<b>Polymers</b>		<b>Solvent and other additives</b>	
PVA	Polyvinyl alcohol	NMP	1-Methyl-2-pyrrolidone
PAA	Poly(acrylic acid)	DMF	N,N-dimethylformamide
P(VDF-TrFE)	Poly(vinylidene fluoride-co-trifluoroethylene)	mpg-C <sub>3</sub> N <sub>4</sub>	Mesoporous graphitic carbon nitride
PEG	Poly (ethylene glycol)	DMAc	N,N- dimethyl acetamide
PVDF	Poly(vinylidene fluoride)	TTIP	Titanium tetraisopropoxide
PMMA	Poly (methylmethacrylate)	EtOH	Ethanol
PDA	Polydopamine	APTES	Aminopropyltriethoxysilane
PES	Polyethersulfone	TEP	triethyl phosphate
PSf	Polysulfone	RGO	Reduced Graphene Oxide
DA	Dopamine	GLUT	Glutaraldehyde
PEGMA	Poly(ethylene glycol) methyl ether methacrylate		
CA	Cellulose Acetate		
<b>Pollutant</b>		<b>Others</b>	
MB	Methylene Blue	HF	Hollow Fiber
IBPR	Ibuprofen	DHLF	Dual Layer Hollow Fiber Membrane
DS	Dichlofenac Sodium	N.D.	Not Detectable
MO	Methyl Orange	ALD	Atomic Layer Deposition
RB	Reactive Blue		
RdB	Rodamine B		
RB5	Reactive black 5		
BSA	Bovine Serum Albumin		
BG	Brilliant green		
BPA	Bisphenol A		
EE2	17 $\alpha$ -ethynylestradiol		
HA	Humic Acid		
POME	Palm oil mill effluent		
NP	Nonylphenol		
Tartz	Tartarine		
SULF	Sulfadiazine		
Ind. Surf	Industrial Surfactant		
EY	Eosin Yellow		
SFMO	Sulfamethoxazole		
Bentz	Bentazon		
		<b>List of Symbols</b>	
		$\theta$	Contact Angle
		$\varepsilon$	Porosity
		Model	Model pollutant
		V <sub>0</sub>	Volume of contaminated solution
		C <sub>0</sub>	Initial concentration
		D	Degradation
		R	Rejection
		k <sub>app</sub>	Apparent reaction kinetic constant
		A	Area of the membrane
		I	Irradiation source and power
		t	Total irradiation time
		P	Performance in terms of $\mu\text{g}$ of pollutant removed per hour per area of PMs
		v	Recirculation velocity

## Annex 2 UV-responsive PMs used in different types of PMRs, their Photocatalytic Activity and other membrane performances with operating conditions

### Annex 3 Batch PMRs Using Flat Sheet PMs

PM type	wt% TiO <sub>2</sub> added	θ (°)		ε (%)		M	C <sub>0</sub> , V <sub>0</sub> , pH,	I, t, A	D (%)	k <sub>app</sub> (min <sup>-1</sup> )	P (μg/(h×cm <sup>2</sup> ))	Observation and other performances	Ref.
		PV DF	PM	PV DF	PM								
TiO <sub>2</sub> /PVDF(2 mM TTIP/ EtOH sol-gel)	0.092 wt% of dry PMs	73	76	7% of porosity reduced		MB	3.2 mg/L, 4 mL	76 W/m <sup>2</sup> UV-A, 2.5 h, 4.12 cm <sup>2</sup> .	100% (2 h)		1.55	Decrease hydrophlicity and porosityof PMs and hence water flux decreased but BSA filtration performance improved	[16]
						IBRF	100 mg/L, 4 mL		45 %		48.5		
						DS	25 mg/L, 4 mL		70%		12.13		
PVA/TiO <sub>2</sub> /P VDF	1%	87	59	44	38	RB	50 mg/L, 150 ml	15W UV-C lamp, 2.5 h, 25 cm <sup>2</sup>	44%	-	26.4	At optimum PVA (3 wt%) and 1 wt% TiO <sub>2</sub> improved mechanical property and hydrophilicty but above this value TiO <sub>2</sub> encapsulate.	[72]
						RdB			45%	-	27		
						MO			48%	-	28.8		
TiO <sub>2</sub> /PAA/P VDF	0.5 (w/v)	116	28			RB5	40 mg/L, 25mL	15W UV lamp, 2 h, 25 cm <sup>2</sup>	30%	0.031	6	Higher flux obtained under UV due to high antifouling property of the membrane. 3 wt% loading of TiO <sub>2</sub> shows best photocatalytic activity.	[28]
	1. 5 (w/v)		23						30%	0.033	6		
	3 (w/v)		N.D						42%	0.042	8		
TiO <sub>2</sub> /PVP/PV DF/DMAc	1%	79	65	70	75	BSA	1000 mg/L, 50 mL.	11W UV, 2 h, 19.3 cm <sup>2</sup>	53%	0.0068	686.5	Addition of TiO <sub>2</sub> decrease surface roughness, and incorporation GO faster the photodegradation of BSA and improve the BSA rejection and water flux.	[73]
GO/TiO <sub>2</sub> /PV P/PVDF/DM Ac	1%		61	70	83				80%	0.0142	1036.3		
TiO <sub>2</sub> nanotube/ PVP/PVDF/ DMAc	0.1%	92	82	28	43	BG	150 mL. pH 7.5,	15W Hg-lamp, 1.5 h, 50 cm <sup>2</sup> .	42% for 1.5 wt%			Best antifouling property obtained for 1.5 wt% TiO <sub>2</sub> content, more than this porosity, mechanical stability and water flux decreased.	[74]
	0.5%		-		47								
	1.0%		73		50								
	1.5%		70		56								
	2%		-		36								
TiO <sub>2</sub> /P(VDF–TrFE)/ DMF	3%	76	88	80	78	MB	3.2 mg/L, 13 mL, pH 6.8	40 W/m <sup>2</sup> LED UVA, 1.5 h, 12 cm <sup>2</sup>	77%	0.018	1.78	8 wt% TiO <sub>2</sub> shows better photodegradation but porosity and hydrophilicity decreased. Inclusion of a zeolite (NaY) can increase hydrophilicity and porosity by aiding microporosity and capillary effect of zeolite.	[21]
	5%		97		77				93%	0.026	2.15		
	8%		97		74				99%	0.037	2.29		
NaY/TiO <sub>2</sub> /P( VDF-TrFE) /DMF	3% NaY		N.D		90				91%	0.023	2.1		
	5% NaY				95				99%	0.04	2.29		
	8% NaY				97				96%	0.033	2.22		
TiO <sub>2</sub> /PVDF/P MMA/PEG/P VP/TEP	0.12 %	110	102	82	80	MB	3.2 mg/L, 500 mL	500W UV lamp, 6.5 h, 16 cm <sup>2</sup>	86%	0.0055	10.58	TiO <sub>2</sub> loading increase the porosity, hydrophilicity, and mechanical properties of PMs.	[75]
	0.25%		98		81				95%	0.0084	11.69		
	0.50%		93		83				99%	0.0117	12.18		
TiO <sub>2</sub> /PVDF/P VP/DMAc	4%	108	61			MB	6.4 mg/L, 50 mL	UVA lamp 4W, 1.5 h, 20 cm <sup>2</sup>	100 %	0.044	10.66	Fabrication of highly porous and photoactive PMs using a bipolymer system through electrospinning process.	[36]
						BPA	5.0 mg/L, 50 mL		96% (4 h)	0.030	3		
						EE2	5.0 mg/L, 50 mL		96%	0.033	8		

## Flat sheet (Dead End)

PM type	wt% TiO <sub>2</sub> added	θ (°)		ε (%)		M	C <sub>0</sub> , V <sub>0</sub> , pH, υ	I, t, A	D (%) or R (%)	k <sub>app</sub> (min <sup>-1</sup> )	Observation and other performances	Ref.
		PV DF	PM	PV DF	PM							
TiO <sub>2</sub> /PVDF/H <sub>2</sub> O	0.1 g/L**	128	127	75	34	RB 19	25 mg/L, pH 3, 0.877 L/min	9W UV-C, 1 h, 59.5 cm <sup>2</sup> .	19%		Negatively charged RB5 favors acidic pH, alkaline condition decreases photoactivity.	[51]
	0.5 g/L		121		39				26%			
	1 g/L		114		40				35%			
TiO <sub>2</sub> /PDA/PVDF	1.5 %	85	65	14	8	ARB	10 mL (cell )	0.12 W/m <sup>2</sup> UV lamp, 3 h, 4.1cm <sup>2</sup>	98%		Increased hydrophilicity, and high antifouling properties	[20]
						ARGs			98%			
Flat sheet (Cross flow)												
TiO <sub>2</sub> /PVDF (CVD)	-	61 90	26	70	-	MB	4.15 mg/L	6 W/m <sup>2</sup> Hg-lamp, 6 h, 14.5 cm <sup>2</sup>	92% (4 h)	0.028	Needed to optimize sputtering conditions. Increased in hydrophilicity improved transmembrane fluxes.	[18]
						DS	29.6 mg/L, 250 mL/min		100%	0.0083		
TiO <sub>2</sub> /PVP/PVDF/DA/DMAc	0.8%	80	30			SULF	100 μg/L, 100 mL. pH 7.5, 30 L/h	1.25 W/m <sup>2</sup> UV, 2 h, 26.12 cm <sup>2</sup> .	91.4%	0.0216	Rejection increased under UV irradiation because of synergistic effect of photodegradation and filtration.	[44]
TiO <sub>2</sub> /LiCl/PVDF/DMAc	0.5%		75			HA	2 mg/L, pH 7.5, 500 mL/min	100 W Hg-lamp, 8 h, 48 cm <sup>2</sup>	82% of R		Better NOM rejection and reduction of membrane fouling with good self-cleaning ability.	[43]
Fe <sub>3</sub> O <sub>4</sub> @TiO <sub>2</sub> /PVDF/NMP	0.5%	A decrease of 10° for PMs		1.5	1.6	HA	20 mg/L, 100 mL. pH 7.5, 100 L/h	19W Hg-lamp, 2 h, 50 cm <sup>2</sup> .	67% of R		Most of the nanoparticles flocculate on the surface of PMs because of magnetic coagulation bath. Under UV, PMs had better antifouling performance against humic acid solution.	[76]
	1%				1.7				69% of R			
HF (Cross flow)												
TiO <sub>2</sub> /PVP/PVDF/NMP	0.5%	88	78	36	40	Ind. Surf.	COD ~1420 mg/L, pH 6.8, 410 mL/min	8 W UV-A, 36 cm <sup>2</sup>	66.7% of R		Under UV-light optimum content of TiO <sub>2</sub> (2 wt%) exhibited desired porosity, hydrophilicity, tensile strength, rejection and long-term stability in salty environment under UV.	[32]
	1%		71		60							
	2%		61		82							
	3%		64		81							
TiO <sub>2</sub> /PVP/PVDF/NMP	0.5%			85	85		6.4 mg/L, 250 mL, rate 0.06 m/s (25 g/L NaCl)	UV-A 18 W, 6 h, 36 cm <sup>2</sup>			Addition of pore-former PEG, increase the hydrophilicity and and hence improved water permeability and stability of PMs under salty water.	[23]
TiO <sub>2</sub> /PVP/PEG/PVDF /NMP	0.5%	86	78	82	83	MB-water MB-NaCl		97%	0.012			
HF (submersed)												
TiO <sub>2</sub> /PVDF/DM Ac	3%	91	79			NP	100 mg/L, 7 L pH 6.8	UV-A 36 W, 4 h, 248 cm <sup>2</sup> (20 HF)	98% of R		7.5 wt% is the optimum TiO <sub>2</sub> content, and above this value decrease antifouling property, porosity and hence flux performance.	[26]
	7.5%		78						96% of R			
	10.5%		75						91% of R			
	15%		67						96% of R			

TiO <sub>2</sub> nanotube/ PVP/PVDF/NMP	0.1%	76	70		(POME)	TC = 742 mg/L, 15 L. pH 3	UV-A 8W, 4 h, 2.89 m <sup>2</sup>	42.3%		The best flux, rejection and anti-fouling property observed for 0.5wt% TiO <sub>2</sub> . High initial concentration decreases photoactivity of PMs, and alkaline condition improve flux.	[53]
	0.3%		65					57.1%			
	0.5%		64					67.3%			
	1%		61					50%			

## Annex 5 Representative of Some Visible-light sensitive PMs used in different types of PMRs, their Photocatalytic Activity, and Other Performances of Membranes including Operating Conditions

### Flat sheet or Hollow fiber membranes in Batch Process

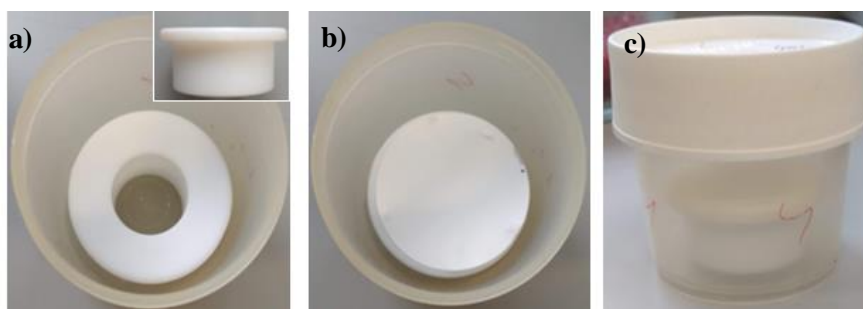
PM type	wt% TiO <sub>2</sub> added	θ (°)		ε (%)		M	C <sub>0</sub> , V <sub>0</sub> , pH, v	I, t, A	D (%) or R (%)	k <sub>app</sub> (min <sup>-1</sup> )	P (μg/(h ×cm <sup>2</sup> ))	Observation and other performances	Ref.
		Sup port	PM	Sup port	PM								
N-TiO <sub>2</sub> / PEG/PVDF/ DMAc (DLHF)	7.5%		70		35.1	BPA	5 mg/L	30W LED	81.6%			N-doped TiO <sub>2</sub> DLHF shows the same photoactivity under UV and solar irradiation. 7.5 wt% is the optimum catalyst dose, above this content, properties improve but catalyst agglomerates due to higher surface tension between the solvent of dope solution.	[77]
	3%		82		9.6	RB5	5 mg/L	Visible light 6h	75% (for 7.5 wt% TiO <sub>2</sub> )				[55]
	7.5%		70		35.1								
	10.5%		69		55.1								
Fe-TiO <sub>2</sub> / PEG/PSF/ DMAc & NMP (4:1)	0.05%					BPA	10 mg/L, 100 mL	500 W Xenon lamp, 3 h, 47.78 cm <sup>2</sup>	90.8% (for 0.2 wt%)		6.33	Enhanced mechanical property and shows self- cleaning ability under visible-light. Optimum catalyst dose is 0.2 wt%. Higher inclusion of TiO <sub>2</sub> (0.25 wt%) decreases membrane mechanical stability.	[78]
	0.1%												
	0.15%												
	0.2%												
	0.25%												
N-TiO <sub>2</sub> / PMAA-g- PVDF/PAN/ DMAc	1%					Bentaz	10 mg/L, 100 mL. pH 7	UV 5063 lux, 3 h, 36 cm <sup>2</sup>	42.9% for 3 wt% TiO <sub>2</sub>		3.97	3 wt% catalyst dose optimum amount, above this value, roughness of the membrane surface increases because nanoparticles began to form lumps. Under basic conditions, positive charged bentazon adsorb more on PMs surface and enhance photodegradation.	[62]
	3%							Sunlight 110000 lux, 3 h, 36 cm <sup>2</sup>	99.8% for 3 wt% TiO <sub>2</sub>		9.24		
	5%												
Pd/N-TiO <sub>2</sub> / PSf/NMP	0.5%	79	66			EY	100 mg/L, 100 mL	450W Xenon lamp, 3 h, 9 cm <sup>2</sup>	92.7%	0.0098	257.5	Improved hydrophilicity, porosity, visible light absorption and photoactivity but higher TiO <sub>2</sub> content increased membrane roughness due to embedded TiO <sub>2</sub> particle aggregation.	[61]
	1%		72					86.7%	0.0084	240.9			
	2%		73					97%	0.0149	269.4			
	4%		76					96.3%	0.0142	267.5			
	7%		77					97.3%	0.0169	270.3			
	Flat sheet (Dead-end)												
Ag-TiO <sub>2</sub> / APTES/PVP /PVDF/DM Ac	0.1%	82	73			MB	3 mg/L, 30 mL. pH 7	Visible light, 1 h	80.3%			Smaller TiO <sub>2</sub> particles caused intracellular damage, and unique antibacterial performance. Meanwhile, Ag has inhibitory effect on the growth of bacteria; Thus these PMs shows excellent antibacterial activity towards E coli. Under visible light, provides a good self-cleaning ability and improved BSA rejection (from 63.43 to 89.80%)	[52]
	0.2%		68					86.7%					
	0.5%		61					90.1%					
Ag-TiO <sub>2</sub> / PVP/PVDF/ DMAc	0.01%	83	78			BSA		450 W Xenon lamp, 9 cm <sup>2</sup>	97.21% of rejection, 8h				[59]
	0.03%		74										
	0.06%		57										

RGO-Ag-TiO <sub>2</sub> /PEG/G LUT/CA	2.5 mg <sup>†</sup>		66		MB	20 mg/L	500 W Xenon lamp,	99% of rejection for all pollutant conditions			Inclusion of 10 mg of catalyst shows good and stable hydrophilicity and water permeability. Simultaneously degrade dye and separate oil–water emulsions under visible-light irradiation in a short time.	[58]
	5 mg		59		MB-oil							
	10 mg		43		RhB	30 mg/L						
	20 mg		37		RhB-oil							
mpg-C <sub>3</sub> N <sub>4</sub> /TiO <sub>2</sub> /PVP/P Sf/NMP	0.2%	71	66		SFMO	10 mg/L, 50ml	300 W Xenon lamp, 30 h, 8.5 cm <sup>2</sup>	49%		0.96	Mechanically stable PMs prepared by adding nanoparticles. Increase hydrophilicity of the PMs, enhance water permeability albeit pore size decreased.	[60]
	1%		58					69%		1.35		
Flat sheet (Cross-flow)												
TiO <sub>2</sub> /PVDF-TrFE/DMF	8%		40		Tartz	10 mg/L	Sunlight, 5 h, (38×12) cm <sup>2</sup> v = 28 mL/s (for all C <sub>0</sub> )	77.77	0.30		Increase in initial feed concentration (10-30 mg/L) reduced the photodegradation of tartzarine (78 to 47%). Meanwhile, increasing the feed flow rate (9.78-28 mL/s) enhance photodegradation efficiency (37-77%) due to larger turbulence from the higher flow rate, which promotes external mass transfer.	[33]
						20 mg/L		57.72	0.18			
						30 mg/L,		46.57	0.12			
3D-TiO <sub>2</sub> /ZnO/PVDF (ALD)		95	40		MB	3.2 mg/L,	200 W Xenon lamp, 19.6 cm <sup>2</sup>	95% (30 min)	0.11	31.02	Super-hydrophilicity of this visible-light active membrane shows enhanced anti-fouling performance.	[57]
					HA	100 mL 300 mL		73% (1 h)		35.75		
L–Histidine/TiO <sub>2</sub> /CdS/P VP/PES/DM Ac	0.1%	63	51		POME	1 g/L, pH 5.5, 150 L/h	500 W Halogen lamp,	100% (30 min)			Better performance observed after incorporation of 0.5 wt% catalyst, because above this load surface becomes rougher and nanoparticle agglomerates. Increase in initial feed concentration (1-5 g/L) reduced the permeation flux (31.4 to 11 kg/m <sup>2</sup> .h). Meanwhile, increasing the feed flow rate (50, 150 L/h) also improved rejection efficiency and permeation flux. Due to turbulence as well as bigger Reynolds number from higher flow rate reduce the concentration polarization and membrane fouling.	[79]
	0.5%		47									
	1%		45									

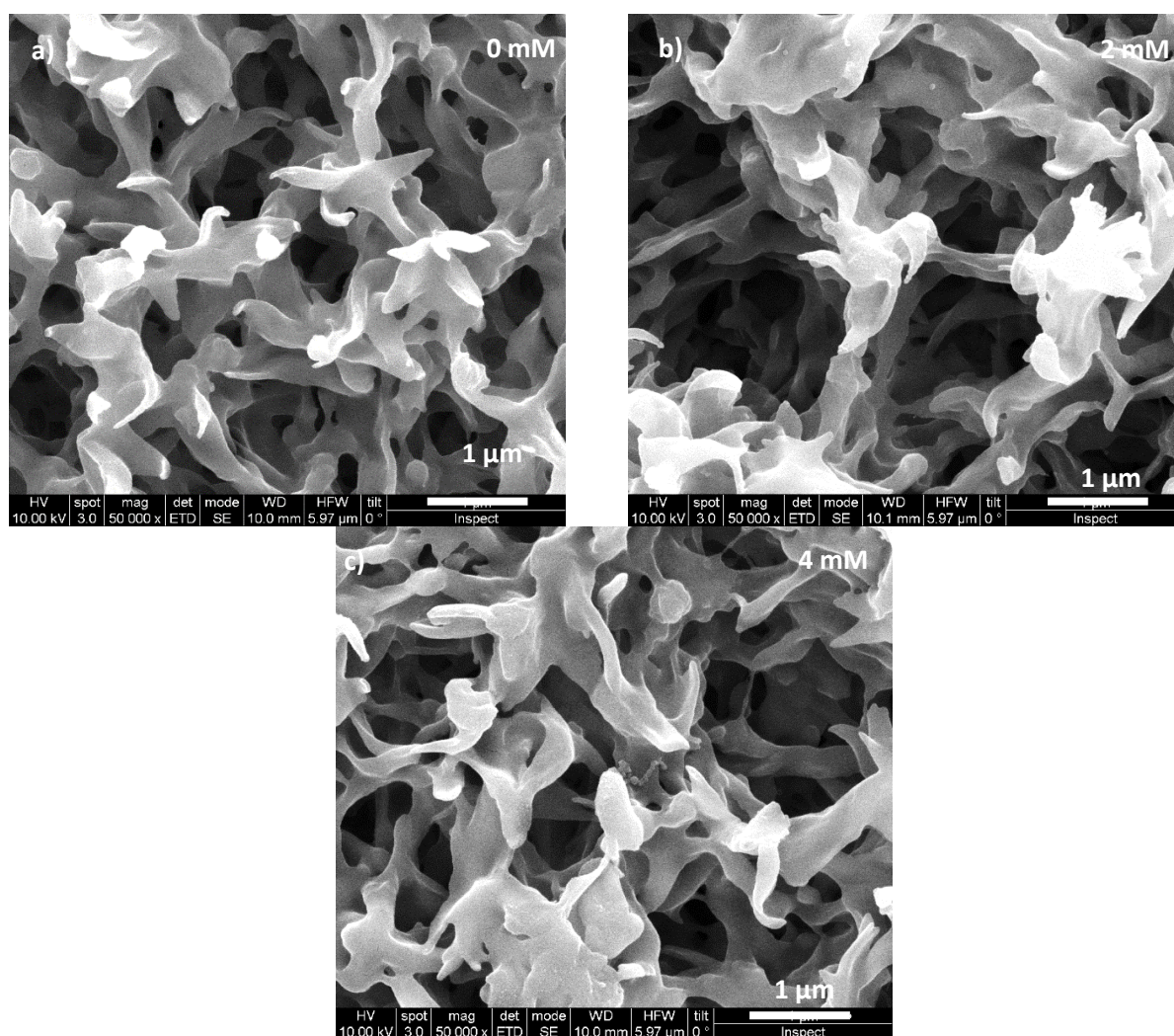
<sup>†</sup> These amounts are deposited onto membrane grafting by PEG and GLUT through vacuum filtration method

<sup>\*\*</sup> These are based on suspensions of TiO<sub>2</sub> on water and then membrane was immersed in this suspension to deposit TiO<sub>2</sub>

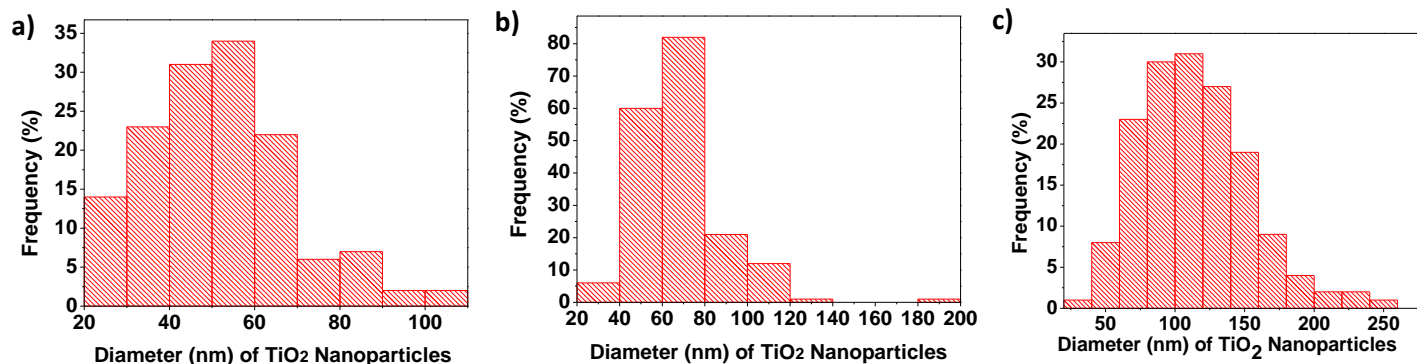
Annex 6 Recipient to crystallize on-site synthesized  $\text{TiO}_2$  nanoparticles on membrane



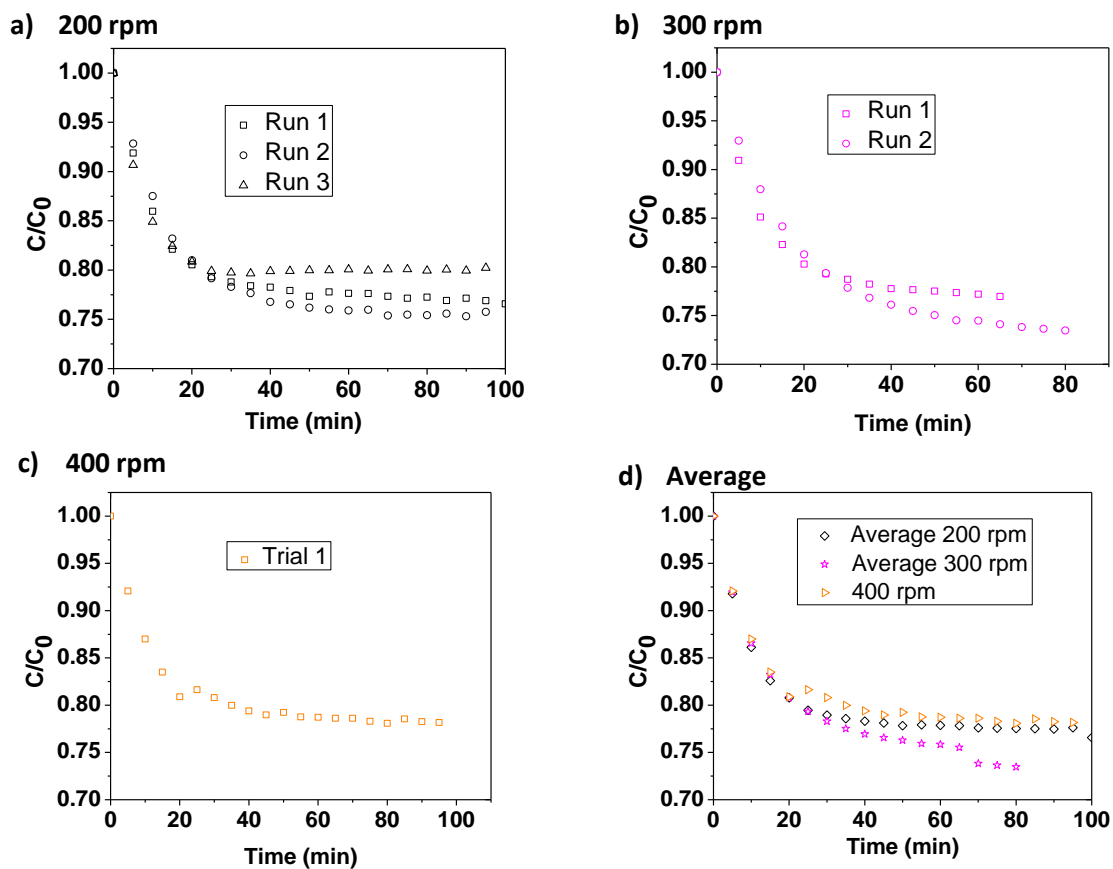
Annex 7 SEM images of the Membranes prepared from lower concentration (2 mM and 4 mM) sol-gel solution; a) Pure PVDF (0mM), b) 2mM, c) 4mM



Annex 8 Histogram of the distribution of diameter of nanoparticles deposited on a) 8mM, b) 16mM, and c) 32mM



Annex 9 Effect of Mechanical Stirring on MB adsorption onto non-coated pure PVDF membrane a) adsorption at 200 rpm for 3 runs, b) adsorption at 300 rpm for 2 runs, c) adsorption at 400 rpm, and d) comparison of average adsorption rates for all runs.



Annex 10 Histogram of the distribution of diameter of nanofibers of a) Electrospun pure PVDF, b) PPTM, and c) PTM

