†Electronic supplementary information (ESI)

Remediation and recycling of inorganic acids and their green alternatives for the sustainable industrial chemical processes

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Fig. S1. Schematic illustration of general working principle of membrane distillation process.

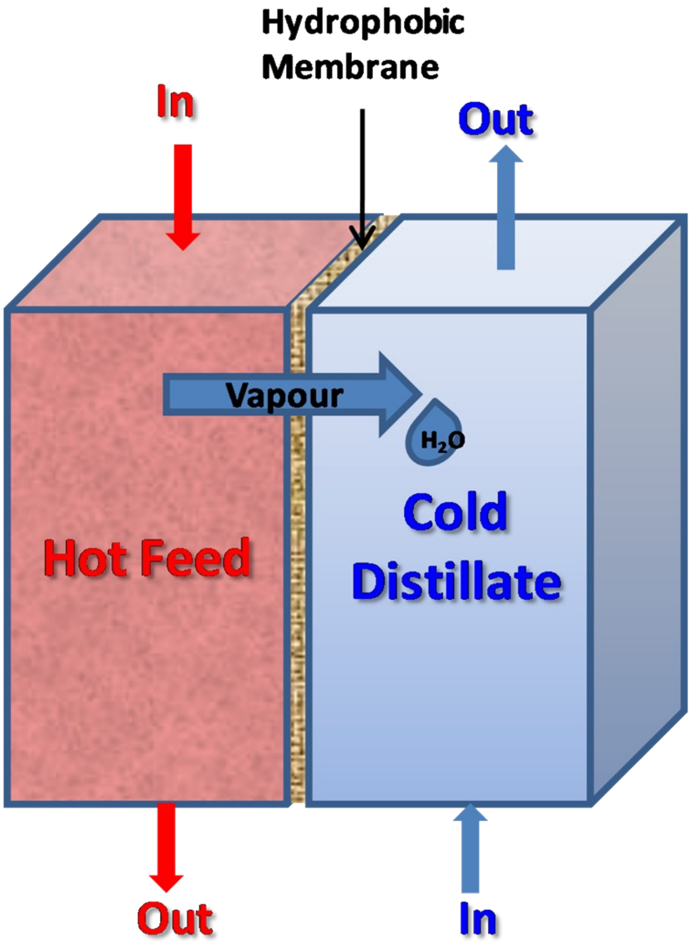

Fig.S2. Conceptual representation of Process Flow Diagram (PFD) of the membrane-integrated process for the recoveries HCl, water and salts from a pickling solution. Adopted from Ref. 336.

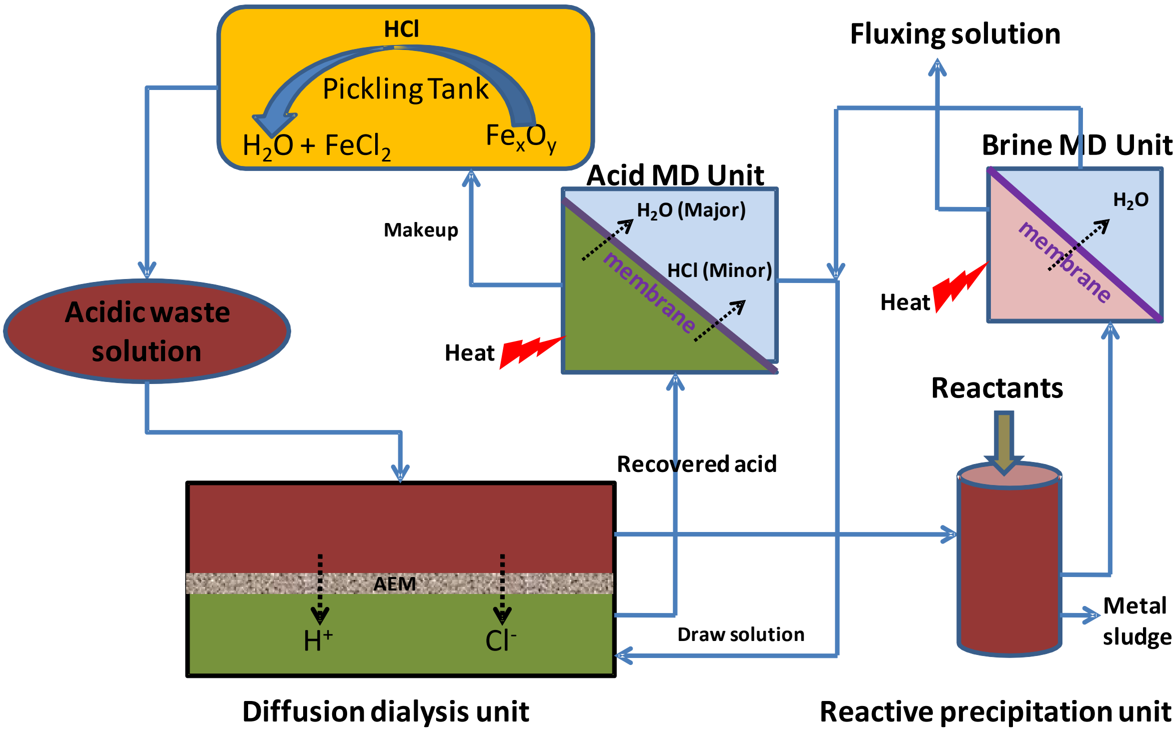

Table S1. Diffusion Dialysis studies and their major outcomes.

Title of the paper	**Aimed application/system studied**	**Membrane composition**	**Major outcome**	**Ref No.**
Recovery of H_2SO_4 from waste acid solution by a diffusion dialysis method	H_2SO_4 recovery in diamond manufacturing process	Selemion DSV	Process found to be feasible and economically viable	1
Hybrid organic-inorganic anion-exchange pore-filled membranes for the recovery of nitric acid from highly acidic aqueous waste streams	High level nuclear waste	PP host with pores filled with polymerized [(3-Acrylamidopropyl) trimethyl ammonium chloride (APMAC) and (3-acryloxypropyl)trimethoxysilane (APTMS)]	Could be successfully demonstrated with simulated waste	2
Tuning the acid recovery performance of poly(4-vinylpyridine)-filled membranes by the introduction of hydrophobic groups	$HCl/NaCl/MgCl_2$	4-vinylpyridine based PP AEM	Performance better than commercial membranes	3
Fabrication of asymmetrical diffusion dialysis membranes for rapid acid recovery with high purity	$HCl/FeCl_2$	brominated poly(phenylene oxide) (BPPO) ultrafiltration membranes with a thin active layer (< 1 micron mm thick)	Process capacity increased with 3 times increase in acid purity	4

Imidazolium functionalized anion exchange membrane blended with PVA for acid recovery via diffusion dialysis process	$HCl/FeCl_2$	Imidazole based membrane prepared by sol-gel process	Higher S than commercial DF-120 membrane	5
One-step fabrication of methylthiazole-functionalized anion exchange membranes for diffusion dialysis	Iron polishing waste solution (1.0 mol·L-1 $FeCl_2$ + 0.2 mol·L^{-1} HCl)	brominated poly(2,6-dimethyl-1,4-phenylene oxide) (BPPO) based membranes with 4-methylthiazole functional groups	U^{H+} and S much higher than commercial DF-120	6
In-situ cross-linked porous anion exchange membranes with high performance for efficient acid recovery	Industrial acid water with HCl (1 mol L^{-1}) and $FeCl_2$ (0.2 mol L^{-1})	Chloromethyl polyethersulfone substrate using N, N, N′, N″, N″-pentamethyldiethylenetriamine as a bifunctional agent for cross-linking and quaternization	Membrane synthesis by one-pot method S two orders of magnitude higher than commercial DF-120	7
Facile surface modification of anion-exchange membranes for improvement of diffusion dialysis performance	HCl (1 mol L^{-1}) and $FeCl_2$ (0.2 mol L^{-1})	Neosepta-AF modified with polypyrrole	S improved with respect to pristine membrane	8
Recovery of H2SO4 from an acid leach solution by diffusion dialysis	H_2SO_4 from an acid leach solution produced	DF120	economic evaluation revealed that an	9

	during the vanadium manufacturing process		investment in this process could be recovered within 27 months	
Recovery of hydrochloric acid from the waste acid solution by diffusion dialysis	$HCl/FeCl_2/ZnCl_2$	DF120	Leakage of other salts observed	10
Separation and recovery of sulfuric acid from acidic vanadium leaching solution by diffusion dialysis	H_2SO_4 from an acid leach solution produced during the vanadium manufacturing process	DF120	Efficient rejection of V, Al and Fe ions wrt H could be shown	11
Poly (triethoxyvinylsilane-co-quaternaryvinylbenzylchloride)/fGNR based anion exchange membrane and its application towards salt and acid recovery	$HCl/FeCl_2$	triethoxyvinylsilane and quaternized vinyl benzyl chloride based AEM modified with functionalized graphene nano-ribbons (fGNR)	Reduction in swelling ratio and improvement in IEC due to functionalized graphene nano-ribbons (fGNR) presence	12

Covalently cross-linked pyridinium based AEMs with aromatic pendant groups for acid recovery via diffusion dialysis	$HCl/FeCl_2$	covalently cross-linked pyridinium based AEMs	performance better than commercial membranes	13
The recovery of sulphuric acid from the waste anodic aluminum oxidation solution by diffusion dialysis	Recovery of H_2SO_4 from waste anodic aluminum oxidation solution	DF120	DD found to be cost effective	14
Recovery of hydrochloric acid from simulated chemosynthesis aluminum foil wastewater by spiral wound diffusion dialysis (SWDD) membrane module	HCl from simulated chemosynthesis aluminum foil wastewater	spiral wound diffusion dialysis (SWDD) membrane	Investment could be recovered within 16.5 months	15
Effective recovery of acids from egg waste incorporated PSf membranes: A step towards sustainable development		egg shell membrane powder loaded on polysulfone membrane	egg shell membrane powder enhances proton permeability of polysulfone membrane	16
Diffusion dialysis membranes with semi-interpenetrating network		semi-interpenetrating networks in polyvinyl chloride (PVC)	performance better than	17

for acid recovery			commercial membranes	
Advanced charged porous membranes with ultrahigh selectivity and permeability for acid recovery	HCl/$FeCl_2$/$AlCl_3$	Porous brominated poly(2,6-dimethyl-1,4-phenylene oxide) (BPPO) membranes	performance better than commercial membranes	18
Anion exchange membranes used in diffusion dialysis for acid recovery from erosive and organic solutions	acid from organic solution containing HCl and glyphosate	quaternized poly(2,6-dimethyl-1,4-phenylene oxide) (QPPO) and polyvinyl alcohol (PVA)	Membranes less erosive and performance better than commercial membrane	19
Graphene oxide embedded "three-phase" membrane to beat "trade-off" in acid recovery	HCl/$FeCl_2$	Three phase graphene oxide sheet based AEMs	Improved efficiency and separation	20
Anion exchange membranes from hot-pressed electrospun QPPO–SiO2 hybrid nanofibers for acid recovery	HCl/$FeCl_2$	quaternized poly(2,6-dimethyl-1,4-phenylene oxide)/Silicon dioxide hybrid material	Performance better than commercial membranes	21
Porous diffusion dialysis	HCl/$FeCl_2$	brominated poly	Performan	22

membranes for rapid acid recovery		(phenylene oxide) (BPPO) ultrafiltration membrane	ce better than commercial membranes	
Porous BPPO-based membranes modified by aromatic amine for acid recovery	$HCl/FeCl_2$	triphenylamine based porous (AEM with brominated poly(2,6-dimethyl-1,4-phenyleneoxide) (BPPO)	Performance comprable to commercial membranes	23
Novel quaternized aromatic amine based hybrid PVA membranes for acid recovery	$HCl/FeCl_2$	quaternized 4,40 -(1,10 -biphenyl-4,40 -diyldioxy)dianiline (QBAPB) polyvinyl alcohol (PVA) based AEM	Performance better than commercial membranes	24
Asymmetrically porous anion exchange membranes with an ultrathin selective layer for rapid acid recovery	$HCl/FeCl_2$	Asymmetric ultrafiltration membrane with a skin layer	Performance better than commercial membranes	25
Improved acid recovery performance by novel Poly(DMAEM-co-γ-MPS) anion exchange membrane via diffusion dialysis	$HCl/FeCl_2$	Novel membranes prepared by free radical polymerization	Performance better than commercial	26

			membranes	
Recovery of sulfuric acid from a stone coal acid leaching solution by diffusion dialysis	sulfuric acid from a stone coal acid leaching solution	DF-120-III	Process found to be feasible	27
Investigation of key process parameters in acid recovery for diffusion dialysis using novel (MDMH-QPPO) anion exchange membranes	$HCl/FeCl_2$	methyl 6-(dimethylamino) hexanoate (MDMH) and poly (2, 5-dimethylphenyl oxide) based AEM	Process parameters studied using full factorial design	28
Quaternized poly(2,6-dimethyl-1,4-phenylene oxide)s with zwitterion groups as diffusion dialysis membranes for acid recovery	$HCl/FeCl_2$	quaternized poly(2,6-dimethyl-1,4-phenylene oxide)s (PPO) anion exchange membranes (AEMs)	Introduction of zwitterionic groups into anion exchange membrane simultaneously improves dialysis coefficient and selectivity	29
Supplementing multi-functional groups to polysulfone membranes using Azadirachta indica leaves powder for effective	$HCl/FeCl_2$	plant waste (Azadirachta indica, neem leaves powder (NP)) as filler to polysulfone (PSf)	Performance better than commercial	30

and highly selective acid recovery		membrane	membranes	
In-situ crosslinked AEMs with self-assembled nanostructure for acid recovery	$HCl/FeCl_2$	tris(2-(2-methoxyethoxy) ethyl) amine (TDA) functionalized bromo poly(2, 6-dimethyl-1, 4 phenylene oxide)	Membranes exhibited better operational stability for 10 consecutive cycles during the entire diffusion dialysis process.	31
High proton selectivity membrane based on the keto-linked cationic covalent organic framework for acid recovery	$HCl/FeCl_2$	keto-linked covalent organic frameworks with high acid-stability was fabricated on the surface of carboxyl-modified polyacrylonitrile (CPAN) ultrafiltration membrane	High separation performance after 10 cycle under strong-acid conditions	32
Fabrication and characterization of pyridinium functionalized anion exchange membranes for acid recovery	$HCl/FeCl_2$	brominated poly (2,6-dimethyl-1,4-phenylene oxide) (BPPO) as a polymer backbone and 4-methylpyridine (MP) based AEM	Performance better than commercial membranes	33

Construction of two dimensional anion exchange membranes to boost acid recovery performances	$HCl/FeCl_2$	2D AEMs were fabricated by layer by layer stacking the graphene oxide nanosheets decorated by imidazolium cations (Im-GO)	High separation performance after 10 cycle	34
Acid-triggered polyether sulfone - Polyvinyl pyrrolidone blend anion exchange membranes for the recovery of titania waste acid via diffusion dialysis	H_2SO_4 recovery from titania waste	PES (polyether sulfone) - PVP (polyvinyl pyrrolidone) blend membranes	Permeability coefficients remain unchanged for 337 days	35
Prepared poly(aryl piperidinium) anion exchange membranes for acid recovery to improve dialysis coefficients and selectivity	$HCl/FeCl_2$	self-organized nanostructured cross-linked AEMs based on poly (aryl piperidinium)	Performance better than commercial membranes	36
Polysulfone/graphene quantum dots composite anion exchange membrane for acid recovery by diffusion dialysis	$HCl/FeCl_2$	chloromethylated polysulfone membranes are prepared with different concentrations of graphene quantum dots	shows potential for acid recovery	37
Novel poly (ionic liquid)-based anion exchange membranes for efficient and rapid acid recovery from industrial waste	$HCl/FeCl_2$	poly (ionic liquids)-based AEMs by the free radical polymerization of 1-butyl-3- vinyl imidazolium bromide, acrylic acid, styrene,	Performance better than commercial membrane	38

		and acrylonitrile under sunlight	s	
Influence of hydrophobic components tuning of poly (aryl ether sulfone)s ionomers based anion exchange membranes on diffusion dialysis for acid recovery	$HCl/FeCl_2$	quaternized poly(arylene ether sulfone)s	Performance better than commercial membranes	39
High-performance porous anion exchange membranes for efficient acid recovery from acidic wastewater by diffusion dialysis	$HCl/FeCl_2$	porous brominated poly (phenylene oxide) membrane	Performance better than commercial membranes	40
Enhanced diffusion dialysis performance of cross-linked poly(aryl piperidine) anion exchange membranes by thiol-ene click chemistry for acid recovery	$HCl/FeCl_2$	quaternized poly(biphenyl piperidine) polymers based AEMs	Performance better than commercial membranes	41
Nanostructured anion exchange membranes based on poly(arylene piperidinium) with bis-cation strings for diffusion dialysis in acid recovery	$HCl/FeCl_2$	AEMs based on poly(arylene piperidinium) with bis-cation strings	Performance better than commercial membranes	42

Internally cross-linked poly (2,6-dimethyl-1,4-phenylene ether) based anion exchange membrane for recovery of different acids by diffusion dialysis	HCl, HNO_3, H_2SO_4	internally crosslinked poly (2,6-dimethyl-1,4-phenylene ether) (PPE) based AEMs	Performance better than commercial membranes	43
Cationic covalent organic framework membranes with stable proton transfer channel for acid recovery	HCl/$FeCl_2$	acid-stable cationic COF (DhaTGCl) layers with intralayer hydrogen bond on the surface of hydrolytic polyacrylonitrile (HPAN) ultrafiltration membrane	High separation performance after 10 cycle	44
Piperazine-functionalized porous anion exchange membranes for efficient acid recovery by diffusion dialysis	HCl/$FeCl_2$	porous chloromethyl polyethersulfone (CMPES) membrane modified by 1,4-dimethylpiperazine (DMP)	Long term excellent thermal stability and acid resistance	45
Investigation on flexible and thermally crosslinked bis-piperidinium-PPO anion exchange membrane (AEM) for electro-kinetic desalination and acid recovery	HCl/$FeCl_2$	Cross-linked membranes prepared using phase di-cationic cross-linking strategy	High dialysis coefficient and separation factor	46
GO-anchored imidazolium based cross-linked composite anion exchange membranes for the enhancement in acid	HCl/$FeCl_2$	graphene oxide (GO) into N-alkylated polyacrylonitrile-co-polyvinyl imidazole copolymer based AEMs	High dialysis coefficient and separation	47

recovery via diffusion dialysis			factor	
Engineering robust RGO/PVA composite membrane for acid recovery via electron beam irradiation	$HCl/FeCl_2$	graphene oxide (GO) and polyvinyl alcohol (PVA) based membranes	Performance comparable to commercial membranes	48
Crosslinking imidazolium-intercalated GO membrane for acid recovery from low concentration solution	$HCl/FeCl_2$	crosslinked imidazolium intercalated GO membrane	Recovery feasible with the synthesized membranes	49
Constructing proton selective pathways using MOFs to enhance acid recovery efficiency of anion exchange membranes	$HCl/FeCl_2$	AEMs based on metalorganic frameworks	Recovery feasible with the synthesized membranes	50
Chemically stable and high acid recovery anion exchange membrane	$HCl/FeCl_2$	N-methyl-4-piperidone (NMPi) based thermoset AEMs	Recovery feasible with the synthesized membranes	51

Table S2. The advantages and `disadvantages of the standalone remediation and recycling methods.

Technology	Method	Advantages	Disadvantages
Remediation method	Neutralization and Precipitation	Simple and inexpensive, suited for low acidity aqueous waste, large volume can be treated	Insufficient for zero waste discharge due to presence of toxic metal ions, slow precipitation and sedimentation, formation of watery sludge or secondary waste
	Pyrohydrolysis	Specific to spent pickle liquor or similar type of aqueous waste	Insufficient for zero waste discharge, energy intensive, generation off toxic gases, secondary waste formation
	Thermal decomposition/ bio-denitrification	Simple and very effective method, suited for low acidity and low volume.	Insufficient for zero waste discharge, Possibility of significant amount of energy consumption, generation of toxic gases, secondary waste formation, and large volume treatment of aqueous waste is not feasible.
	Coagulation and flocculation	Simple, economical and does not require separate unit, often coupled with precipitation.	Secondary waste generation, incomplete metal ions removal, may need pH control to remove metal ions.
	Crystallization	Crystallization is an effective way to get rid of ferrous chloride in water after hydrochloric acid pickling.	The crystallization requires considerable energy, it is challenging to remove from the waste acids strong metallic ions like Fe ions, there isn't a economically viable way to handle the crystal that was removed.
Regeneration method	Solvent extraction	Acid can be selectively removes with appropriate extractant	Requires special plants, high purity acid may not be achieved, capital intensive, does not suit to low water consumption and zero waste discharge.
	Ion-exchange	Selective for ions removal, large volume can be processes	High purity acid not recovered, large volume of aqueous waste may generate, does not suit to low water consumption and zero waste discharge
Membrane technology	Nanofiltration and Reverse	High purity water can be obtained from RO,	RO may be suited for concentrating aqueous waste

	Osmosis	NF can separate acid from multivalent metal ions. Process is faster than any other membrane	not acid recovery, NF membranes are not stable in high acidic condition, and high purity acid is not recovered. Zero waste discharge not possible due to the presence of metal ions in the rejected water.
	Membrane Distillation	Well suited for recovery of high purity water from acidic waste, low waste heat utilization	HCl can vaporize and pass through membrane with water, not suited for recovery of acid from the waste.
	Diffusion dialysis	High purity acid can be recovered and recycled, low energy consumption, economical and simple	Process is slow and only 50% acid from feed can be recovered.
	Electrodialysis	With monovalent selective membrane, electrodialysis can separate acid from aqueous waste	Well suited for the salt removal, and H^+-ions selective cation exchange membrane is required, proton leakage through anion-exchange membrane is problem, and reverse flow of water is also problem, if concentration difference is higher between two compartments separated by the membrane.

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