

## 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(vinylidenefluoride-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	Aminopropyltriethoxsilane
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
DS	Dichlofenac Sodium	N.D.	Membrane
MO	Methyl Orange	ALD	Not Detectable
RB	Reactive Blue		Atomic Layer Deposition
RdB	Rodamine B	θ	
RB5	Reactive black 5	ε	
BSA	Bovine Serum Albumin	Model	<b>List of Symbols</b>
BG	Brilliant green	V <sub>0</sub>	Contact Angle
BPA	Bisphenol A		Porosity
EE2	17 $\alpha$ -ethynylestradiol	C <sub>0</sub>	Model pollutant
HA	Humic Acid	D	Volume of contaminated
POME	Palm oil mill effluent	R	solution
NP	Nonylphenol	k <sub>app</sub>	
Tartz	Tartzarine	A	Initial concentration
SULF	Sulfadiazine	I	Degradation
Ind. Surf	Industrial Surfactant	t	Rejection
EY	Eosin Yellow	P	Apparent reaction kinetic
SFMO	Sulfamethoxazole		constant
Bentz	Bentazon	v	Area of the membrane
			Irradiation source and power
			Total irradiation time
			Performance in terms of $\mu$ g
			of pollutant removed per
			hour per area of PMs
			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 48.5 12.13	Decrease hydrophobicity and porosity of PMs and hence water flux decreased but BSA filtration performance improved	[16]				
PVA/TiO <sub>2</sub> /PVDF	1%					IBRF	100 mg/L, 4 mL		45 %							
						DS	25 mg/L, 4 mL		70%							
						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 hydrophilicity but above this value TiO <sub>2</sub> encapsulate.	[72]			
TiO <sub>2</sub> /PAA/PVDF	0.5 (w/v)	116	28			RdB			45%	-	27					
	1. 5 (w/v)					MO			48%	-	28.8					
	3 (w/v)					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]			
TiO <sub>2</sub> /PVP/PVDF/DMAc	1%	79	65	70	75	BSA	1000 mg/L, 50 mL.		30%	0.033	6					
GO/TiO <sub>2</sub> /PVDF/DMAc	1%			61	70				42%	0.042	8					
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> .	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]			
	0.5%			-	47				80%	0.0142	1036.3					
	1.0%			73	50				42% for 1.5 wt%							
	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> /PVDF-TrFE/DMF	3% NaY	N.D		90		MB	3.2 mg/L, 50 mL	500W UV lamp, 6.5 h, 16 cm <sup>2</sup>	91%	0.023	2.1	TiO <sub>2</sub> loading increase the porosity, hydrophilicity, and mechanical properties of PMs.	[75]			
	5% NaY			95					99%	0.04	2.29					
	8% NaY			97					96%	0.033	2.22					
TiO <sub>2</sub> /PVDF/PMMAP/PEG/PVP/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	Fabrication of highly porous and photoactive PMs using a bipolymer system through electrospinning process.	[36]			
	0.25%			98	81				95%	0.0084	11.69					
	0.50%			93	83				99%	0.0117	12.18					
TiO <sub>2</sub> /PVDF/PVP/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)

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 Visible light 6h	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] [55]
	3%		82		9.6				75% (for 7.5 wt% TiO <sub>2</sub> )				
	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%								99.8% for 3 wt% TiO <sub>2</sub>				
	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)

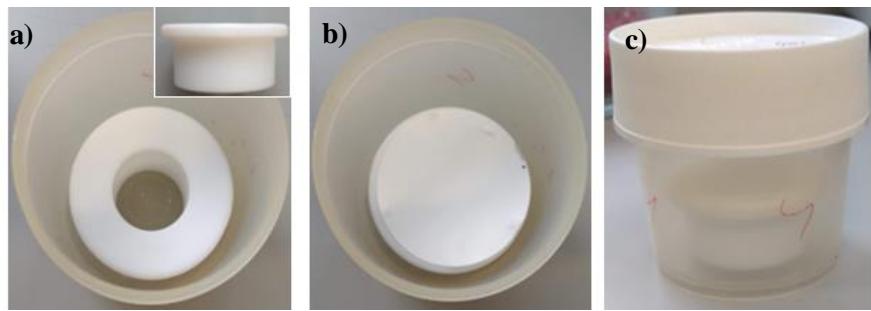
Ag-TiO <sub>2</sub> / APTES/PVP /PVDF/DM Ac	0.1%	82	73		MB	3 mg/L, 30 mL. pH 7	Visible light, 1 h	80.3%				Smaller TiO <sub>2</sub> particles caused intracellular damage, and unique antibacterial performance. Meanwhile, Ag has inhibitory effect on the growth of bacteria; Thus these PMs shows excellent 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		
<b>Flat sheet (Cross-flow)</b>												
TiO <sub>2</sub> /PVDF-TrFE/DMF	8%				Tartz	10 mg/L 20 mg/L 30 mg/L, $v = 28 \text{ mL/s}$ (for all $C_0$ )	Sunlight, 5 h, (38×12) cm <sup>2</sup>	77.77	0.30		Increase in initial feed concentration (10-30 mg/L) reduced the photodegradation of tartazarine (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]
								57.72	0.18			
								46.57	0.12			
3D-TiO <sub>2</sub> /ZnO/PVDF (ALD)		95	40		MB HA	3.2 mg/L, 100 mL 300 mL	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]
								73% (1 h)		35.75		
L-Histidine/TiO <sub>2</sub> /CdS/P VP/PES/DM Ac	0.1% 0.5% 1%	63	51 47 45		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]

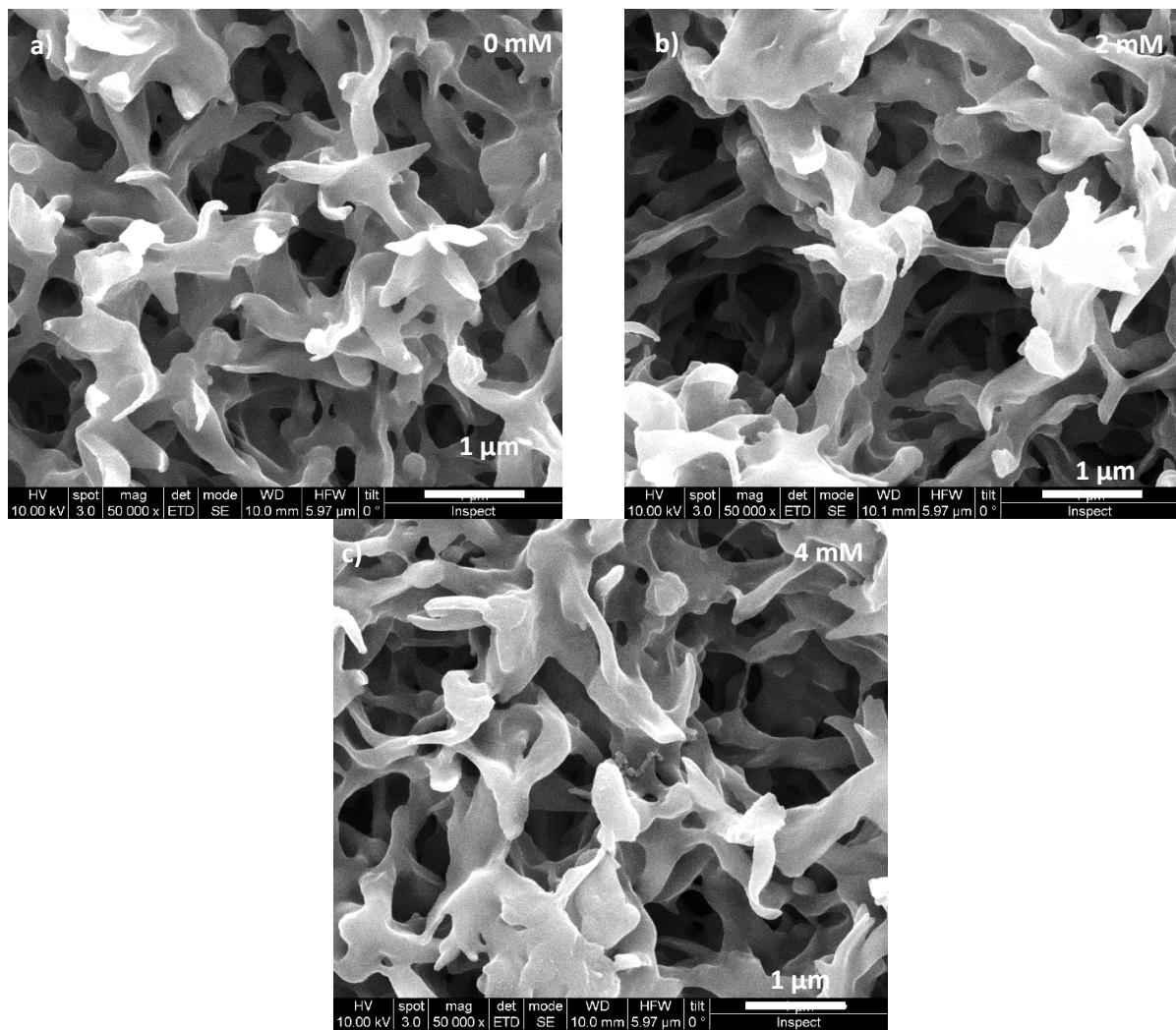
<sup>†</sup> These amounts are deposited onto membrane grafting by PEG and GLUT through vacuum filtration method

<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>

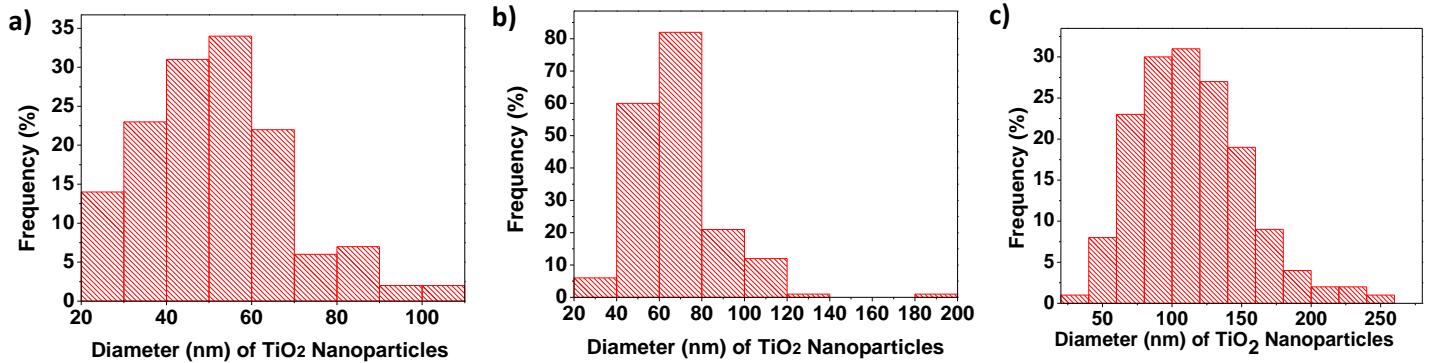
Annex 6 Recipient to crystallize on-site synthesized TiO<sub>2</sub> nanoparticles on membrane



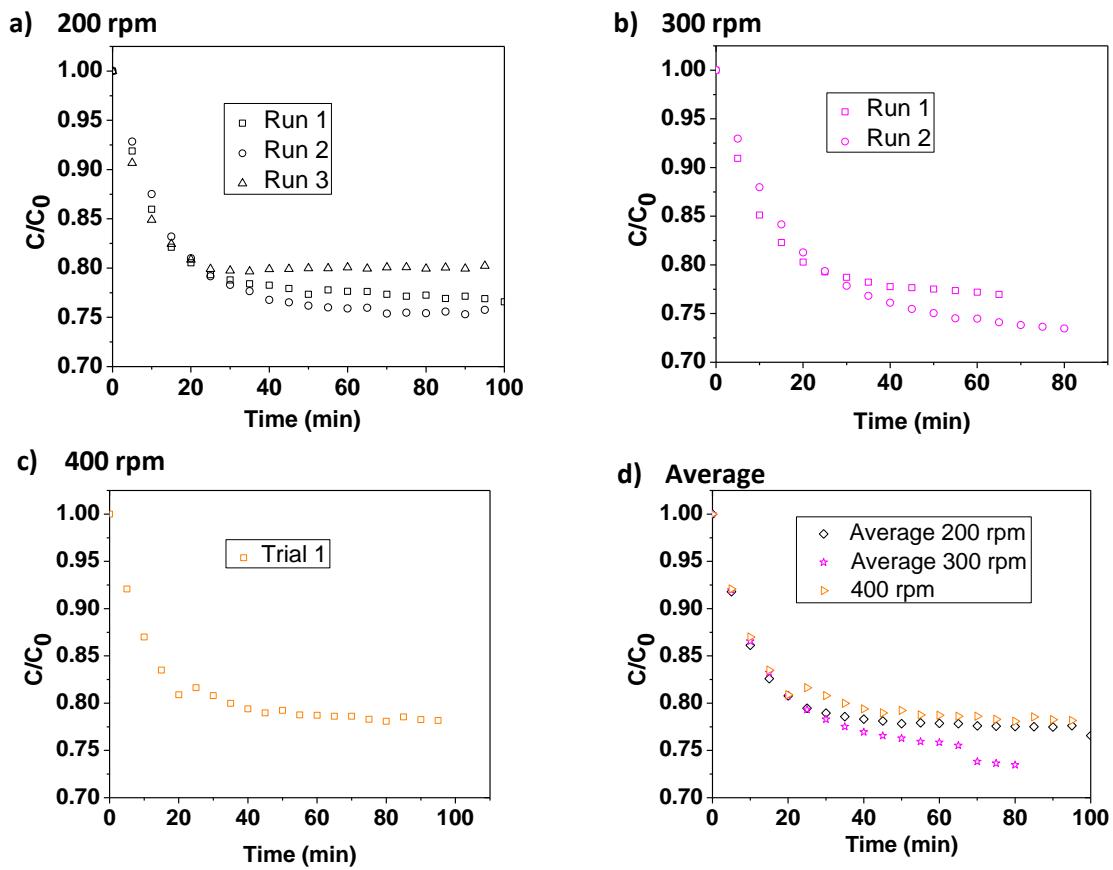
Annex 7 SEM images of the Membranes prepared from lower concentration (2 mM and 4 mM) sol-gel solution; a) Pure PVDF (0mM), b) 2mM, c) 4mM



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

