

# Recent Advances in the Recovery of Ionomer Membranes from Used Membrane Electrode Assemblies: A Review

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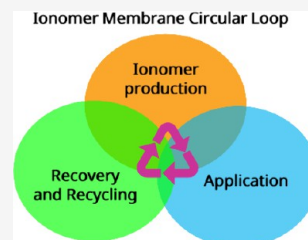
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**ABSTRACT:** The growing demand for proficient, sustainable energy storage and supply technologies has sparked the development of advanced ionomer membranes, electrodes, and metal catalysts. These membranes have critical functions in electrochemical devices, such as fuel cells, electrolyzers, and batteries, by facilitating proton transfer and providing essential reactant barriers. Perfluorosulfonic acid (PFSA) membranes are often favored for their superior proton conductivity and chemical, mechanical, and thermal stability. In proton exchange membrane fuel cells (PEMFC), they feature a sophisticated multilayer system, membrane electrode assembly (MEA) comprising the ionomer membrane, electrode, catalyst layer, and gas diffusion layer (GDL), with bipolar plates (BPs), and end plates used to connect several fuel cells in a stack. However, the lack of recycling processes for the deteriorated ionomer membrane from end-of-life (EoL) secondary sources leads to the accumulation of costly membranes that pose environmental risks and present a significant sustainability challenge due to high material costs. To address these issues, research is focused on sustainable recovery processes for ionomer membranes through dissolution using less hazardous solvents, aiming to minimize waste, maintain membrane integrity, and reduce power input. The objective of this review article is to present an overview of the recent recovery and recycling processes of ionomer membranes from EoL fuel cells and highlight some of the limitations of the membrane recycling processes as sustainable measures.



## 1. INTRODUCTION

Until now, the high global energy demand has intensified the incineration and combustion of fossil fuels, resulting in elevated carbon emissions and consequential degradation of the environment, which are major factors that contribute to climate change.<sup>1–3</sup> The accelerated depletion of nonrenewable energy resources has triggered efforts from the international community to explore reliable or convenient alternative power sources that are sustainable, cost-effective, and environmentally friendly.<sup>4–7</sup> Renewable energy sources available through wind, solar photovoltaics, geothermal systems, hydropower, bioenergy, and fuel cells have previously been identified as attractive potential solutions.<sup>2,8,9</sup> Among these, the fuel cell technologies stand out as one of the most attractive, widely recognized for their ability to convert the chemical energy of fuel directly into electrical energy.<sup>10,11</sup> Unlike combustion energy-generating systems, fuel cells are not limited by the Carnot cycle, due to the absence of mechanical parts,<sup>12</sup> which underlies their high conversion efficiencies.<sup>11–14</sup> Although invented in the 18th century, fuel cells have gained significant scientific research interest since the mid-20th century.<sup>15,16</sup> Over the years, they have demonstrated desirable advantages that include high energy density,<sup>17</sup> clean energy production (low emissions),<sup>18</sup> outstanding efficiency, and compatibility with low-cost fuels such as hydrogen,<sup>19,20</sup> hence making them environmentally friendly.<sup>21–24</sup>

Fuel cells operate on a principle similar to that of batteries, where the electrolyte facilitates the separation of reaction sites

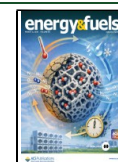
for electrochemical reactions in independent oxidation and reduction phenomena, occurring at the anode and cathode, respectively. This reaction facilitates the separation or generation of electrons and protons.<sup>10</sup> In contrast to batteries, which store energy internally, fuel cells can maintain prolonged, uninterrupted device operation only under a continuous supply of hydrogen fuel and oxygen oxidant.<sup>21</sup> Fuel cell technologies are generally categorized into the following five different groups according to the operating temperature and electrolyte they employ: solid-oxide fuel cells (SOFCs), molten carbonate fuel cells (MCFCs), phosphoric acid fuel cells (PAFCs), alkaline fuel cells (AFCs), and proton exchange fuel cells (PEMFCs).<sup>15</sup> However, direct methanol fuel cells (DMFCs) are sometimes considered as the sixth type of fuel cell technology.<sup>16,25,26</sup> Figure 1 presents different fuel cell technologies, with operating conditions and power density. In this context, the simplicity of the fuel cell design is a key factor behind their integration into transportation,<sup>4,17,27</sup> portable mobile,<sup>28,29</sup> and stationary power-generating systems.<sup>30–33</sup> The widespread integration of fuel cells is expedited

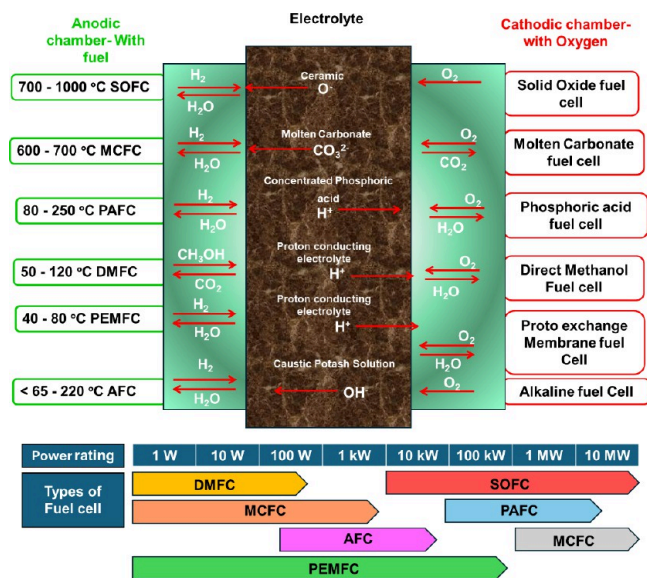
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**Figure 1.** Different fuel cell technologies with their characteristics and operating conditions.

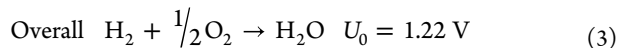
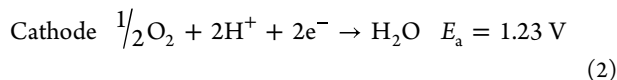
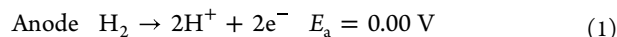
by stacking multiple cells to achieve maximum power output<sup>34–36</sup> Figure 2, with a single cell typically producing a voltage as high as 1.20 V, which is insufficient to meet most current load demand.<sup>34,37</sup> Their integration into daily devices has strengthened global initiatives to reduce carbon dioxide (CO<sub>2</sub>) emissions.<sup>38</sup>

Predominantly, PEMFCs represent the most advanced innovation approaching commercialization more than before. They have drawn substantial attention from many researchers in contrast to other fuel cell technologies.<sup>39,40</sup> PEMFCs offer attractive energy conversion systems with wide power ranges from a few milliwatts to several kilowatts,<sup>41</sup> producing no emissions except water and heat as the other end products.<sup>2</sup> By 2023, the power density of PEMFC-based vehicles had exceeded 3 kW L<sup>-1</sup>, in commercial electric vehicles, demonstrating their scalable potential for sustainable energy solutions.<sup>42</sup> The main advantages of PEMFCs include low operating temperatures, low emissions,<sup>43</sup> rapid start-up,<sup>40</sup> scalability, fast fueling,<sup>4</sup> and large range power (high power density).<sup>44</sup> These advantages, combined with the favorable

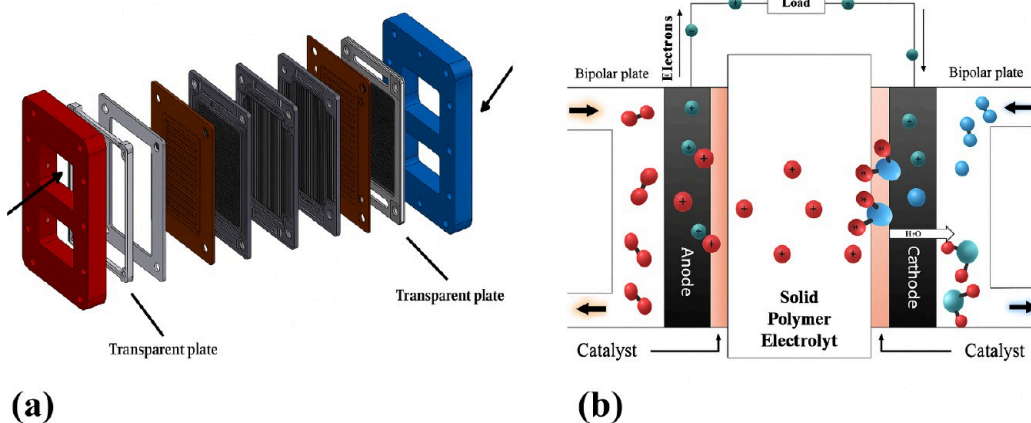
weight-to-power ratio of 0.86 kW/kg,<sup>15,45,46</sup> and high energy conversion rate of approximately 40–60%,<sup>34,47</sup> enable production of versatile energy systems and are environmentally benign.<sup>48</sup>

Despite the appealing advantages, the complete commercialization<sup>49</sup> continues to lag behind other renewable energy options for several reasons including: (i) high cost of main materials,<sup>50–52</sup> (ii) poor durability that results to short-life term,<sup>31,48</sup> (iii) high sensitivity to impurities, requiring ultrapure fuel, (iv) water management resulting in mass transport issues,<sup>30,48,53</sup> (v) fuel cell starvation from low hydrogen availability,<sup>22,54–56</sup> (vi) catalyst aggregation (vii), and membrane degradation,<sup>30</sup> which ultimately contribute to their delayed implementation in major residential and commercial energy systems. The hydrogen fuel introduced is oxidized at the anode, resulting in the formation of protons and electrons, with an activation energy ( $E_a$ ) of 0.00 V.<sup>57</sup> These protons traverse the electrolyte membrane to reach the cathode via membrane channels, while the electrons are directed through the external circuit present between the anode and cathode.<sup>58</sup>

At the anode, hydrogen gas undergoes oxidation to yield protons and electrons with an activation energy ( $E_a$ ) of 1.23 V. The interaction of these electrons and oxygen during the oxygen reduction reaction at the cathode, facilitated by a catalyst, forms water, exhibiting an electrode potential ( $U_0$ ) of 1.22 V, which is subsequently expelled from the fuel cell as water vapor and air.<sup>39</sup> The electrochemical reactions occurring within the PEMFC device are represented by the following equations:<sup>36,59</sup>



PEMFCs primarily feature a membrane electrode assembly (MEA)<sup>60,61</sup> composed of an electrolyte membrane, electrodes, catalyst layers, and gas diffusion layers (GDLs).<sup>52</sup> Additionally, the device structure includes bipolar plates (BPs) and end plates to hold the fuel cell intact and for charge collection.<sup>62,63</sup> Previous research aimed at establishing PEMFCs as reliable



**Figure 2.** (a) Fuel stack. Reproduced with permission from ref 93. Copyright 2020. Elsevier. (b) Structure of the PEMFC device with the operation mechanism. Adapted with permission from ref 94. Available under a CC-BY 4.0 license. Copyright 2020, by Silaa, M. Y.; Derbeli, M.; Barambones, O.; Chekane.

Table 1. Recent Reviews of the Focusing on Recycling of Fuel Cells and Ionomers

review article	publication year	description
Shoaib et al. <sup>80</sup>	2024	provide a comprehensive review of advancements in organic and inorganic electrolytes in RFBs; the study elucidates major challenges associated with deploying Nafion in energy storage devices, and further elucidates LCA studies to identify stages requiring effort to minimize pollution associated with their production
Ye et al. <sup>81</sup>	2024	offer a comprehensive review of the performance of vanadium redox flow batteries (VRFBs) and examine advances in alternative ionomer membranes with superior performance compared to commercially available PFSA membranes; they highlight the shortcomings of PFSA membranes in batteries and their potential environmental impact upon end-of-life disposal
Xun et al. <sup>82</sup>	2025	thoroughly discusses factors influencing the supply of critical materials in fuel cells and water electrolyzers with a focus on the circularity of critical materials; the major challenges for alkaline electrolyzers include resource depletion pressure and substitutability, while polymer electrolyte membrane electrolyzers face supply disruption risks
Zante et al. <sup>83</sup>		the article focuses mainly on the recovery of critical metals from energy storage devices mentions briefly the challenge of recovering polymer electrolyte membranes (PEMs) from EoL materials, which are degraded during the metallurgical recovery process using sulfuric and hydrochloric acids; while acknowledging the presence of other materials, including PVDF, EVA, epoxy resins, and Nafion; ionic liquids may be deployed to detach the organics from the hydrophobic catalyst
Du et al. <sup>84</sup>	2025	discuss the advancements of VRFBs, with emphasis on efforts devoted to improving the performance of Nafion membranes; they highlight that significant modifications and enhancements of Nafion's properties are needed to achieve sustainable applications
Ebner et al. <sup>85</sup>		discuss the environmental impact of VRFB production, including the consequences of Nafion membrane production; they emphasize that recycling critical materials from EoL technologies using less toxic processes could enhance the sustainability of these materials
Ameduri et al. <sup>86</sup>	2023	this manuscript examines the degradation processes applicable to PFSA-based membranes; it further concentrates on the previous recovery of Nafion membranes from dissolution, alongside declaration of sustainable EoL targets by Chemours Company; in general, the manuscript discusses the environmental impact of fluorinated polymers
He et al. <sup>87</sup>		this review focuses on recent developmental efforts to advance the production of both PEMWEs and PEMFCs; the strategies to offset or lessen the cost of production are addressed; the review thoroughly discusses the versatility of Nafion in both PEMWEs and PEMFCs, acknowledging the limitations impacting commercialization due to these shortcomings
Kisti et al. <sup>88</sup>	2025	this review focuses on the recent development efforts to advance the production of both PEMWEs and PEMFCs; the efforts to offset or improve the cost of production are mentioned
Staudacher et al. <sup>89</sup>	2025	this article provides insights into processes involved in the recycling of individual components of PEMWEs, including the nondestructive and automated disassembly of catalyst-coated membranes (CCMs); furthermore, it emphasizes the efficiency of mechanical recycling processes; the variety of PEMWE designs poses a challenge in developing a universal recycling method, and several stack designs are thus discussed
Li et al. <sup>90</sup>	2024	this article provides insights into processes involved in the recycling of individual components of PEMWEs, including the nondestructive and automated disassembly of CCMs; furthermore, it emphasizes the efficiency of mechanical recycling processes; the variety of PEMWE designs poses a challenge in developing a universal recycling method, and several stack designs are thus discussed
Mvokwe et al. <sup>91</sup>		the review presents the recent methods investigated for the recovery electrocatalyst metallurgical process; the study also discusses the efficiency of previous studies as well as challenges
Kusoglu et al. <sup>92</sup>		this study presents detailed review of PFSA membranes, including their properties and performance

energy systems is earmarked by significant progress and breakthroughs toward improving their stability and durability.<sup>64,65</sup> Achieving a desirable combination of cost,<sup>66</sup> reliability, efficiency, durability, and performance is essential to afford PEMFCs as a viable solution to address the high global energy demand.<sup>67</sup> The Nafion membrane, initially produced by DuPont, has rapidly become one of the most widely established and used membranes in fuel cell technologies presently.<sup>44</sup> Despite their attractive advantages, Nafion faces several challenges, including high cost, complex maintenance requirements, hydrogen infrastructure, reliability, and durability concerns. The Nafion membrane itself accounts for approximately 30% of the total cost of electrochemical devices, while the noble catalyst materials and BPs contribute to the remaining high-cost fuel cells.<sup>15</sup>

The nondegradable nature of ionomer membranes poses a considerable environmental threat if end-of-life (EoL) membranes are discarded without appropriate recycling or proper disposal processes.<sup>68</sup> Moreover, fuel cells contain hazardous materials that present environmental and health risks upon disposal.<sup>69</sup> Particularly, some of these materials can persist in the environment for thousands of years, and their global warming potential (GWP) values are 4 orders of magnitude higher than that of CO<sub>2</sub>.<sup>2</sup> A significant limitation of fuel cell hydrogen (FCH) technologies is the lack of effective EoL management strategies that minimize the demand and utilization of raw materials. For sustainability considerations, it is critical to develop recycling methods that are both economical and environmentally viable.<sup>70</sup>

Recyclability signifies one foremost sustainability goals that ensures access to essential materials, while concurrently reducing environmental impacts associated with material utilization and waste disposal.<sup>69</sup> The PEMFCs exemplify sophisticated devices that contain both precious metals in the nanoparticle form and complex polymers, making them equally challenging to recycle and thereby presenting a meaningful challenge for the recycling industry. To date, most published studies address hydrometallurgical recovery, focusing mainly on the platinum recovery from the electrocatalyst.<sup>3</sup> The recycling of platinum group metal-based (PGMs) electrocatalysts, together with other critical materials, is of significant environmental importance because of their scarcity.<sup>71</sup> Recent studies have focused on reducing the production costs of fuel cells by investigating the recyclability of the main fuel cell components.<sup>72,73</sup> However, these studies have prioritized recycling the PGMs electrocatalysts owing to their limited accessibility, which is unevenly distributed across Zimbabwe, Russia, South Africa, and the United States.<sup>74</sup> These valuable metals are considered rare metals due to their scarcity. Pyrometallurgy and hydrometallurgical processes are among established methods for recycling electrocatalyst from spent MEAs of fuel cells.<sup>73,75,76</sup>

The recycling of proton electrolyte membrane (PEM) through dissolution using organic solvents and recasting has previously been reported.<sup>69</sup> The high electronegativity and small atomic radii of the fluorine atom that confer a short and quite stable carbon–fluorine (C–F) bond, fluorinated products have remarkable chemical inertness and thermal stability (the dissociation energy of this C–F bond is approximately 120 kcal mol<sup>-1</sup>).<sup>77</sup> Some attempts have been undertaken to recycle noble catalyst materials and electrolyte membranes in MEAs of PEMFCs.<sup>71</sup> Several patents have already been issued about the methods addressing the

recycling of perfluoro sulfonic acid (PFSA) [perfluoro (4-methyl-3,6-dioxane-7-octene-1-sulfonyl fluoride)]<sup>78</sup> membranes, yet few research articles have investigated the solution properties of PFSA polymers and preparation techniques.<sup>79</sup> In pursuit of enhancing the sustainability, circular hydrogen economy, and environmental compatibility of fuel cell technologies, the recycling of ionomer membranes would have to be studied. In recent years, significant scientific interest has been dedicated to recycling PFSA ionomer membranes, aiming to offset the costs of the membrane, mitigating the environmental risk, and ensuring a sustainable supply of critical materials. Major issues in reusing recycled membranes include cost effectiveness and feasibility of processes involved in preparation of the ionomer solutions, recasting, and quality evaluation of the recast membrane, primarily in terms of mechanical and ion transfer properties.<sup>79</sup> It is noteworthy that the PFSA membrane may potentially be a toxic membrane due to fluorine emissions.<sup>71</sup> While significant progress has been documented, the field now stands at a pivotal point where understanding the interplay between synthesis, stability, and large-scale application is crucial for commercialization.

As one of the global commitments to reducing CO<sub>2</sub> emissions and transitioning to cleaner energy solutions, a comprehensive exploration of material recyclability in fuel cells and other technologies is required. However, the reliance on recycling only PGMs presents a risk that may impede the large commercialization of fuel cells or lead to an explosion of the market, as historically evidenced by the sharp price increase of catalytic converters. Thus, it is imperative to investigate the recyclability of other materials within fuel cells, particularly those of a higher value. To clearly outline the contribution of this review paper, Table 1 compares its scope and focus against existing key reviews in the field of recycling ionomer membrane and fuel cells. Although multiple studies have concentrated on the reclamation of metals from fuel cells, there is a need to explore progress in the recycling of ionomers from varied energy storage systems such as redox-flow batteries (RFBs), electrolyzers, and fuel cells. The number of publications on the development of ionomer membranes has steadily increased over the years, as evident in Figure 3. This review seeks to address a critical gap in understanding the

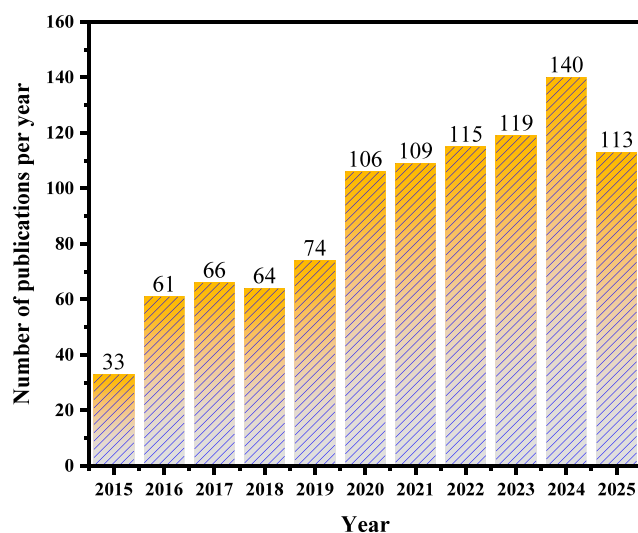


Figure 3. Number of publications per year on the development of ionomer membranes from 2015 to 2025.

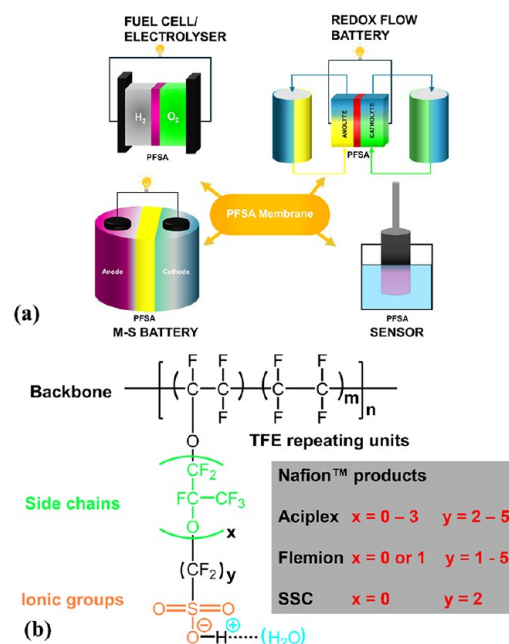
recycling of ionomer membranes, presenting key considerations and frameworks for these processes. Emphasizing the critical role of membrane recycling in the adoption of sustainable fuel cell technologies as energy sources. This systematically discusses different fuel cell membranes and the recovery and recycling of MEAs. Specifically, more focus is placed on the dissolution of used membranes under environmentally friendly conditions, highlighting challenges with potential implementation for industrial upscaling. It includes introducing membrane degradation mechanisms. Additionally, a section was organized to provide a brief discussion on life cycle assessment (LCA) studies of the EoL membrane. This article aims to provide valuable insights into emerging experimental limitations to complete the commercialization of fuel cell technologies, while supporting sustainable and economic pathways.

## 2. BACKGROUND

### 2.1. Proton-Exchange Membranes

In MEAs, proton-exchange membranes serve as an integral component, sandwiched between catalyst layers,<sup>28</sup> with GDLS on either side completing the device structure.<sup>95,96</sup> Membranes must exhibit high chemical and mechanical stability to withstand harsh operating conditions and multiple electrochemical cycles.<sup>2</sup> Commercially, membranes have significantly reduced thickness in micrometers to reduce ohmic proton polarization, decrease the size and volume, and improve power density.<sup>42</sup> The ionomer has key functions in isolating diffusion of reactant gases within the catalyst layers.<sup>38,40,97</sup> Furthermore, it simultaneously prevents the crossover of negatively charged electrons, allowing only the movement of protons through the anode.<sup>33,98</sup> The selective electron flow to the external circuit generates electrical energy, allowing electrons to reach the cathode through the external circuit. Affirmatively, the membrane exhibits high ionic conductivity for protons, which migrate across toward the cathode,<sup>99</sup> combining with oxygen to produce water and heat.<sup>100</sup> Additionally, the membrane also provides support to the catalyst-containing electrode layers.<sup>101</sup> The side chain of the ionomers directly influences proton conductivity and oxygen permeability.<sup>102,103</sup>

Numerous ionomers with different molecular weights have been successfully developed to date.<sup>104</sup> These materials are generally characterized by main parameters: (i) equivalent weight (EW), (ii) ion exchange capacity (ICE), and (iii) side-chain length, as depicted in Figure 4. Studies have shown an inverse relationship between EW and membrane stability, with lower EW membranes demonstrating reduced degradation resistance.<sup>103</sup> The most widely used membranes for PEMFCs are the PFSA-based membranes such as the Flemion, Aciplex, Gore-select, Aquivon, Hayflon, and Nafion<sup>45,101,105,106</sup> as shown in Figure 4. These membranes are typically constructed by the copolymerization of unsaturated perfluoroalkyl sulfonyl fluoride (PSF) with tetrafluoroethylene (TFE)<sup>107–109</sup> generally forming a semicrystalline main chain polytetrafluoroethylene hydrophobic backbone<sup>5,41</sup> with repeating sulfonic ionic groups (SO<sub>3</sub><sup>-</sup>) hydrophilic side chains.<sup>110,111</sup> They exhibit high proton conductivity under the hydrated state and stability at temperatures up to 100 °C; however, their performance significantly declines under low humidity conditions at higher temperatures above approximately 150 °C.<sup>32,108,112</sup> The presence of fluorine atoms with high electronegativity and a

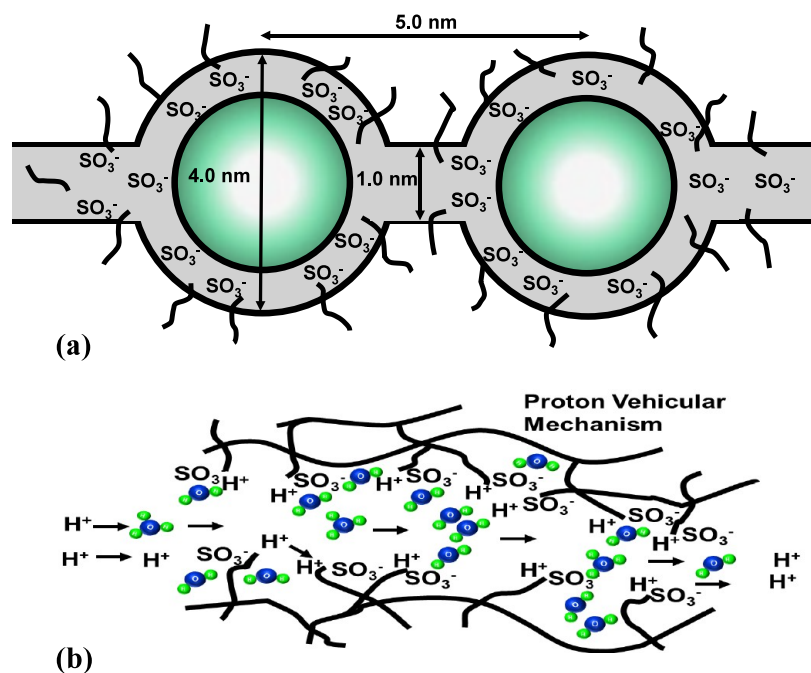


**Figure 4.** (a) Electrochemical devices with the PFSA membrane and (b) structure of the PFSA membrane with different side-chain lengths.

small radius contributes to the exceptional mechanical properties.<sup>86</sup>

The sulfonic groups at the end of the side chains provide the membrane with ion-conducting properties, while hydration of the membrane ensures sustained conductivity.<sup>27,113</sup> The sulfonic ion clusters are interconnected by channels which are used for mass transport and gas transport, with phase separation from the hydrophobic backbone and hydrophilic sulfonic side chains.<sup>114</sup> This phase-separated morphology affords the PFSA with its unique solvent-transport characteristics and outstanding ion-conducting performance.<sup>38</sup> Additionally, the high crystallinity of the membrane inhibits dissolution in water, especially under humid conditions, thereby improving membrane durability,<sup>103</sup> as shown in Figure 5. However, their practical application is hindered by reduced conductivity, notable under low relative humidity, and gas permeability at elevated temperatures.<sup>32</sup> Additionally, the Nafion membranes exhibit inherent limitations in permeating fuel crossover from the anode to the cathode, as demonstrated with methanol crossover in DMFCs.<sup>115</sup> Moreover, ohmic losses occur when the membrane loses its ionic conductivity, particularly under dehydration conditions.<sup>112</sup>

Membrane with a PFSA general structure is widely utilized in various electrochemical applications, including RFBs, direct alcohol fuel cells (DAFCs), polymer electrolyte membrane water electrolyzers (PEMWEs), regenerative fuel cells (RFCs), and both low- and high-temperature fuel cells.<sup>107</sup> Additionally, PFSA has extended use in the chlor-alkali industry as a sodium-ion conductor.<sup>92</sup> Among the listed PFSA-based ionomers that are commercially available, Nafion and Aquivon<sup>106</sup> are the two most common. whereas long-side chain (LSC) Nafion membrane, formerly produced by DuPont (now Chemours), exhibits exceptional proton conductivity of 0.008–0.130 S/cm<sup>27,29,53</sup> at 100% relative humidity and 75 °C, and durability of above 60,000 h.<sup>98</sup> Meanwhile, short-side chains (SSC) Aquivon exhibit delayed structural changes and resistance to less dry-wet after each cycling while maintaining enhanced



**Figure 5.** (a) Representation of the water channels in the ionomer membrane. (b) Diagram of proton diffusion and transportation in typical pristine membrane.

crystallinity.<sup>116</sup> Aquivion ionomers have previously gained attention due to their higher degree of crystallinity and thermal stability, positioning them as both suitable for high-temperature fuel cell technologies and a dispersion in catalyst material. Attention is being paid to SSC. This membrane offers favorable advantages, including reduced production cost, enhanced proton exchange, lower reactant or gas crossover, exceptional mechanical properties, and chemical stability.<sup>116</sup> Comparative studies have established a significant preference for Aquivion over its long-sided counterpart, Nafion, in terms of stability and optimum performance under harsh conditions.<sup>117</sup> Table 2 summarizes the categories of various ionomers used in fuel cell technologies along with their characteristics.

The pursuit to improved efficiency and commercialization of PEMFCs has driven the development of alternative ionomer membranes including sulfonated polyether ether ketone (SPEEK),<sup>120</sup> sulfonated polyether sulfone (SPES),<sup>121,122</sup> and sulfonated polyphenyl sulfone (SPPSU),<sup>123,124</sup> sulfonated poly(arylene ether)s (SPAES)<sup>125</sup>/sulfonated poly(arylene ether sulfone) (SPAES),<sup>12</sup> and polybenzimidazole (PBI)<sup>126</sup> Figure 6. These advanced membranes aim to address the limitations of conventional PFSA membranes and achieve a superior fuel cell performance.

## 2.2. Membrane Degradation

Numerous studies have been conducted on the degradation mechanism of fuel cells.<sup>101</sup> The fuel cell experiences some irreversible degradation to some extent; the degradation gradually increases with the aging of the components, thus reducing the efficiency of the fuel cell device.<sup>45</sup> The performance of the fuel cell decreases over time due to the mechanical, thermal, and chemical degradation.<sup>31,127</sup> Degradation can occur from the early life of the fuel cell until the total breakdown of the device occurs,<sup>15</sup> due to manufacturing errors.<sup>128</sup> Fuel performance decreases significantly during

degradation, owing to increased internal ionic resistance and poor efficiency.<sup>34</sup>

Membrane degradation in PEMFCs results from different processes within the device itself. Electrochemical aging, chemical, thermal, and mechanical conditions are some of the main causes of membrane failure, leading to end-of-life MEAs.<sup>129</sup> The performance of the fuel cell diminishes as major components deteriorate.<sup>130</sup> Different factors affecting the condition or status of the membrane can induce specific degradation mechanisms, which are categorized into mechanical, thermal, and chemical degradation.<sup>36</sup> Therefore, it is crucial to understand the different degradation modes occurring within fuel cells. Mechanical degradation of the membrane triggers premature failure of fuel cell stacks when tears, cracks, and delamination occur. Fabrication defects are the major factors that affect the integrity of the membrane beforehand. The thermal and humidity changes during the cell operation lead to uneven microscopic dimension changes to the membranes, which exert mechanical localized stress on the membranes.

Generally, device degradation occurs during the fuel cell operation under harsh conditions that exert prolonged stress on the components.<sup>131</sup> Damage experienced on the membrane is noticeable with the formation of cracks on the catalyst layers and a pinhole appearance on the membrane. These defects potentially lead to the crossover of reactant gases.<sup>45</sup> The surface morphology of the Nafion sampled examined with scanning electron microscopy (SEM) micrographs images shows substantial degradation that hinders the performance of the membrane in fuel cells; Figure 7. The comparison between the cross-sectional views of the pristine Nafion 112 MEA and the degraded MEA is depicted by using SEM images in Figure 7a,b. In the pristine MEA, there is an absence of irregular conformities, such as fractures or delamination, between the membrane and the highly porous catalytic layer. The ionomer membrane exhibits a compact structure with no visible signs of

Table 2. Types of Ionomer Membranes with Properties<sup>a</sup>

type of membrane	structure	advantages	disadvantages	operating temperature (°C)	ionic properties
perfluorinated	<ol style="list-style-type: none"> <li>1. fluorinated backbone</li> <li>2. presence of the SO<sub>3</sub>H proton exchange group of the side chain</li> </ol>	<ol style="list-style-type: none"> <li>1. high proton conductivity</li> <li>2. excellent chemical and physical stability in fuel cell conditions</li> <li>3. high durability in fuel cells</li> </ol>	<ol style="list-style-type: none"> <li>1. high price</li> <li>2. poor performance in DMFC (methanol crossover)</li> </ol>	<100 °C with long chains	0.01–0.03 S/cm <sup>2,50</sup>
partially fluorinated	<ol style="list-style-type: none"> <li>1. fluorinated base</li> <li>2. hydrocarbon or aromatic side chain</li> </ol>	<ol style="list-style-type: none"> <li>1. lower price</li> <li>2. proton conductivity increases by grafting the proton exchange group in the structure of the side chain</li> </ol>	<ol style="list-style-type: none"> <li>1. significant performance decrease at high temperature, and anhydrous conditions</li> <li>1. low durability value compared to the perfluorinated membrane</li> <li>2. low performance</li> </ol>	>150 <sup>50</sup>	
nonfluorinated	<ol style="list-style-type: none"> <li>1. hydrocarbon base, functionalized with proton-conducting groups</li> </ol>	<ol style="list-style-type: none"> <li>1. high mechanical strength</li> </ol>	<ol style="list-style-type: none"> <li>1. poor chemical and thermal stability</li> </ol>	<200 <sup>o,119</sup>	~0.01 S/cm <sup>2,119</sup>
acid–base blend	<ol style="list-style-type: none"> <li>1. grafting of the acid component onto the alkaline polymer base</li> </ol>	<ol style="list-style-type: none"> <li>2. low price</li> <li>1. good chemical and thermal stability</li> </ol>	<ol style="list-style-type: none"> <li>2. poor proton conductivity</li> <li>1. poor durability</li> </ol>		
ionic liquid-based	<ol style="list-style-type: none"> <li>1. organic cation or inorganic anion</li> </ol>	<ol style="list-style-type: none"> <li>2. good proton conductivity compared to Nafion</li> <li>1. high proton conductivity</li> <li>2. high chemical and thermal stability</li> </ol>	<ol style="list-style-type: none"> <li>1. difficult to obtain a solid membrane</li> </ol>		

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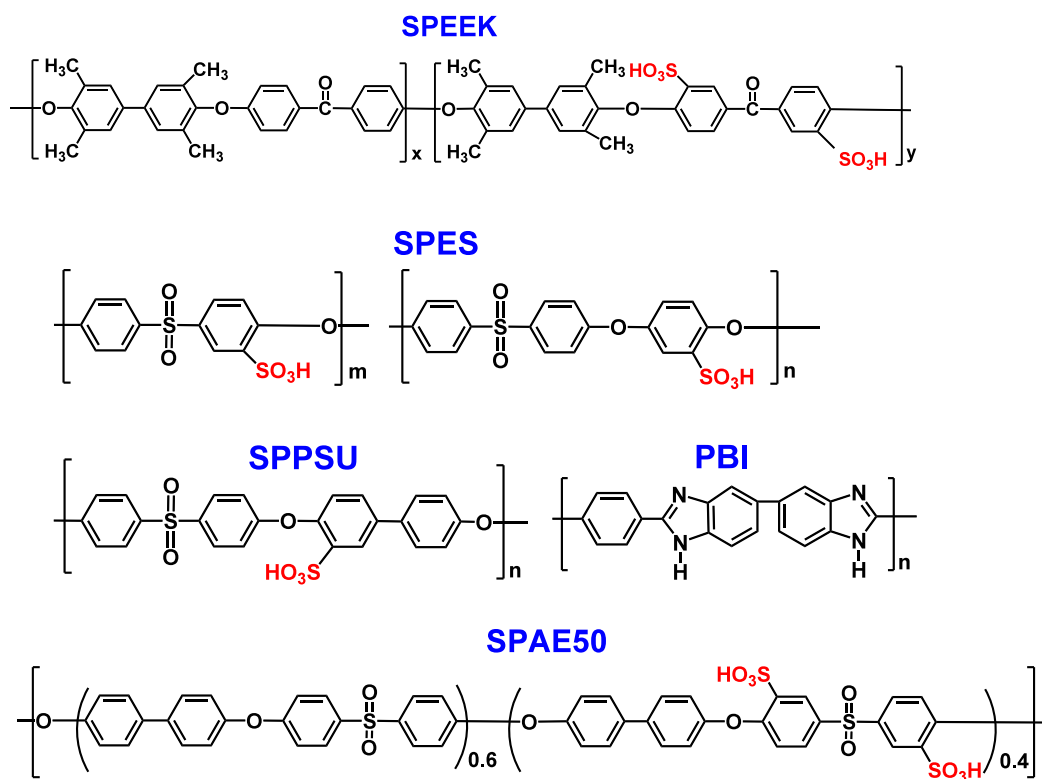
deterioration. However, the cross-sectional images in Figure 7b reveal alterations in the membrane structure when compared to the pristine MEA.<sup>132</sup> Additionally, membrane thinning is noticeable, which may affect gas crossover and result in ohmic losses. The thinning of the membrane facilitates the possible formation and propagation of pinholes. As illustrated in Figure 7c–g, pinhole formation occurs within a membrane subjected to accelerated stress testing (AST), which simulates the degradation of the membrane under actual fuel cell operating conditions.<sup>45,133</sup>

### 3. EOL OPTION FOR USED IONOMER MEMBRANE IN FCH TECHNOLOGIES

The use of high-value materials in FCH technologies raises serious concerns over the long-term handling of EoL or flawed Nafion over the long term. Accumulation of these materials presents a challenge that will likely be addressed either by reprocessing or by disposal. However, the latter process poses an environmental threat.<sup>134</sup> One of the proposed strategies for handling PEM from spent FCH technologies is reusing and recycling with appropriate, sustainable, eco-friendly methods<sup>135</sup> Figure 8. The application of fuel cells in fuel cell vehicles requires the following qualitative steps in their recycling efforts: (i) collection, (ii) dismantling, (iii) disassembly, and (iv) preprocessing.<sup>136</sup> Each of the steps plays an important role in the recycling cycle to achieve a sustainable circular economy. The use of high-value critical materials significantly compromises the economic feasibility of hydrogen technologies. It is necessary to optimize EoL strategies that fall within the sustainability, environmental friendliness, and feasibility targets.<sup>137</sup> Although the recyclability of PFSA-based membranes within various energy and electrochemical technologies has been demonstrated, this does not inherently result in fully established sustainability. Instead, it serves as a standard for a prospective circular hydrogen economy and substantial recycling efforts, which can facilitate the adoption of FCH technologies for commercial applications, while mitigating the demand for critical materials and reducing the costs of fuel cell technologies. Further optimization of the circularity could serve as a strategic framework for advancing sustainable practices while adhering to both economic and ecological considerations.

#### 3.1. Disassembly of FCH Technologies and PEM Delamination

The collecting and dismantling of fuel cell stacks from energy systems (stationary or mobile) necessitate the discharge and other treatment processes before the systems are dismantled.<sup>136</sup> Disassembly of decommissioned PEMFC stacks into constituent components is critical for accessing and evaluating EoL materials' condition. This process primarily involves a labor-intensive process to expose and separate individual cells from the fuel cell stack. Recent studies have copulated the process of dismantling fuel cells, as demonstrated in Figure 9. Disassembly of fuel cells serves as an important step in implementing a sustainable circular economy through repurposing, recycling, and remanufacturing.<sup>139</sup> In a recent study, Robert et al. investigated the various routes for the separation of membranes and electrodes from spent MEAs, as a primary step for membrane recycling. In this study, diluted isopropyl alcohol solutions under stirring are used in the proposed two-step process. The process has reported a high



**Figure 6.** Structures of examples of other available ionomer membranes with different degrees of sulfonation.

recovery rate of approximately 95% PFSA, avoiding emissions and requiring low energy consumption.<sup>140</sup>

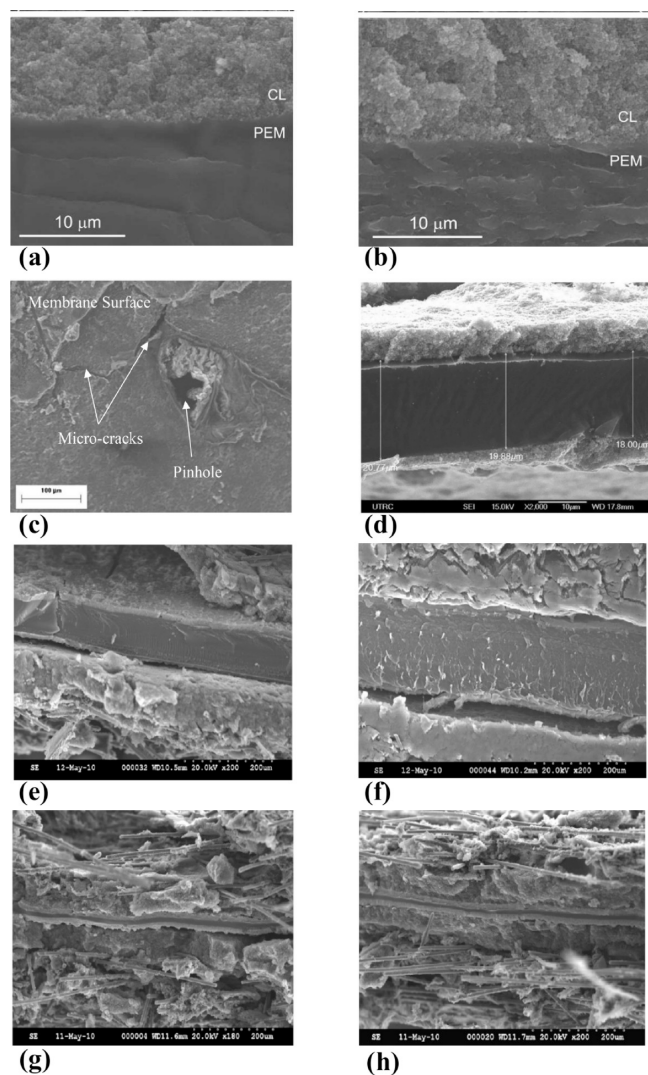
Delamination or isolation is a required preprocessing step for recycling the ionomer membrane through solvent treatment in lower alkyl alcohol. Historically, several studies have reported on the successful isolation of MEA using chemical methods that include soaking the MEA in 50% isopropyl alcohol or in an aqueous alcohol solution.<sup>141</sup> Shore, in 2009,<sup>142</sup> discovered that the lower alkyl alcohol can be used to separate the ionomer membrane from the electrode materials. Furthermore, the alcohol can dissolve the membrane when heated under mild conditions, with the ultimate step involving the filtration of the noble metal catalyst. The process is effective in separating the MEA into individual materials, namely, the PEM, CL, and GDL, with an additional process required to refine the individual materials. For instance,<sup>143</sup> delaminated PFSA membrane from a used MEA by immersing in boiling isopropyl alcohol, while Sreeraj achieved the membrane delamination by immersing the MEA in isopropyl alcohol (IPA). Improper delamination may potentially lead to loss of PGMs<sup>136</sup> and could additionally pose a health risk to personnel working on the stacks.<sup>144</sup> Similarly, Sharma et al.<sup>69</sup> conducted the delamination of MEAs through a solvent wetting process using 96% (v/v) ethanol solution, which was followed by a physical separation. This method enabled the recovery of PEM and the removal of the PGM catalyst, as documented in the studies executed by ref 145.

The delamination of the MEA is consistent with challenges as encountered during the initial process during the recovery of the Pt from the spent PEMFC stack, MEA being intact on the GDL by as Nafion as a binder.<sup>141</sup> Carmo et al.<sup>146</sup> demonstrated membrane delamination using an isopropanol/water solution in a custom reactor, achieving complete separation within 10–30 min; Figure 10. In 2019,<sup>147</sup> reported

membrane separation was reported by cyclic wetting/drying (15 min in water or 0.1 M HNO<sub>3</sub> followed by drying at 80 °C, repeated 3–5 times). Additionally, Parnian et al.,<sup>134</sup> achieved delamination of the PEM from the catalyst layer via dissolution of the Nafion membrane in a water/ethanol solution under high-pressure conditions (240 °C, 6 h, N<sub>2</sub> atmosphere). Recovering the ionomer membrane from FCH technologies can significantly promote the sustainability of this technology.

### 3.2. Recent Advances in EoL PEM Handling

Under normal circumstances, undamaged BPs and EP are often reused directly after sequential structural assessment and cleaning. This approach of recycling material from EoL sources instigated efforts to recover high-value material, including those that degraded to minimize environmental threat due to exposure and, most importantly, reduce the cost of FCH technologies. Such advancements not only make them desirable but also increase adoption as sustainable energy supply systems. Currently, four proven recovery strategies are available for EoL MEAs: (i) a high-temperature combustion process, (ii) an acid dissolution process, (iii) an electrochemical (process-based) recovery route, and (iv) an alcohol treatment method. However, these techniques are more recognized primarily for electrocatalyst material recovery.<sup>149</sup> The recovery of the PGMs has technically matured more than PEM and other materials, such as GDL. This is evident with the use of metallurgical methods to leach or recover Pt electrocatalyst materials from fuel cell electrodes. Furthermore, the pyrometallurgical process requires a calcination step and treatment with aqua regia, therefore generating toxic fumes and requiring energy input of energy. Currently, the environmental risks associated with poly-/per-fluoro alkyl substances (PFAS) materials and Nafion inherently hinder widespread application and full-scale establishment of fuel cells.<sup>92</sup> Section 2.2



**Figure 7.** (a) SEM images of the cross-section of degraded MEAs, (b) Nafion 112 MEA adapted with permission from ref 132. Copyright 2011. Elsevier. (c) Pinhole and crack formation and (d) thinning of the membrane, respectively. Reproduced with permission from ref 45. Copyright 2019. Elsevier, (e) N115, (f) N117, (g) NR211, and (h) NR212 ionomer membrane after degradation. Reproduced with permission from ref 133. Copyright 2012. Elsevier.

comprehensively discusses the morphological and structural changes resulting from membrane degradation. These alterations render direct reuse of PEM after Pt/C removal without regeneration via chemical treatment undesirable.<sup>144</sup> More recently, Robert et al. proposed a recovery process for used PFSA-based membranes after physical delamination and separation of the Pt and carbon material. Subsequently, the membrane dissolution was carried out at 75 °C, leading to the recovery of the membrane as an ionomer dispersion.<sup>140</sup>

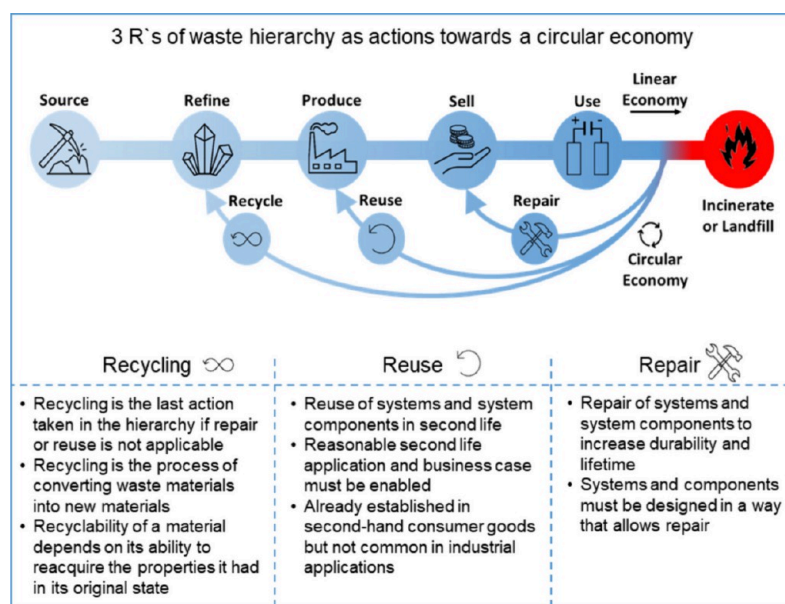
There is a growing demand for the establishment of a circular economy to address the global waste accumulation challenges predominantly through repair or material prolongation of EoL devices.<sup>150</sup> To reduce the environmental threat posed by Nafion and other PFAS materials, a circular economy approach has been proposed, applicable across the material lifecycle. This approach is divided into three strategies: (i) smart product manufacturing and use, (ii) product or component advancement to extend functional durability, and

(iii) diverse material application. Besides the economic benefits of reusing high-value materials from the EoL technologies, it also offers an opportunity for developing newer technologies from recycled materials.<sup>149</sup> In the next section, the terms “recovery” and “recycling” are distinguished as follows: **Recovery** refers to the chemical processing of used fuel cell stack components into solutions, while **Recycling** denotes the subsequent reapplication of the material dissolutions via recasting, yielding a new membrane for potential second-life applications.

**3.2.1. Recovery of EoL Membranes.** The initial phase of membrane reclamation focuses on the selective dissolution of the ionomer membrane itself from the EoL sources. This section presents the organic solvent recovery method with more focus on the efficacy of membrane dissolution exhibiting degradation. The recovered membrane can be processed by chemical treatment to dissolve the membrane into an ionomer dissolution or dispersion. However, the PFSA membrane remains insoluble at lower temperatures and requires high temperature and high pressure to successfully prepare the desired ionomer dissolution for membrane recast.<sup>134</sup> The method of recovering an ionomer membrane from used sources through dissolution in organic solvents is the main method. This approach was first described in earlier studies by Xu et al.<sup>151</sup> In the work, the use of the PFSA membrane Nafion is subsequently recovered as a recast membrane after being dissolved in dimethyl sulfoxide (DMSO) at 170 °C. Herein this study, the use of organic solvents is met with several disadvantages, such as the dissolution of Nafion membranes from the EoL fuel cell stack was achieved using DMSO under ambient conditions. Additionally, the dissolved membrane was inherently reproduced into a recast membrane at 170 °C. This studies further report that the recast membrane demonstrated similar performance to the untreated pristine membrane in terms of water uptake, membrane resistance, water uptake and exchange capacity, however, with superior strength compared to the pristine membrane.<sup>151</sup>

Koehler et al.<sup>152</sup> reported on the recovery of ionomer membrane from used CCMs under critical conditions. In the actual experiment, a three-layer CCM was processed in a stainless-steel autoclave with a stirrer, at a temperature of 374 °C and a pressure of 240 bar for a duration of 7 h. The resulting green solution with black residuals was obtained and filtered. Whereas the greenish solution was concentrated with the possibility of being cast into recycled membranes or in the production of new catalyst inks. Notably, Li et al.<sup>153</sup> demonstrated the effective solubility of Nafion powder using aprotic polar solvents such as DMSO during the preparation of Nafion-composite membrane casting solution.

Bharti et al.<sup>149</sup> reported on the recovery of ionomer form used MEAs as powder. However, this process could potentially lead to the emission of toxic HF arising from fluorinated-containing materials (PFSA and PTFE). Presented a low-temperature hydrothermal treatment using a 50:50 v/v of water and isopropanol (IPA) solution under ambient temperature. Further adds that the treatment with the IPA: water solution facilitates the detachment of the Nafion membrane from the coated Pt/C catalyst material and additionally the dissolution of the membrane via hydrothermal treatment, with subsequent separation of the Pt/C by filtration. In contrast, Robert et al.<sup>154</sup> successfully separated aged membrane from the MEA using a brush soaked in a water/ethanol solution.



(a)



(b)

**Figure 8.** (a) Circular economy flow diagram reproduced from a previous study.<sup>138</sup> Available under a CC-BY 4.0 license. Copyright 2024 by Matz, L.; Bensmann, B.; Hanke-Rauschenbach, R.; Minke, C. (b) Life cycle of the used PFSA membrane. Adapted with permission from ref 135. Copyright 2024. Elsevier.

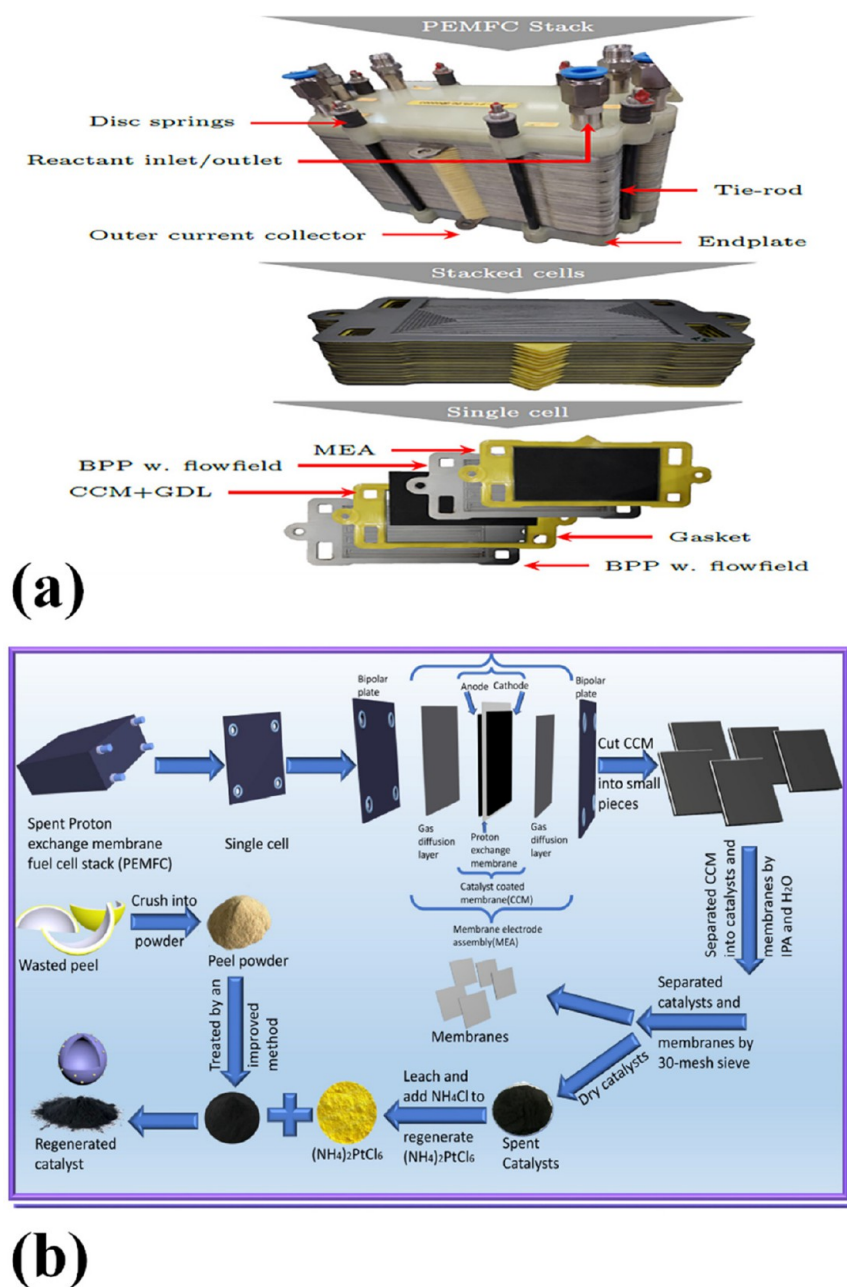
Subsequently rehydrated the membrane was immersed in deionized water overnight.

Zeng et al.<sup>148</sup> also reported on the separation of the PEM from CCM by immersion in deionized (DI) water and IPA (1:1, v/v) and stirred for 1 h. The mixture was filtered using a 30-mesh sieve, and the PEM was obtained by dissolution, whereas the Pt/c suspension was obtained. The PEM solution was further processed by sonication for 10 min in 30 mL of DI water and IPA (1:1, v/v), with complete recovery after drying at room temperature. Xu et al.<sup>71</sup> immersed/dipped CCMs in 0.5 M Sulfuric acid for 2 h at 150 °C, with the resulting sample crushed and dried at 80 °C under a vacuum oven. The PFSA resin and Pt black sediments were separated in a centrifuge at 6000 rpm for 10 min. Dissolution of ionomers [Nafion D2020 (EW 1000), Aquivion D98-25BS (EW 980), and Aquivion D83-24B (EW 830)] in solubilized at 0.2 g/mL in anhydrous dimethylacetamide (DMAc) in the process of producing blend membranes as a binder for catalyst ink is reported.<sup>155</sup>

Pratama et al.<sup>156</sup> prepared an ionomer solution by dissolving 3 M PFSA powder in *N*-methyl-2-pyrrolidone (NMP) at 25 °C under moderate stirring. Subsequently, the PFSA solution was cast onto a glass substrate using a doctor blade. Ma et al.<sup>157</sup> reported on the preparation of solid Nafion resins by dissolving dried Nafion in the following solvents: IPA-H<sub>2</sub>O (4:1), *N,N*-dimethylformamide (DMF), MeOH-H<sub>2</sub>O (4:1), NMF, ETOH-H<sub>2</sub>O (4:1), and DMAc. Tsai et al.<sup>158</sup> also reported the dissolution of chopped Nafion in DMF in the process of preparing/in preparation of Nafion-based composite membranes. The formation 5 wt % Nafion solution for 12 h at temperatures near 200 °C in a Teflon-lined stainless-steel autoclave. Parnian et al.<sup>134</sup> adopted a similar procedure for preparing Nafion dissolution when preparing ZrO<sub>2</sub> nano-composite membrane by using a high-pressure reactor (Berghof – BR-100) at 240 °C for 6 h under a nitrogen atmosphere to dissolve weighted pieces of Nafion 117 in a water: ethanol (50:50) solution. Additionally, the Nafion dissolution was concentrated and introduced in DMAc with

temperatures increased to above 90 °C to remove the water and ethanol solvents. Pozio et al.<sup>159</sup> report dissolution of Nafion 112 with an EW of 1020 g mol<sup>-1</sup> in DMF using a sonicator. The study dissolved Nafion 117 membranes in a water/ethanol solution (50:50) using high-pressure conditions (240 °C, 6 h, N<sub>2</sub> atmosphere) to regenerate the Nafion ionomer, enabling membrane recycling. The dissolved solution was filtered to remove impurities before further processing (Table 3).

In the most recent study by Kutter et al.<sup>135</sup> demonstrated a solvent-free hydrothermal method for dissolving PFSA membranes (800 EW and 1000 EW) using only water at 230 °C under pressure in a Parr Instruments pressure vessel. Unlike previous approaches that relied on organic solvents (e.g., alcohols and dimethyl sulfoxide) or harsh acids, this method avoids toxic chemicals and simplifies scalability. The process yielded a homogeneous ionomer dispersion, which was then used for recasting. Spectroscopic analysis (FTIR, Raman, NMR) confirmed that the ionomer's chemical structure remained intact, indicating no degradation during dissolution. This method is particularly advantageous for industrial recycling due to its environmental safety and efficiency. Both short side chain (SSC) and long side chain (LSC) ionomers with EWs of 800 and 1000 were investigated. In contrast to previously published methods, we were able to recover the PFSA ionomer by using water instead of high-boiling organic solvents such as DMF or DMSO. The recycled membranes were obtained from the recycled PFSA ionomer solution by doctor-blading. They were investigated by in situ Raman techniques for studying water uptake and analyzed by FTIR techniques to get a more detailed insight into chemical change. In single-cell tests at standard (80 °C) and enhanced (up to 130 °C) PEMFC operating temperatures, the recycled membranes showed increased cell performance as compared to the pristine 3M-800 EW membrane. Solid-state NMR measurements confirmed that no chemical changes in the membrane occurred due to the heat-treatment process. In



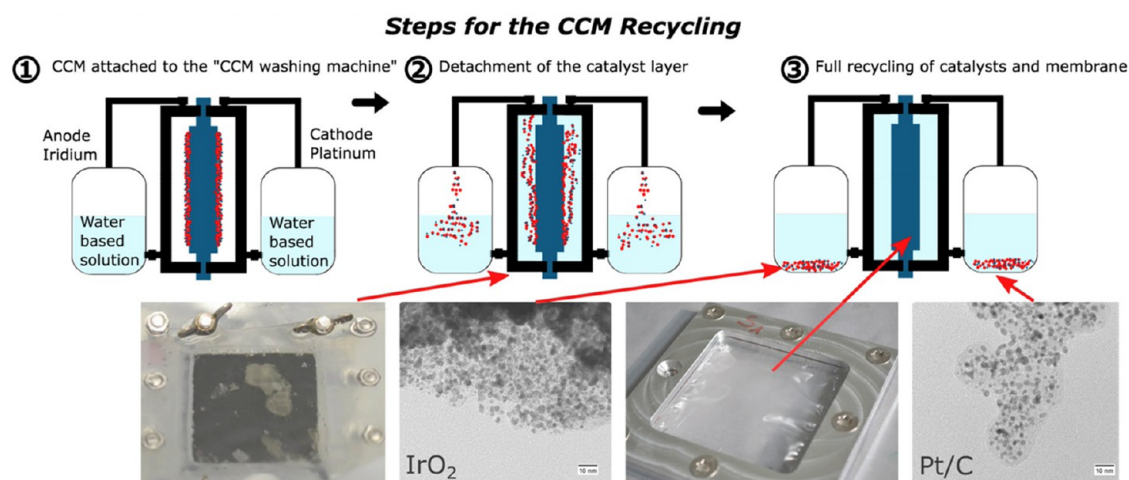
**Figure 9.** (a) Photograph of the dismantled fuel cell stack. Reproduced with permission from ref 139. Copyright 2023. Elsevier. (b) Schematic diagram of dismantling and recycling PEMFCs. Reproduced with permission from ref 148. Copyright 2022. Elsevier.

order studies Carmo et al.,<sup>146</sup> The study employed sequential boiling treatments ( $3\% \text{H}_2\text{O}_2 \rightarrow 1 \text{M} \text{H}_2\text{SO}_4 \rightarrow \text{DI water}$ ) for purification. The properties of the cleaned membrane, characterized by electrochemical testing, demonstrated retained functionality with only a 40 mV performance loss at  $2 \text{A}/\text{cm}^2$  versus pristine membranes. The delaminated membrane was processed through boiling hydrogen peroxide (3 wt %) and sulfuric acid (1 M) treatments, followed by thorough water rinsing. The properties of the cleaned membrane, characterized by electrochemical testing in reassembled cells, demonstrated retained functionality with only a 40 mV performance loss at  $2 \text{A}/\text{cm}^2$  compared to those of pristine membranes. Table 4 summarizes the recovery efficiency of PFSA using different alcohol-based solvents. Uekert et al. suggest that recycling the PFSA membrane via the dissolution route may currently offer a more progressive and cost-effective

comparison of the inherent PFSA manufacturing. Despite the promising results, this process requires a design that will enable a closed-fluorine loop to prevent fluorine emissions to the environment.<sup>73</sup> This challenge needs to be overcome to remain compliant with international emission policies.

### 3.2.2. Recycling of EoL Membrane by Casting.

Succeeding in the recovery of the ionomer membrane, the recycled membrane undergoes either the recasting process or the preparation as a binder material. This section mainly evaluates the recasting procedure. Key considerations include analysis of the functional group, water uptake, mechanical strength, and electrochemical properties of the reclaimed membrane. The complete recycling of PEM is driven by the following factors: (i) a complicated fabrication process that is also time-consuming, (ii) their high economic value, and (iii) ecological benefits that minimize the demand for raw material



**Figure 10.** Delamination of the MEA. Adapted with permission from ref 146. Copyright 2019. Elsevier.

**Table 3. Summary of the Recovery Efficiency of PFSA Using Different Alcohol Solvents in Patent Work Adapted from a Previous Study<sup>161</sup>**

membrane material	solvent	efficiency (%)
PFSA membrane	ethylene glycol	80.0
PFSA membrane	propylene glycol	85.0
PFSA membrane	1,2-butanediol	81.0
PFSA membrane	1,3-butanediol	25.0
PFSA membrane	1,3-butanediol	75.0
PFSA membrane	1,4-butanediol	10.0
PFSA membrane	1,5-butanediol	35.0
PFSA membrane	1,5-butanediol	85.0
CCM <sup>162</sup>	water, alcohols, acetonitrile, diethyl ether, dioxane, dimethyl ether	98.9
MEAs <sup>163</sup>	-	16.2

and waste generation. Moreover, the recycling procedure developed for PEM, particularly dissolution, can be adapted to PEMWEs and other technologies that can be adapted to PEMWEs and other technologies that use similar materials, thus this underscores the broader industrial relevance.<sup>164</sup>

Currently, the most promising method for recycling remains the dissolution or dispersion of EoL ionomer membrane in organic solvents.<sup>165</sup> In 2005,<sup>166</sup> had already demonstrated successful drying casting of Nafion membrane from Nafion dispersions prepared by ethyl glycol, DMSO, and DMF. Sreeraj et al.<sup>68</sup> demonstrated the recycling of recovered PFSA membranes through solvent casting. The purified membrane was dissolved in a high-boiling polar solvent (e.g., dimethyl sulfoxide or alcohol/water mixtures) at elevated temperatures (80–150 °C) to form a homogeneous ionomer dispersion. The solution was then cast onto a flat substrate using a doctor blade, dried at 60–80 °C, and annealed at 120–180 °C to enhance mechanical stability. Post-treatment included protonation in 1 M H<sub>2</sub>SO<sub>4</sub> and thorough rinsing to remove residual acid. This process, while effective, increased membrane crystallinity due to sulfonic anhydride formation, reducing

water uptake and proton conductivity. The study highlighted the trade-off between recyclability and performance retention in solvent-based recycling methods.

Grot and Grot<sup>162</sup> presented patent report on the recovery of PFSA ionomer membrane from the slurry of CCM materials, using water and solvent solutions at temperatures between 190 and 290 °C and high pressure in an autoclave for extended periods. This process involves the reapplication of both the membrane and catalysts to prepare a new CCM. Carmo et al.<sup>146</sup> The study formulated a comprehensive recycling methodology for all CCM components, achieving recovery rates exceeding 96% for both platinum and iridium catalysts. The characterization of the recycled materials, conducted via TEM and electrochemical testing, revealed a preserved catalyst particle size with only minor performance degradation, indicated by a 40 mV increase at 2 A/cm<sup>2</sup>. The membrane recycling process effectively circumvented the emission of hazardous HF and exhibited the potential for large-scale application, with projections suggesting adequate material availability for GW-scale electrolyzer deployment utilizing this method. The properties of the entire recycling system, evaluated through mass balance calculations and economic analysis, demonstrated its viability for industrial implementation, achieving recovery rates of 97.9% for Ir and 96.8% for Pt. Parnian et al.<sup>134</sup> The recovered Nafion ionomer was mixed with DMAc, cast onto glass plates, and dried (100 °C, 2 h) to produce recast membranes. Post-treatment included boiling in H<sub>2</sub>O<sub>2</sub>, H<sub>2</sub>SO<sub>4</sub>, and DI water. Characterization showed that the recast Nafion retained key chemical groups (FTIR) but had lower proton conductivity than pristine Nafion due to potential sulfonic group degradation during dissolution. SEM confirmed dense, defect-free microstructures. While the recast membrane maintained structural integrity, its reduced conductivity highlights the need for optimization to match commercial Nafion performance. The study primarily used this method to prepare ZrO<sub>2</sub> nanocomposites.

The dissolved Nafion was recycled into new membranes using three distinct methods, as illustrated in Figure 11.<sup>143</sup> In Method 1, the Nafion solution was concentrated by evaporation, mixed with DMAc, cast onto glass plates, and thermally treated at 100–145 °C. Method 2 employed a NaOH pretreatment step to enhance the membrane's thermal stability before recasting, while Method 3 involved neutralizing the solution with NaOH, drying to obtain Nafion crystals, and

Table 4. Summary of Recovery and Recycling of Ionomer Membrane by Various Chemical Methods

material	recovery process	solvent	remarks	references
EoL MEAs	hydrothermal treatment	isopropanol aqueous solution	Pt/C 98.7% recovered, and Nafion membrane recovered as an ionomer-enriched solution; ionomer powder was obtained after heat treatment of the ionomer solution for 12 h at 80 °C	149
N115	dissolution and recasting	DMSO	membrane recasting by heat treatment at 150–170 °C under N <sub>2</sub> atmosphere	151
800 EW	hydrothermal process	propanol: water (95:5)	increased water uptake and proton conductivity	135
1000 EW	hydrothermal process	ethanol: water (60:40)	showed maximum power density at 80 °C and low relative humidity	135
Waste Nafion	microwave heating	alcohol: water	the formed Nafion membrane resin was then reinforced with PTFE to form composite membranes	171
MEA membrane	microwave heating	methanol, ethanol, isopropanol, and butanol: water	filtration was used to remove the catalyst and carbon residue from the ionomer solution	172
PEM samples	microwave synthesizer	ethanol	supercritical conditions were employed for the dissolution of the ionomer membrane	69
Nafion D2020	dissolution	DMAc	the Nafion powder was dissolved in DMAc as a casting solution, which was modified with SPAEKs, to form SPAEK-Nafion composite membranes	173
Nafion	refluxing	DMSO	the Nafion-DMSO solution was cast into new membranes at a temperature of approximately 120 °C	174
Nafion 177	dissolution	ethanol and DMSO	the used Nafion from vanadium flow batteries was dissolved in 50% ethanol solution, resulting in membrane dispersion; DMSO was used to replace the dispersion solvent and to cast a recycled membrane with enhanced mechanical properties	99
Nafion	dissolution	dimethylformamide (DMF)	new membrane was cast from the Nafion-DMF solution on a Petri dish at temperatures around 60 °C	175
Nafion	dissolution	DMF, NMP, DMSO, ethanol, and water.	the Nafion resin was redissolved in the solvents: DMF, DMSO, ethanol/water, and NMP; membranes were cast at temperatures between 100 and 180 °C	176

## Nafion® Membrane Recycling Methods

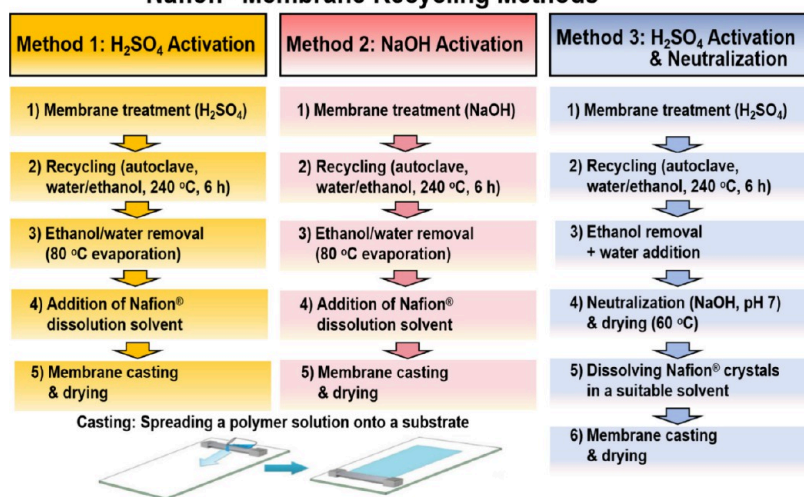


Figure 11. Recycling method investigated developed by Alipour Moghaddam et al.<sup>143</sup> Reproduced with permission from ref 160. Copyright 2025. Elsevier.

redissolving in DMAc prior to membrane formation. The recast membranes exhibited key properties comparable to pristine Nafion 117, including ion exchange capacities of 0.88–0.90 m equiv/g, temperature-dependent proton conductivity, and dense, defect-free microstructures confirmed by SEM. While the recycled membranes demonstrated slightly reduced chemical stability in Fenton tests, attributed to residual catalyst ions, they maintained satisfactory mechanical strength, particularly those prepared via Method 1. These findings demonstrate that properly recycled Nafion membranes can achieve performance characteristics approaching those of commercial Nafion, validating their potential for fuel cell applications. In contrast, Sharma et al.<sup>69</sup> approached the recovery of membranes from delamination by dissolving them

in ethanol under supercritical conditions within a microwave synthesizer (utilizing a CEM Discovery SP). The reinstatement of sulfonic groups was attempted on the N-3R01 sample through sulfonation via reflux in 2 M aqueous sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) for 6 h. Additionally, sulfonation was attempted on the N-3R02 sample using a 2 M chlorosulfonic acid (ClSO<sub>3</sub>H) solution in dichloromethane (CH<sub>2</sub>Cl<sub>2</sub>) for 20 h.

Safronova et al.<sup>167</sup> studied the properties of Nafion membrane recast from NMP dispersion solvent. It was found that the Nafion membrane cast from the NMP dispersion demonstrated higher conductivity than commercially available Nafion 212 membranes. Additionally, both membranes illustrated similar ion-exchange capacity and water uptake performance/properties. Kutter et al.<sup>135</sup> After dissolution, the

Table 5. Comparison of Performance of the Pristine and Recycled Membranes

type of membrane	description of the recycled membrane
800EW, 800EW_re <sup>135</sup>	<ul style="list-style-type: none"> <li>the thickness decreased from <math>35 \pm 2</math> to <math>32 \pm 3</math> <math>\mu\text{m}</math> for the pristine and recast membranes, respectively</li> <li>there was a slight decrease in the IEC from 1.21 for the pristine 800 EW membrane to <math>1.20 \text{ mmol g}^{-1}</math> 800 EW_re</li> <li>there was an increase in equivalent weight (EW) from <math>831 \pm 15</math> to <math>833 \pm 15 \text{ g mol}^{-1}</math></li> </ul>
1000EW, 1000EW_re <sup>135</sup>	<ul style="list-style-type: none"> <li>the thickness decreased from <math>75 \pm 3</math> to <math>72 \pm 3</math> <math>\mu\text{m}</math> for the pristine and recast membranes, respectively</li> <li>there was a slight increase in the ion exchange capacity (IEC) from 1.12 for the pristine 1000 EW membrane to <math>1.13 \text{ mmol g}^{-1}</math></li> <li>additionally, there was an increase in equivalent weight (EW) from <math>852 \pm 15</math> to <math>854 \pm 15 \text{ g mol}^{-1}</math></li> </ul>
Nafion 212, Nafion 212 <sup>68</sup>	<ul style="list-style-type: none"> <li>the crystallinity of the pristine membrane was measured at <math>22 \pm 3\%</math>, whereas the recycled membrane exhibited a crystallinity of <math>29 \pm 6\%</math></li> <li>the amorphous region of the recycled membrane accounted for <math>71 \pm 6\%</math>, in contrast to the pristine membrane, which had <math>78 \pm 3\%</math></li> <li>the mechanical strength of the recovered membrane was enhanced</li> <li>a decline in fuel cell performance was observed as depicted in the polarization curve</li> <li>there was a 32% increase in crystallinity in comparison to the pristine membrane, with a strength augmentation of 35% and reductions in free volume, as well as percentage decreases in water uptake and ion conductivity of 22 and 35%, respectively</li> <li>the recycled membrane displayed a reduction in proton conductivity by 45%</li> </ul>
Nafion 117 <sup>143</sup>	<ul style="list-style-type: none"> <li>reduction results in decreased water uptake by the recycled membrane, which can directly impact its conductivity</li> <li>the chemical stability, as evaluated by weight loss, of the recycled membrane is superior during the Fenton test compared to the recast membrane produced by method 1</li> <li>recycled Nafion exhibited an IEC of 0.846 m equiv/g, whereas the recast (Re-1) and commercial membranes demonstrated IECs of 0.881 and 0.0903 m equiv/g, respectively</li> </ul>
— <sup>69</sup>	recovered membrane used in the fabrication of gas diffusion electrodes; similar FTIR results was observed for recovered membranes compared to the commercial membrane; N-3R01 demonstrated sulfur content (%S) 9% below the pristine membrane

recovered ionomer dispersion was recast into new membranes using the same solvent-based method as for pristine membranes (propanol/water for 800 EW, ethanol/water for 1000 EW). The reprocessed membranes exhibited similar chemical properties but showed reduced mechanical strength (lower Young's modulus, decreased glass transition temperature). However, they displayed improved water uptake and retention, enhancing performance in fuel cells under high-temperature and low-humidity conditions. This suggests that hydrothermal recycling effectively preserves ionomer functionality while slightly altering the membrane microstructure, making it a viable approach for sustainable membrane reuse. Table 5 shows the summary of the recovery and recycling of ionomer membrane by various chemical methods.<sup>140</sup> suggests that ionomer membranes exhibiting severe chemical degradation have the potential for reuse in purification technologies, including antifouling coating, pervaporation, and ultrafiltration. Consequently, following characterization, these recovered ionomer membranes may serve as essential materials in alternative technological applications. Meanwhile, a previous report<sup>69</sup> demonstrated the second life application on recovered membrane, both with and without sulfonation, for the preparation of new gas-diffusion electrodes. Such a development could positively influence the membrane circular economy and underscore the versatility of these recovered membranes.

### 3.2.3. Potential Primary Sources for PFSA Recycling.

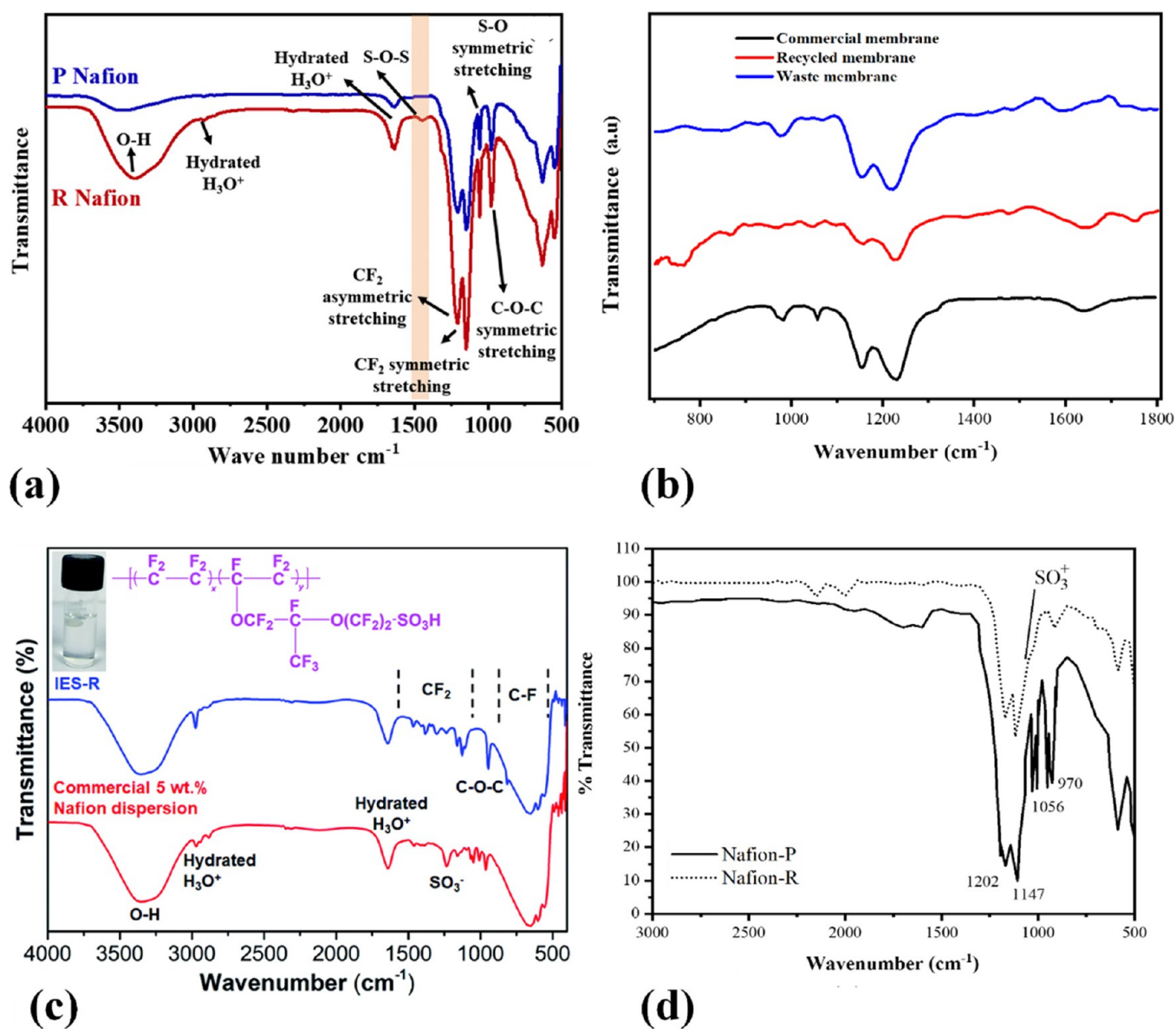
The scope of this review solely focuses on the recovery of PFSA-based membranes from fuel cells. These membranes are fundamental materials that have a critical application across a variety of technological energy sources. Their application in PEMWEs is on the rapid rise due to advancements heeding calls to transition to renewable energy, more especially green hydrogen.<sup>168–170</sup> The calls to transition or enhance the deployment of hydrogen technologies do not sufficiently consider the anticipated high-volume demand for PFSA membranes. Furthermore, the transportation of materials pertinent to FCH technologies, as well as hydrogen gas itself, must be addressed. The extensive utilization of these materials

in RFB designs is well-documented in the literature, demonstrating substantial performance in VRFBs.<sup>99</sup> reported on the recovery of waste Nafion 117 using ethanol following a VFB lifespan test, noting vanadium concentrations in the dispersion of approximately 74 mg/L. In the VRB, Nafion is employed to maintain electrical balance by separating electrolytes. The degradation of the battery necessitates efforts to mitigate costs associated with the replacement of new membranes, which consequently lead to vanadium losses. The objective of reclaiming the waste membrane and suitably recycling vanadium resources can significantly promote resource circularity. Notably, the literature lacks publications concerning the recovery of waste membranes from spent VFBs. Thus, the findings and insights from this Perspective can potentially influence or drive the recovery of PFSA from technologies beyond FCH technologies. Given the component similarities between PEMWEs and PEMFCs, advancements in the recycling of PFSA from fuel cells might be applicable to electrolyzers.<sup>89</sup> conceptualized the recycling chain of PEMWEs, placing emphasis on recycling with minimal disruption during dismantling and mechanical processing. Any advancement in membrane circularity is poised to impact subsequent technologies, thus potentially alleviating the high volume demand for PFSA-based membranes.

### 3.2.4. Impact of the EoL Chemical Processing/Treatment on Membrane Properties.

Chemical treatment influences the chemical and physical properties of the PEM, particularly influencing the ionic transport characteristics, crystallinity, and mechanical strength. Research studies report that the PFSA-based ionomer membrane does not fully dissolve in organic solvents, but rather leads to the formation of dispersion particles, which have a significant effect on various colloidal particles or dispersion aggregates of ionic groups.<sup>92,177</sup>

In order to evaluate the effects of solvent and thermal treatment on membrane recasting, FTIR spectroscopy was used to characterize the chemical change (presence of functional groups) in both recast and commercial membranes. As illustrated in Figure 12, the FTIR results show that the main

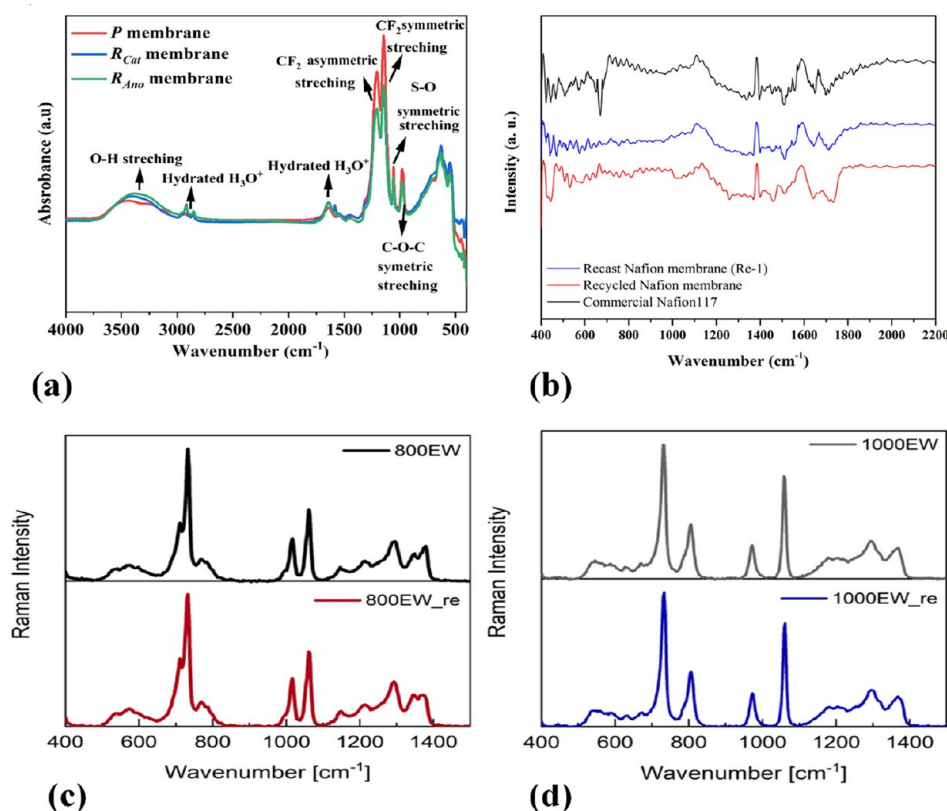


**Figure 12.** FTIR spectra of recycled membrane (a) Results of *p* and *r*-Membrane. Adapted with permission from ref 68. Copyright 2021. Elsevier. (b,c) Recovered and commercial Nafion dispersion. Reproduced with from ref 99. Available under a CC-BY 4.0 license. Copyright 2022 Chen, W. S.; Chen, Y. A.; Lee, C. H.; Chen, Y. J. (d) Recovered and pristine membrane. Reproduced with permission from ref 71. Copyright 2010. Elsevier.

functional groups in the PEM recycled and commercial membrane show no significant changes in the bond vibration before and after recasting. Figure 12a shows the presence of anhydride formation from the dehydration of fuel cells during operation, associated with the peak observed at  $1440\text{ cm}^{-1}$ . The bands observed at  $980$ ,  $1060$ ,  $1100$ , and  $1200\text{ cm}^{-1}$  can be related to the symmetric stretching of C–O–C, S–O, CF<sub>2</sub>, and asymmetric stretching of CF<sub>2</sub>, respectively. Additionally, the presence of peaks at  $3500\text{ cm}^{-1}$  indicates the retention of an important hydrogen-sulfonic bond or the presence of water; Figure 12a,c. However, this could result from the sulfonation of the membrane with the treatment of H<sub>2</sub>SO<sub>4</sub>. However, this predicament is not applicable to all other recast PFSA ionomer membranes, with variations in recovery (dissolution) and recycling (recast) conditions leading to various changes in membrane function groups. Figure 12b shows the FTIR spectra of the membrane recast by different methods. As

shown in Figure 12, the recast membrane showed a comparable chemical composition to the pristine.

Furthermore, absorbance studies and Raman spectroscopy in Figure 13 confirm the FTIR results from previous studies, which showed no significant variations in functional groups. EoL processes of the ionomer have detrimental effects on the ionomer chain. The absorbance of the sulfonyl group in the pristine membrane was observed at  $1060\text{ cm}^{-1}$ ; however, in the recycled membrane, the sulfonyl groups were reduced. Moreover, the peak presence at  $1440\text{ cm}^{-1}$  is attributed to the anhydrides. The intensity of the C–O–C peaks at  $980\text{ cm}^{-1}$  was lower. Figure 13b Nafion 117 membrane. With notable vibrations at  $1100$ – $1200\text{ cm}^{-1}$  attributed to the CF<sub>2</sub>. The peak observation at  $1055$  and  $1228\text{ cm}^{-1}$  were assigned to the symmetry and asymmetry stretching vibrations of SO<sub>3</sub><sup>-</sup>, respectively. Whereas the peak at  $975\text{ cm}^{-1}$  corresponds to C–O–C, the Raman spectra of recycled and commercial membranes in Figure 13a,b show similarities between the



**Figure 13.** FTIR spectra of (a) pristine membrane, anode, and cathode side of recycled membranes. Reproduced with permission from ref 178. Copyright 2023. Elsevier. (b) Nafion 117, recast Nafion membrane (Re-1), and recycled Nafion membrane adapted with from a previous study.<sup>99</sup> Available under a CC-BY 4.0 license. Copyright 2022, by Chen, W. S.; Chen, Y. A.; Lee, C. H.; Chen, Y. J. The Raman spectra of recycled and commercial membrane (c) 800 EW and 800 EW-re, and (d) 1000 EW and 1000 EW-re. Reproduced with permission from ref 135. Copyright 2024. Elsevier.

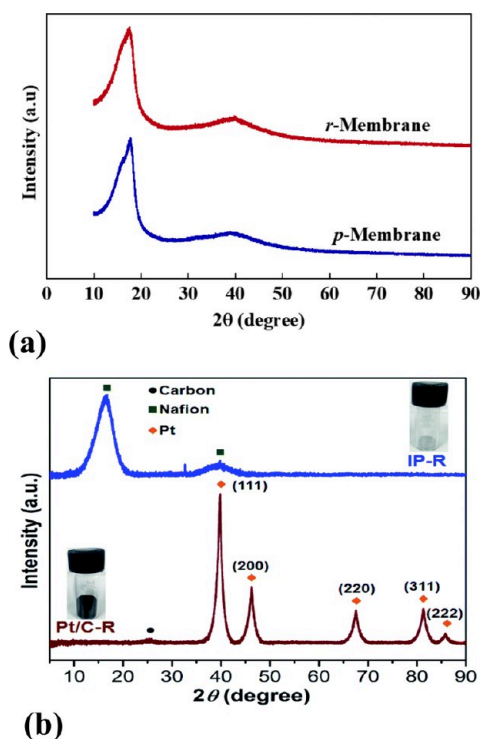
spectra 800 EW-re and 1000 EW-re with their respective pristine membrane. While higher crystallinity of the ionomer indicates higher mechanical strength, is previously stated inverse relationship between chain length and conductivity. Hence, ionomer membrane crystallinity reduces the ion conductivity and mobility properties.

X-ray diffraction (XRD) analysis, as illustrated in Figure 14 shows that the recycled membranes have a higher crystallinity, comparable to that of the commercial membrane. Figure 14a shows that the recycled membrane demonstrated an increase in crystallinity of about 29%, while the pristine membrane only showed 22%. The increased crystallinity can be associated with the S–O–S cross-linked, which signifies that the presence of anhydrides is attributed to enhanced mechanical strength in recycled membranes, affects their water uptake and ion exchange capacity (IEC), while improving durability. Figure 14b reveals the X-ray diffractogram of the recovered membrane via evaporation of solvents from the IES-R solution and Pt/C catalyst (Pt/C-R). Analysis of the results showed peaks attributed to Nafion at 16.6° and 39.9°. Meanwhile, the Pt/C-R exhibits face-centered cubic (fcc) Pt lattice, notable with the following peaks 39.8°, 46.3°, 67.5°, 81.4°, and 85.9° corresponding to the (111), (200), (220), (311), and (222) planes of the fcc Pt lattice. Although recycled membranes may not exhibit exact properties of commercially available membranes, they are reported to retain some properties of interest for potential use in similar technologies. Additionally,

the crystallinity changes on the backbone chain contribute to increased ionic performance of the recast membrane.

Water uptake studies, in Figure 15a, show the effects of different recycling processes on the water uptake of the recast membrane. Furthermore, Figure 15b shows that the pristine membrane exhibits a higher water uptake compared to recycled water. Figure 15b Recycled membrane illustrates reduced water uptake compared to a commercial membrane, mainly from the loss of the sulfonyl groups. Hence, increased crystallinity indicates the loss of sulfonic acid groups. The studies conducted by ref 143, whereby the demonstrated increase in water uptake by recast membranes (Re-1, Re-2, and Re-3) between temperatures 30–80 °C. This observation can be attributed to induced chain movement at higher temperatures, which ultimately accelerates the diffusion of water into the membrane. Figure 15c,d demonstrates the increase of water content within the membrane with increased rate of the relative humidity, with both the 800 EW\_re and 1000 EW\_re showing similar performance to their pristine counterparts.

The membrane chemical properties have a direct effect on the mechanical properties of the membrane, which ultimately influence the stress–strain modes of the membrane, as shown in Figure 17. The stress–strain relationship in Figure 16a shows the analysis of uniaxial tensile loading with a constant strain rate. Meanwhile, Figure 16b shows that the recovered membrane exhibits higher resistance to mechanical strength than the pristine membrane, likely due to alteration of the side chain with chemical processing. The improved mechanical



**Figure 14.** XRD diffractograms for (a) Pristine and recovered membrane *p*- and *r*-Membrane adapted with permission from ref 68. Copyright 2021. Elsevier with deconvoluted peaks of *r* and *p* Membrane, and (b) the recovered membrane via evaporation of solvents forms the IES-R solution and Pt/C catalyst (Pt/C-R) and electrode solution images with pristine and recovered membrane (Inset). Reproduced from ref 149. Available under a CC-BY 3.0 license. Copyright 2020, by Bharti, A.; Natarajan, R.

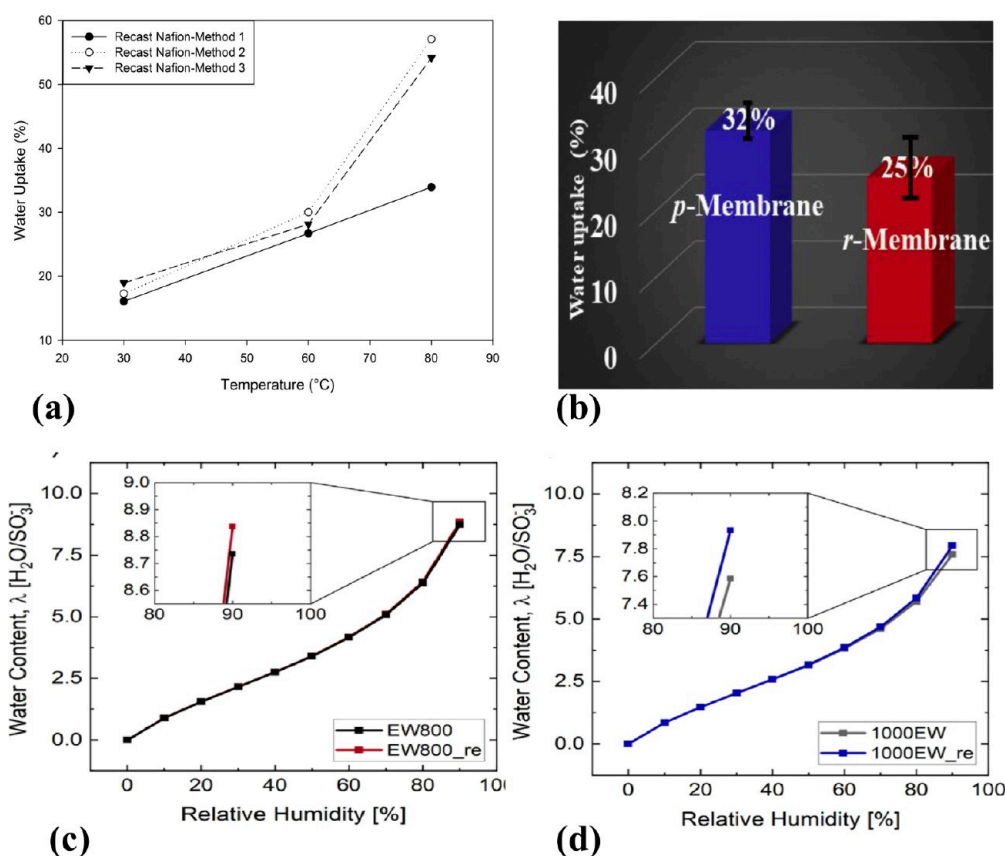
strength is observed for the recycled membrane, due to the presence of anhydride linkage and S–O–S cross-links. Similarly, Figure 16c shows the decreased tensile strength of the recast membrane and the stress of the recast membrane from different solvents and temperatures. Figure 16d illustrates the stress–strain curve for both the recast Nafion membrane and Nafion 117. As shown, the strength of the membranes prepared by methods 2 and 3 (Re-2 and Re-3) demonstrates a significant decrease in mechanical strength compared to the Re-1 membrane and Nafion 117. Furthermore, lower mechanical strength induced by a high degree of water uptake reduces the swelling ratio. While the impact of the solvent on the chemical properties of the membrane is not presented, Figure 16e demonstrates that lower alcohol induces the largest shrinkage on the membrane, mainly due to the lower dielectric constants of the solvents and possible collapse of the hydrophilic domain and water channel. Recast membrane demonstrates various mechanical properties: tensile strength of  $9.8 \pm 0.35$  MPa, Young's modulus,  $165.6 \pm 15.6$  MPa, maximum elongation of  $76.9 \pm 16.2\%$ , and breaking time of 20 h for recast Nafion.<sup>173</sup>

The thermogravimetric analysis (TGA) profiles presented in Figure 17a,b provide essential insights into the thermal stability of Nafion membranes subjected to annealing at 150 °C for a duration of 90 min. A comparative evaluation of membranes stored at 25 °C under 37% relative humidity versus those in distilled water environments reveals three distinct degradation phases: an initial mass loss below 250 °C associated with the evaporation of bound water, a secondary decomposition stage

occurring between 270 and 360 °C linked to the degradation of sulfonic acid side-chains, and a final decomposition of the backbone above 400 °C. Importantly, membranes processed using alcohol–water solvent systems exhibit approximately 15–20% greater water retention capacity, as evidenced by the more pronounced mass loss below 250 °C, compared to those prepared with conventional aprotic solvents. This improved hydration behavior is consistent with the acknowledged proton conductivity benefits of alcohol-processed membranes. Variations in the onset temperatures for side-chain decomposition, ranging from 10 to 15 °C across different solvent systems, indicate that processing parameters significantly impact the thermal resilience of ionic domains. Figure 17c,d illustrates that the decomposition patterns of LSC and SSC closely resemble those of the pristine membrane.

The assessment of weight loss for recast Nafion membranes and the commercial Nafion 117 post-Fenton testing, depicted in Figure 17e, indicates that all recast membranes possess slightly reduced chemical stability relative to their commercial counterpart. Among the recast membranes, Re-2 exhibited the least weight loss, suggesting enhanced resistance to oxidative degradation. This improved stability is likely attributable to the incorporation of NaOH prior to the high-temperature dissolution process. Figure 17f summarizes the percentage of changes in the properties of *r*-Membrane with respect to those of *p*-Membrane. Generally, the recycling process induces changes to the PTFE backbone and the side chain, the percentage changes of properties (chemical and mechanical properties) with 32% crystallinity changes, on other changes of properties (current density, water uptake). These findings highlight the significant interplay between membrane fabrication protocols, water management properties, and thermal stability, which are crucial considerations for the optimization of fuel cell performance under operational conditions. Key considerations are required to make an ionomer for widespread applications; this includes recovering and recycling the membrane while retaining the physical properties of the original membrane. Moreover, an efficient route is established to improve the chemical properties during the recovery face that would be similar to or outperform the commercial membrane.

The Grotthuss mechanism enables fast transport through weakly bonded water, increasing the conductivity with hydration. However, higher crystallinity restricts water-proton mobility, forcing tortuous pathways and reducing conductivity. Figure 18a shows an IEC of 0.57 m equiv/g is observed for the recycled membrane, compared to 0.88 m equiv/g for the pristine membrane, with IEC measured as the rate of sulfonic acid groups protonated. Figure 18b,c shows that the proton permeability is reduced in the recycled membrane; hence, higher crystallinity and lower water conductivity reduce the free volume expansion proton push. The proton conductivity of the recast membrane in Nafion 117 is presented in Figure 18d. Re-2 and Re-3 match Nafion 117's performance, especially at 80 °C. Conductivity rises with temperature due to enhanced proton/chain mobility, influenced by dissolution routes. The proton conductivity conducted between 30 and 80 °C supports the relationship between water uptake and conductivity, whereby the spherical clusters formed within a well-hydrated membrane are demonstrated. Ion conductivities of the membranes. The recycled membrane exhibited half the conductivity of the commercial one ( $6.6 \times$  higher than the



**Figure 15.** Water content results (a) membrane recast with different method adapted with permission from ref 143. Copyright 2018. Elsevier. (b) Pristine and recycled membrane reproduced with permission from ref 68. Copyright 2021. Elsevier. (c,d) Water content in relative humidity. Reproduced with permission from ref 135. Copyright 2024. Elsevier.

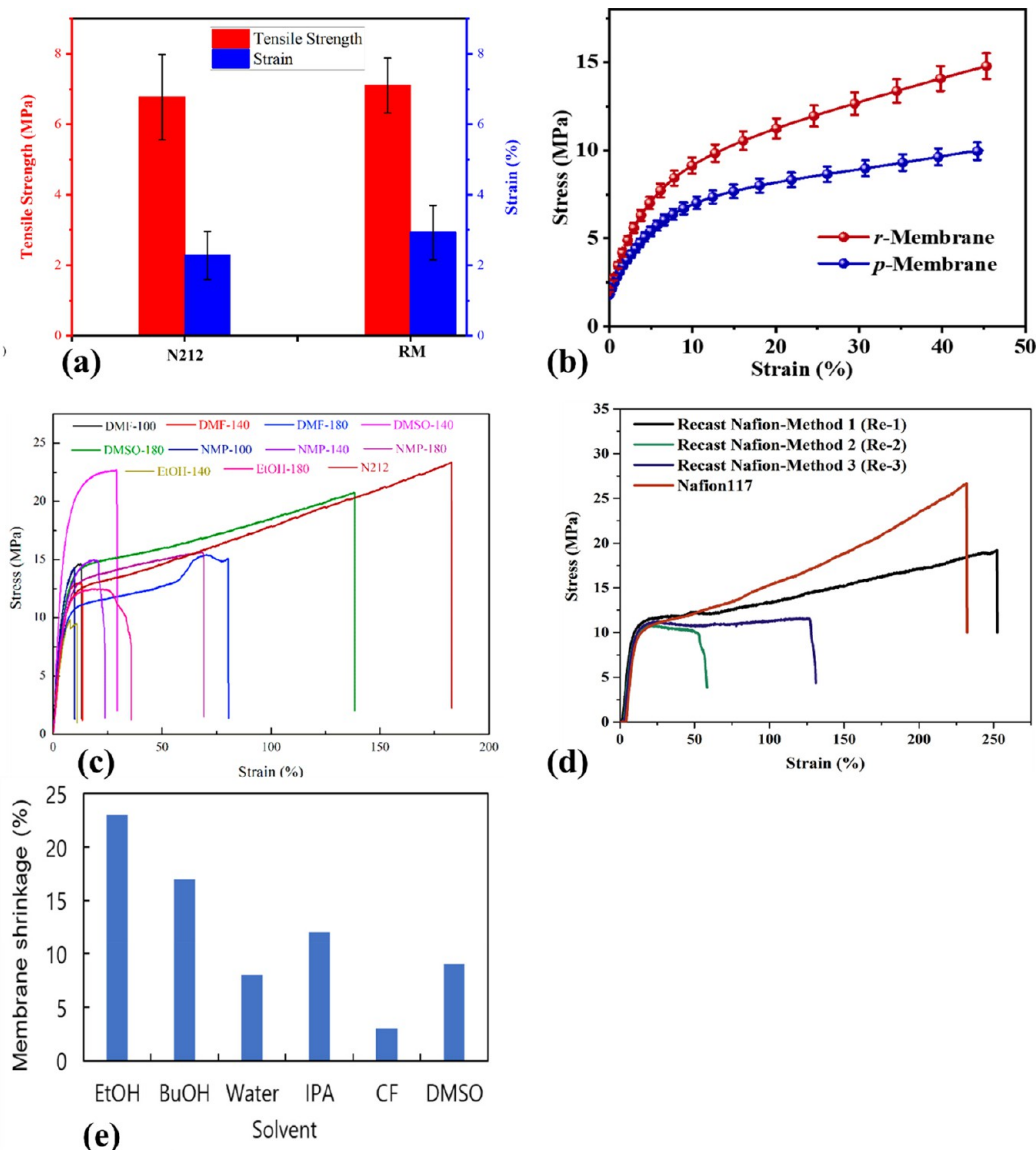
waste membrane), likely due to microstructural changes during vanadium removal and recasting. As shown in Figure 18e.

Figure 19a shows the polarization and power density curve comparison to those of the pristine and reprocessed membrane. Meanwhile, the Nyquist plots of commercial membrane, recycled membrane, and waste membrane are presented in Figure 19b–d, respectively. Contrastingly, it is indicated that the current density of the reprocessed membrane increased, Figure 19e. The performance of a recycled membrane with a pristine catalyst layer was investigated. Carmo et al.<sup>146</sup> demonstrated that the fabrication of CCMs using a recycled membrane. Whereby the CCMs demonstrated higher cell voltage compared to newly prepared CCMs. However, the study does not report on the lifetime of the recycled membrane; rather, it reports on the performance. Fenton test and AST have previously been carried out on the recycled membrane to simulate their chemical and physical stability under simulated conditions. Moreover, a literature review on the industrial application of a recycled ionomer membrane highlights the need for industrial applications of recovered and recycled ionomer membranes.

The morphology studies on recycled membranes indicate that the methodology employed may affect the smooth homogeneous surface of the membrane, leading to inconsistent membrane thicknesses, as illustrated in Figure 20a–c.<sup>143</sup> The recycling method yields membranes with thicknesses ranging between 116–123  $\mu\text{m}$ . Although the membrane thickness impacts the water content, the thickness of the membrane can increase in a hydrated state, thereby also affecting the

membrane's conductivity. The small-angle X-ray scattering (SAXS) analyses for recycled membranes from various studies are presented in Figure 20 d,e.<sup>135,143,181</sup> The spatial arrangement of hydrophobic domains is investigated perpendicular to the plane of the membrane, whereby the recast membrane demonstrates strong scattering, indicating significant ionic aggregations in the ionic hydrophilic domains when in a hydrated state.<sup>181</sup> A summary of the comparison of the performance of the pristine and recycled membranes is presented in Table 5.

It is imperative to determine the molecular weight of the recovered and recycled PFSA ionomers. To date, several methods, including asymmetrical flow field-flow fractionation (AF4), static multiple light scattering (SLS)/ static multiple light scattering (SMLS), and gel permeation chromatography (GPC), have been utilized to determine the molecular weight of polymers.<sup>182–185</sup> GPC is an effective technique for determining the molecular weights of both hydrophilic and hydrophobic oligomers and polymers.<sup>186,187</sup> Similarly, the AF4 method is employed to demonstrate the operation of these techniques, as evidenced by its capability to separate nanoparticles and characterize polymeric materials. However, its application is constrained by the lack of thorough characterization and the small sample size.<sup>188</sup> Moreover, the technique's effectiveness is limited by the sensitivity of the coupled detectors and the low data observed at low concentration samples.<sup>189</sup> Nonetheless, this technique provides direct size characterization in addition to separation capabilities.<sup>190</sup> Since Nafion does not form true solutions,



**Figure 16.** Mechanical properties. (a) Tensile strength and strain of recycled membrane. Reproduced from ref 179. Available under a CC-BY 4.0 license. Copyright 2022, by Xu, Q.; Chen, X.; Wang, S.; Guo, C.; Niu, Y.; Zuo, R.; Yang, Z.; Zhou, Y.; Xu, C. (b) Stress–strain of pristine and recovered membrane Adapted with permission from ref 68. Copyright 2021. Elsevier. (c) Stress–strain of different membrane recast. Adapted with permission from ref 113. Copyright 2017. Elsevier. (d) Stress–strain relationship of recycled membrane. Reproduced with permission.<sup>143</sup> Copyright 2018. Elsevier. (e) Membrane shrinkage in various solvents. Reproduced from ref 180. Awaiting copyright permission.

AF4 remains a technique employed to characterize colloidal particles.<sup>191</sup> SMLS utilizes the principles of photon mean free paths according to Mie theory and the Beer–Lambert law to determine particle size.<sup>184</sup> The SLS can measure the size of Nafion in a transparent solution.<sup>185</sup> The dispersion state of Nafion in its aqueous dispersion properties can be analyzed using SAXS and SLS, as well as dynamic light scattering (DLS).<sup>192</sup>

Further research studies are required to investigate the long-term performance of the recycled ionomer membrane. This includes feasibility studies and environmental and efficiency evaluations. So far, studies have reported on membrane recycling through recast, while this membrane demonstrates mechanical strength, chemical stability, and ionic mobility are affected by the dissolution process. Thus, optimization following established emissions policies is required to establish

a reliable and efficient recycling process for ionomer membranes.

#### 4. ENVIRONMENTAL CONCERNS

The recycling of polymer waste is regarded as a sustainable strategy that has the potential to reduce environmental impact, as well as energy and material demand, in contrast to methods such as incineration or landfill disposal.<sup>193</sup> In order to enhance and facilitate the reuse and recycling of materials, the design of disassembly-friendly materials and devices would significantly improve the process.<sup>89</sup> Although metallurgical processes for the extraction of PGMs electrocatalysts from spent sources are well-established,<sup>194</sup> these processes are not economically viable for the recovery of noble catalysts from PEM-containing electrocatalyst materials due to the incineration of PFSA ionomers, which releases environmentally hazardous fluorocarbons.<sup>145</sup>

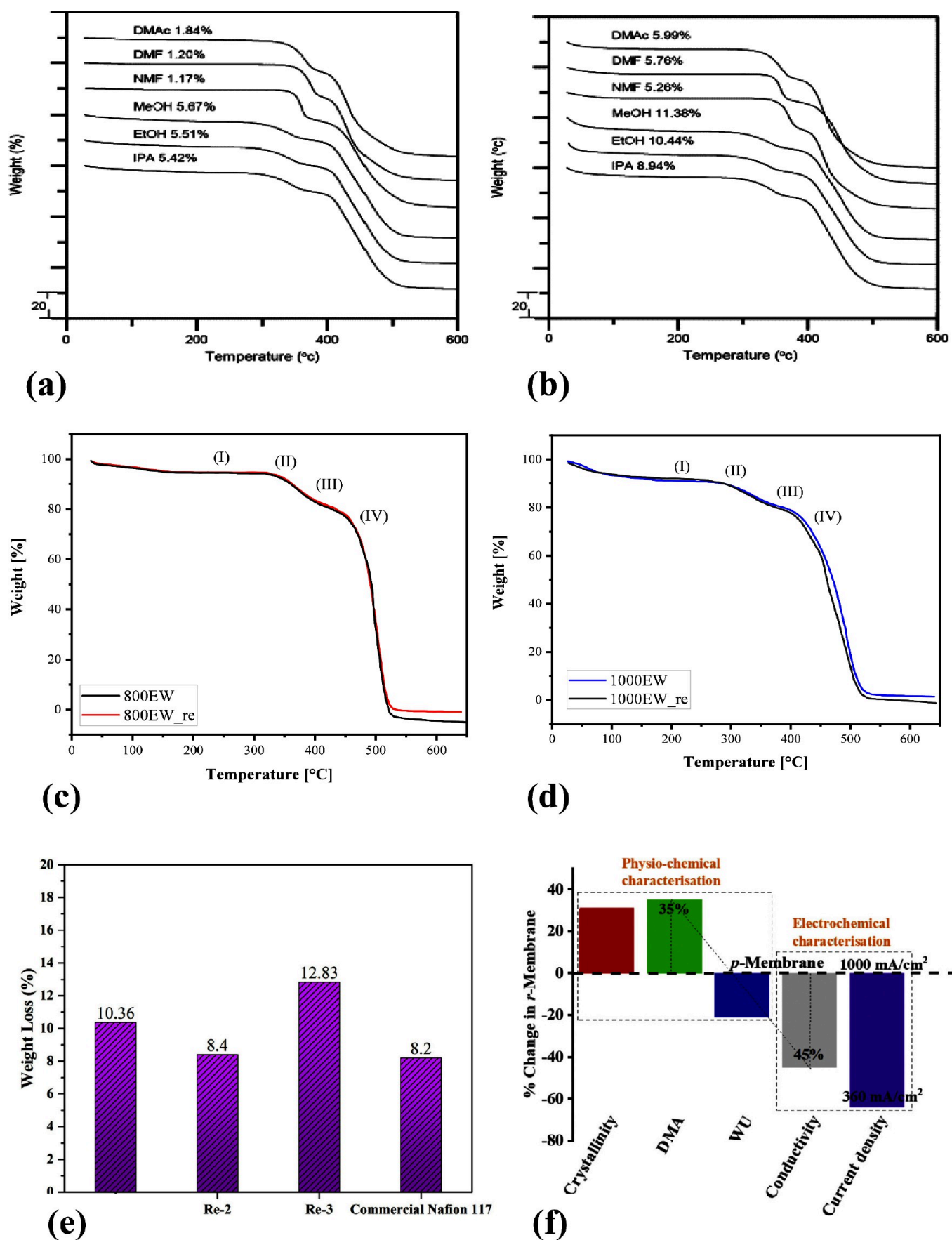
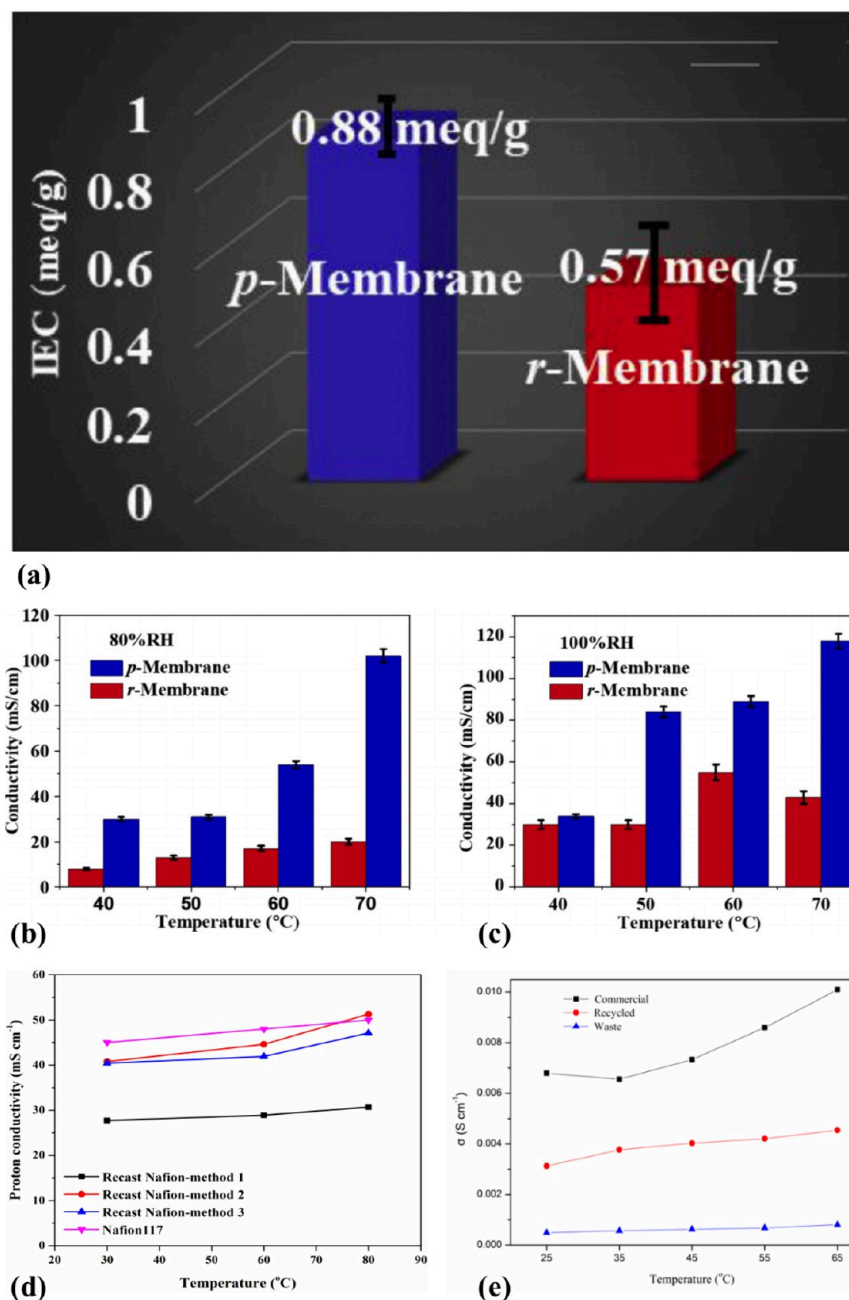


Figure 17. TGA data (a) Nafion annealed at 150 °C for 90 min, (b) Nafion annealed at 150 °C for 90 min in various solvents reproduced with permission from ref 157. Copyright 2009. Elsevier. (c) and (d) show the thermal stability of the pristine and reprocessed membranes tested by

Figure 17. continued

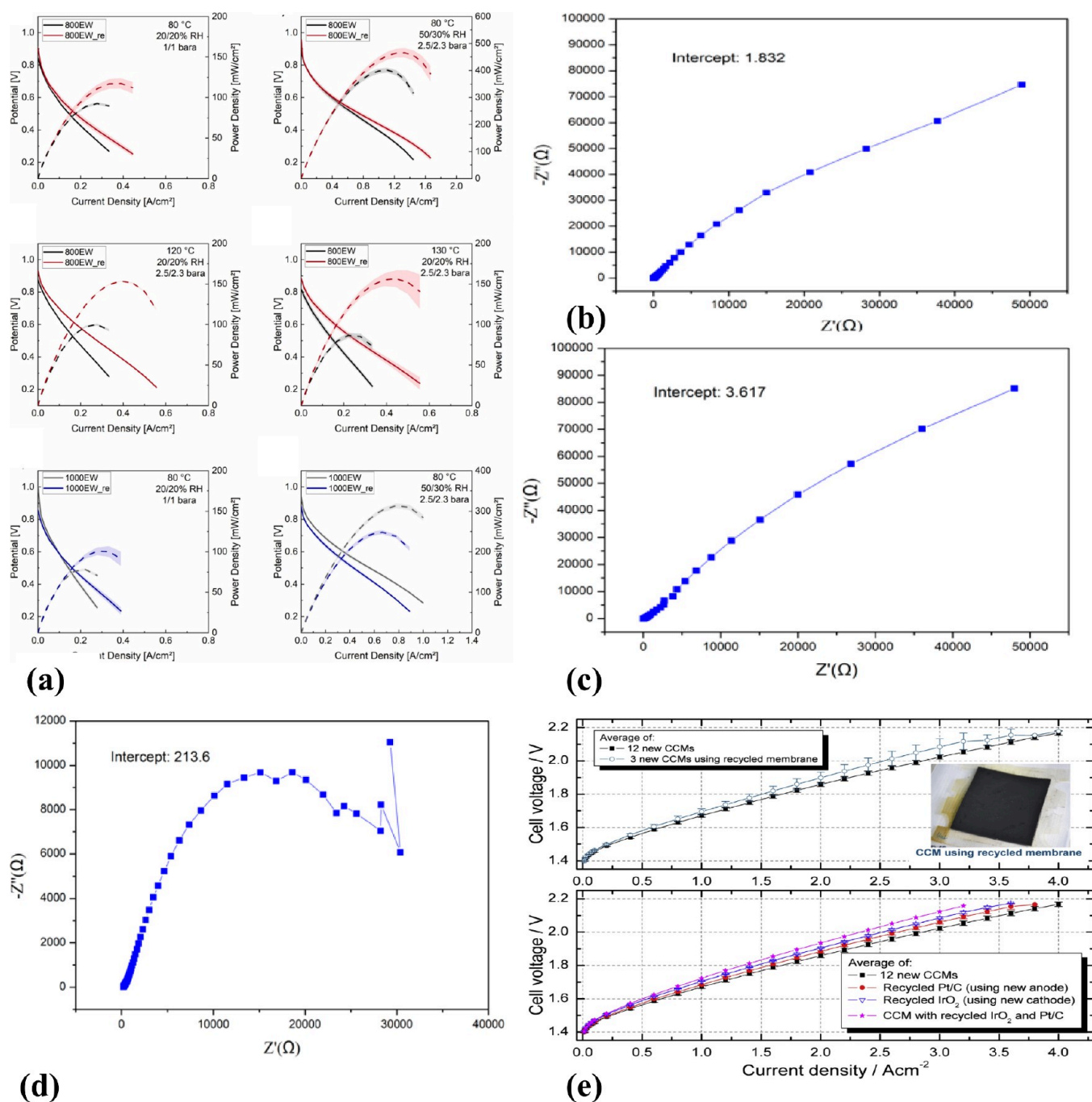
DMA. Reproduced with permission from ref 135. Copyright 2024. Elsevier. (e) Recycled membrane weight loss analysis. Reproduced with permission from ref 143. Copyright 2018. Elsevier. And (f) Crystallinity vs recycled membrane properties. Reproduced with permission from ref 68. Copyright 2018. Elsevier.



**Figure 18.** (a) Ion exchange capacity results of pristine and recycled Membrane, Conductivity measurements of *p* and *r*-membrane at (b) 80% RH and (c) 100%. Adapted with permission from ref 68. Copyright 2021. Elsevier. (d) Conductivities of recast Nafion and Nafion 117 membranes in relation to temperature. Adapted with permission from ref 143. Copyright 2018. Elsevier. (e) Comparison of ion conductivity of commercial, waste, and recycled membrane. Reproduced from ref 99. Available under a CC-BY 4.0 license. Copyright 2022, by Chen, W. S.; Chen, Y. A.; Lee, C. H.; Chen, Y. J.

The importance of inherent primary raw materials and the environmental obligations associated with hydrogen and fuel cell technology motivate the development and adoption of more sustainable EoL processes and handling options in a circular economy. There is an ongoing pursuit for viable alternative EoL processes to enhance the applicability and

implementation of hydrogen technologies and related alternatives, aiming to reduce costs.<sup>146</sup> Consequently, several environmental studies have been conducted on the potential threat posed to both humans and ecosystems by the presence of PFSA/PFAS polymers in the environment. The high mobility of PFAS in water results in their eventual leaching

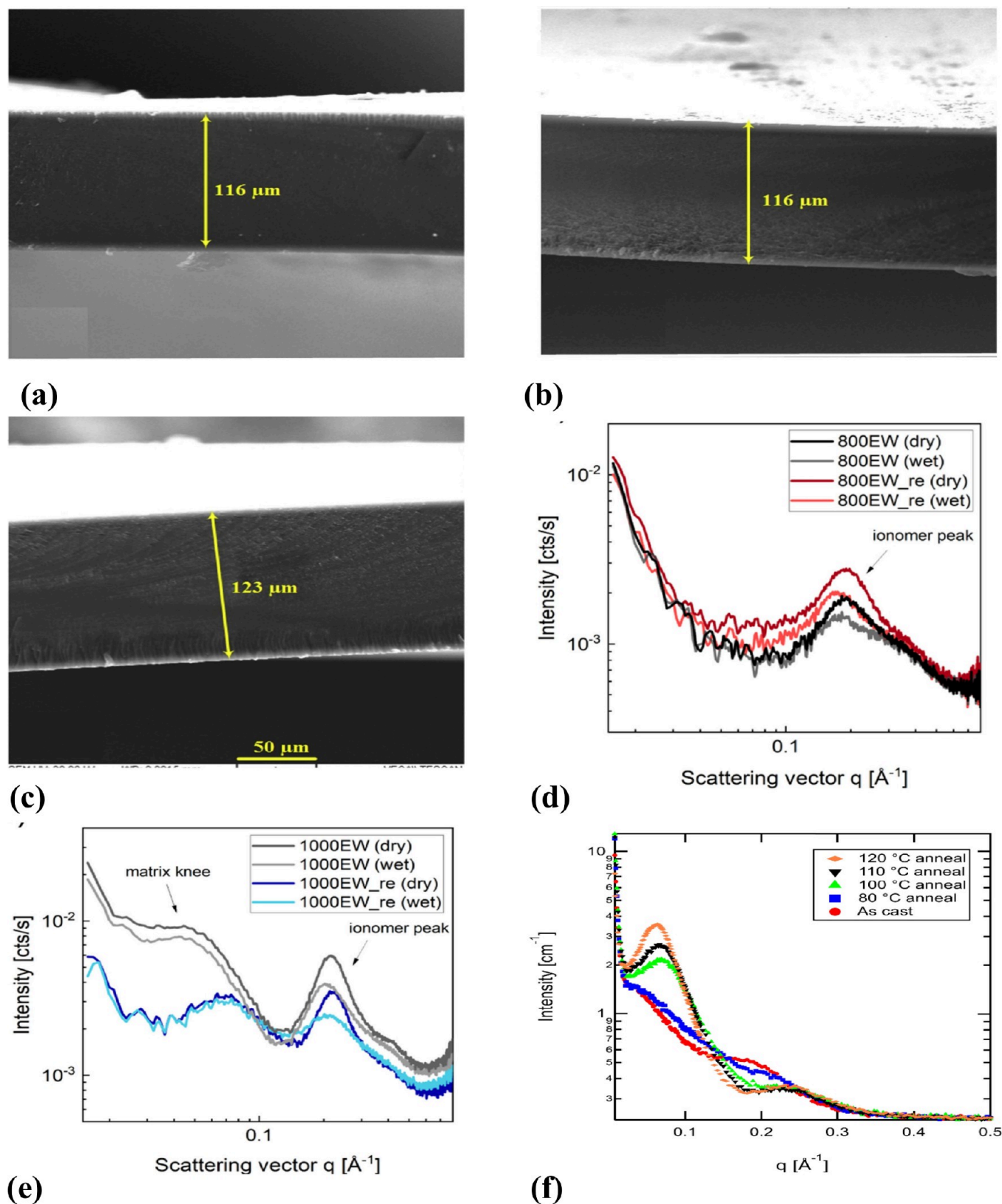


**Figure 19.** (a) Polarization curve and current density of the pristine membrane. Adapted with permission from ref 135. Copyright 2024. Elsevier. Nyquist plots of different membranes: (b) commercial membrane, (c) recycled membrane, and (d) waste membrane. Reproduced from ref 99. Available under a CC-BY 4.0 license. Copyright 2022, by Chen, W. S.; Chen, Y. A.; Lee, C. H.; Chen, Y. J. (e) Cell voltage of CCM prepared with recycled membrane and catalyst. Reproduced with permission from ref 146. Copyrights 2019. Elsevier.

into water systems.<sup>195,196</sup> The stability of fluorinated polymers, such as PFASs, allows them to persist and accumulate in the environment over time.<sup>196</sup> Figure 21 represents the fluoride formation in fuel cells, indicating potential emission into the environment under normal cell conditions.

The disposal of fluorinated materials, particularly polymers like PFASs and perfluorinated membranes, poses a risk of bioaccumulation of fluorine.<sup>195</sup> The emission of fluorine during dissolution poses a risk to the environment, as illustrated in Figure 21. PFSA features suffer drawbacks due to environmental hazards related to the use and recovery of

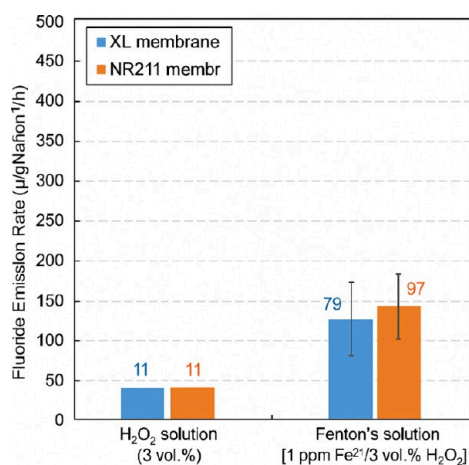
(per)fluorinated compounds.<sup>198</sup> Nafion, another fluorine-based polymer, can present a significant environmental risk upon disposal.<sup>199</sup> Notably, there have been reports of Nafion byproducts being identified in the blood samples of young children participating in studies.<sup>195</sup> The chemical integrity of Nafion renders it a source of pollution, as it tends to persist in the environment when disposed.<sup>196</sup> Efforts to make PEMFCs more attractive have involved the design of PEMs with optimal performance while maintaining environmental sustainability and exploring the potential for recycling and reutilization of the material.<sup>107</sup> There are recognized health and environmental



**Figure 20.** SEM images of the recycled membrane (a–c). Reproduced with permission from ref 143. Copyright 2018. Elsevier. The SAXS data (d) 800EW SSC, and (e) For pristine and recycled 1000EW membrane. Reproduced with permission from ref 135. Copyright 2024. Elsevier. (f) SAXS data for the annealed membrane at different temperatures. Reproduced with permission from ref 181. Copyright 2021. Elsevier.

threats associated with the manufacture of PFASs.<sup>200</sup> It is important to note the significance of separating PFSA ionomer

from electrodes prior to metallurgical processing as a crucial step, due to the economic value of the material.<sup>139</sup>



**Figure 21.** Fluoride emission rate of Nafion XL and NR211 membranes under chemical stress. Reproduced with permission from ref 197. Copyright 2020. Elsevier.

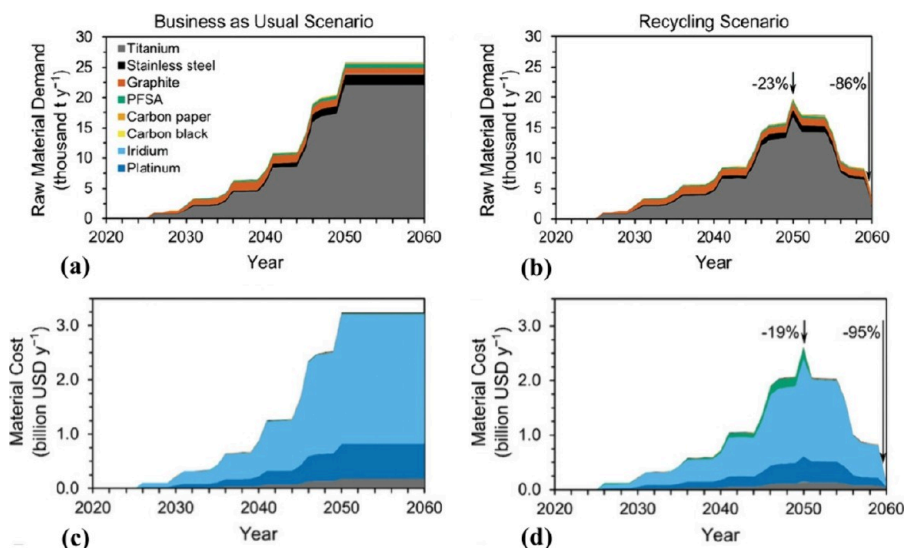
## 5. ECONOMICAL ANALYSIS

The market value of ionomer membranes is anticipated to increase significantly with the expanded application and introduction of PEMWEs and PEMFCs for large-scale applications. The Business-as-Usual Scenario and Recycling Scenario economical implication for FCH technologies data is depicted in Figure 22 highlights an expected rise in the demand for raw materials necessary for the development of FCH technologies from 2020 to 2060, as illustrated in the Business as Usual Scenario in Figure 22a.<sup>73</sup> Furthermore, the recycling scenario in Figure 22b reveals that recycling can reduce the demand for raw materials by at least 23% in the year 2050, and by an additional 86% by 2060. According to the report, the cost of materials for FCH is projected to exceed 3 billion by 2050, which is neither sustainable nor attractive for the implementation of FCHs, as shown in Figure 22c. Currently, the price of Nafion is \$500/m<sup>2</sup>, accounting for 50% in MEAs of the total cost of the fuel cell stack, behind the metal catalyst.<sup>140,200</sup> This projection underscores the significance of designing FCH technologies that are efficient, cost-

effective, and, most importantly, recyclable. It has been reported that optimized recycling methods are crucial, as increased adoption of FCH will increase the cost with the demand for critical materials, including platinum, iridium, and PFSA membranes. If performance and design objectives are achieved, an appropriate recycling process will be complementary to the establishment of FCH technologies as reliable energy sources, as demonstrated in Figure 22d. However, policies and laws concerning emissions are anticipated to play critical roles in establishing a green hydrogen circular economy. Components and materials that are recoverable are technically feasible; the materials are classified as waste streams and are disposed of through incineration or landfilling.<sup>201</sup> Table 6 represents the criticality of the ionomer membrane used in fuel from the LCA. A more compliant transition toward policy-driven incentives is needed to promote compliance and advance efforts toward sustainability.<sup>202</sup>

The simultaneous recycling of both catalysts and ionomer membranes from scrap and waste sources is considered optimal. Despite various recycling efforts being pursued, numerous studies have aimed to identify potential impurities, degradation, molecular conformations, and performance optimizations to enhance reusability. While there are several funded projects, akin to BEST4Hy, targeting a recovery rate of  $\geq 80\%$  of platinum metal and ionomer through hydrometallurgical processes and alcohol dissolution processes, respectively, the recovery project is solely devoted to recovering fuel cell technologies and platinum. In contrast, the project accessible at<sup>203</sup> specifically focuses on the recovery of Ir and ionomer from PEMWE technologies, which can potentially be supported by other EU-funded projects.

Moreover, the objective of the project found at<sup>205</sup> is to minimize the high demand and dependency on critical raw materials, thereby alleviating the carbon footprint, costs, and emissions related to technologies. Johnson Matthey, with extensive expertise in emerging PGM technologies and as a leading institution for recycling both hydrogen fuel cells and electrolyzers to reinforce the hydrogen economy, has reported an 80% reduction in the carbon footprint compared to conventional PGM recycling processes through HyRefineTM



**Figure 22.** Annual FCH technology (a,b), raw material demand, and (c,d) material cost projections. Reproduced from ref 73. Available under a CC-BY 4.0 license. Copyright 2024, by Uekert, T.; Wikoff, H. M.; Badgett, A.

Table 6. List of Electrolytes Commonly Used in PEMFCs<sup>a</sup>

electrolyte	component	methodological assessment			
		material classification	price	material value	material criticality
PFSA	electrolyte	hazardous <sup>201</sup>	medium <sup>70</sup>	medium	medium
S-PEEK		nonhazardous		medium	low
PBI	membrane	hazardous (corrosive)	high <sup>70</sup>	medium	low
PSSA	electrolyte	nonhazardous	low <sup>70</sup>	low	medium

<sup>a</sup>Adapted with permission from ref 204. Copyright 2019. Elsevier.

technology. Additionally, the company's primary goal is to achieve circularity by exclusively focusing on the integrated recycling of PGMs and ionomer, as well as PGM refining for reapplication in new technologies without the use of virgin materials.<sup>206</sup>

Accordingly, Project BEST4Hy,<sup>207</sup> one of the initiatives established by the European Union, has been initiated to explore closed-loop recycling for fuel cell technologies. However, a significant challenge remains to demonstrate the integration of recycled materials (ionomer solution and Pt/C catalyst) into newly produced MEAs for PEMFCs, achieving a performance level of approximately 80% compared with MEAs derived from virgin materials. The project includes collaboration with fuel cell manufacturers such as Elcogen and EKPO Fuel Cell Technologies to incorporate simulations of standard production procedures for closed-loop recycling. Despite the challenges associated with recycled materials, it has been suggested that these materials could be potentially suitable for alternative applications beyond fuel cell technologies. The CEA, under the French Alternative Energies and Atomic Energy Commission, plays a crucial role in the BEST4HY project by contributing to the development, characterization, and evaluation of recycled materials. This involves the development of novel methodologies for fuel cell dismantling and Pt recovery from scaling up at the laboratory scale. Established in 1998 in Aschaffenburg, Germany, Hensel Recycling is recognized as one of the global leaders in PGM recycling. It recovers Pt, Pd, and ruthenium from various EoL sources, including automotive catalytic converters, electronic scrap, oxygen sensors, and fuel cell materials. The company's operations adhere to the German Closed Substance Cycle and Waste Management Act (KrWG) as well as German emissions law (BimSchG). In addition to recycling Pt salts, the company enhances its success through the recovery of platinum ink together with ionomer membranes from fuel cell stacks.<sup>206</sup>

Heraeus precious metals is another entity that has committed efforts to the recycling of critical materials within the circular hydrogen economy. It is among the largest refiners of PGMs globally. Their recycling initiatives encompass components of fuel cell technologies, including BPs, porous transport layers (PTLs), MEAs, and CCMs, as well as defective production materials. The reclamation of end-of-life materials not only bolsters the supply of critical materials but also facilitates the retrieval of valuable substances.<sup>208</sup>

The reported carbon dioxide footprint of recycled PGMs is up to 99% lower compared with that of mined PGMs. In alignment with the Paris Agreement, the Critical Raw Materials Act redefines the European Union's commitment to addressing the risks and challenges associated with demand and dependency on imported materials. This commitment includes efforts to reclaim critical materials, termed strategic materials, and enhance the circularity of these materials, as documented by.<sup>209</sup> In 2023, BASF Environmental Catalyst and Metal

Solutions, a leading producer of materials such as membranes, PGMs, and catalysts for high-temperature PEMFCs, entered into an agreement with Advent Technologies Holdings, Inc. to scale up the production of MEAs and provide recycling services for key materials, including their trademarked Celtec-Z and new Ion Pair membrane technologies. BASF and Advent Technologies have signed an agreement to establish a comprehensive supply chain for hydrogen fuel cell systems in Europe.<sup>210</sup>

Chemours Company,<sup>211</sup> a leading global manufacturer of PFSA ionomers, has committed to achieving net-zero emissions by 2050. The organization has outlined three principal objectives to contribute to its sustainability goals: (i) attain a 60% reduction in CO<sub>2</sub> emissions from manufacturing operations, (ii) achieve net-zero CO<sub>2</sub> operational emissions by 2050, and (iii) align with the global climate goals as articulated in the Paris Agreement's 1.5 °C guidelines. Among the proposed strategies is the management of PFSA membranes, aimed at versatile integration into the hydrogen economy and supporting product development. The company envisions a prospective process for the secondary application of ionomers: (i) Recycling ionomers involves recapturing ionomer products from EoL devices, (ii) The repair process entails restoring damaged ionomer sections through chemical treatment, (iii) Restart refers to the recycling of fluorine by deconstructing ionomers into their constituents, and (iv) Repurposing ionomers involves deploying recaptured ionomers for the development of alternative technologies. These efforts are in line with attempts to achieve sustainability through the management of in-process scrap and defects or early product stages.

Chemours<sup>212</sup> has expressed a commitment to circularity and sustainable development by advocating for recycling and reuse among critical stakeholders and customers. The company is prepared to rapidly reduce waste and environmental impacts through effective management strategies. As part of their initial proposed plan, there is an emphasis on actively integrating product designs and business operations with sustainable circular principles involving stakeholders and customers. Notably, one of the company's significant initiatives is investment in the Dordrecht facility to reduce PFAS emissions by 99% by 2030. However, it is reported that the presence of impurities in postindustrial ionomer membranes from the chlor-alkali industry may necessitate chemical treatments specifically tailored to the Nafion ionomers derived from various applications. A transformative effort involves enhancing the properties of existing membranes to decrease waste generation. The involvement of stakeholders and Nafion production companies in the recycling of ionomer membranes is an essential requirement for ensuring environmental friendliness and fair economic competition.

## 6. CHALLENGES AND PERSPECTIVES

There is an increased research interest in PEMFCs as shown in Figure 3. Burke et al.<sup>213</sup> report that the sale of PEMFC-based vehicles is less competitive than combustion vehicles. This is based on the low volume of production of fuel cell vehicles and high prices. However, it is predicted that the cost of the FC could reach 40\$/kW by 2040.<sup>213</sup> With the extended application of the Nafion in VRFB, fuel cells, and water electrolyzers. It is expected that the amount of the EoL membrane could increase.

The following are some internationally recognized ionomer membrane manufacturers: Chemours, Solvay SA, BASF, AGC Chemicals, Asahi Kasei Corporation, Celanese Corporation, Fumatech BWT GmbH, and Dongyue Group Ltd. The main challenge in the commercialization of PEMFCs is high cost and a lack of EoL strategies.<sup>70</sup> To commercialize PEMFCs, several strides have been taken into consideration to enhance their durability, but several factors such as cost effectiveness, efficiency, and environmental friendliness, are yet to be fully addressed to date. In context, some recommendations and highlights about the commercialization of PEMFCs are mentioned below as follows:

1. Extensive research is necessary to eliminate the environmental threat the EoL ionomer membrane poses, as the application of PEMFCs and other electrochemical devices that use Nafion membrane expands. Recycling strategies of EoL MEAs have a significant role in their commercialization efforts.
2. The state/current conditions of the MEA have a considerable impact on the performance of FC technologies. PGM metal catalysts and PFSA-based ionomer membranes are continuously employed in PEMFCs. However, despite their impressive performance in the electrochemical reactions within the Fuel cell devices, their current cost and low availability are some of their limitations. Therefore, more research efforts are thus required to reduce the cost without compromising the performance of PEMFCs, with composite membranes based on Nafion being some of the leading candidates.
3. Inability to replace defective single components in PEMFCs is considered as the core factor contributing to less distribution and establishment of the devices as into existing power systems.
4. Efforts toward reducing cost and ensuring continuous access to critical materials can be considered as more significant to strengthening the long-term adaptation of PEMFCs. Dissolution methods offer potential solutions to the cost reduction of fuel cell technologies, despite the risk of fluoride emission to the environment. To cope with the current challenges involved in recycling the ionomer membrane, more green methods and less toxic solvents could ultimately recover the Nafion membrane, therefore have application in the circular hydrogen economy.
5. The simulation and modeling studies are critical, transformative processes toward PFSA ionomer membrane recycling. They may contribute to the progress and advancement of recycling by offering optimization, simulation of the effect of solvent, dissolution conditions, and impact of ionomer colloidal properties on the recycled ionomer membrane.

## CONCLUSIONS

In conclusion, the increasing demand for efficient and sustainable energy storage and supply technologies has driven the development of advanced ionomer membranes, electrodes, and metal catalysts. This review provides and summarizes an overview of the present membrane recycling technologies from EoL devices, with key established processes reported. Although limited information about the quantification of the recovery methods is not available. The available literature contains key information about the recyclability of the ionomer membranes in conjunction with other fuel cell components. The recovery methods of the ionomer membranes prove to require more investigation by more environment-friendly and sustainable methods.

Considering the reported results of the dissolution process of the ionomer membrane, there is a future plan for a more efficient method to be established. It is further unknown about the performance of the recycled membrane in devices outside the controlled environment. Recycling critical materials from EoL devices has more ecological benefits than the economy, and it further promotes that fuel cell devices are future energy sources. A challenge yet to be solved is the safety of the recovery process as compared to the manufacturing of this material for energy devices. A better understanding of the recycling process of the membrane can pave the way for green, sustainable operation that will alleviate the potential threat of the disposed material in the environment.

## ASSOCIATED CONTENT

### Data Availability Statement

Available upon request.

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### Notes

The authors declare no competing financial interest.

## Biographies

**Prof. Edson L. Meyer** is a full professor and Director. Research covered aspects of Solar Energy (solar cells, modules and systems), Bio Energy (biomass gasification, biogas digestion), Smart Energy Systems and Sustainable Development Studies (sustainable cities and resources, building integrated photovoltaics for sustainable architecture). He has now established the SolarWatt Park on the Alice campus, where essential research is conducted on various aspects of renewable energy, solar PV and thermal, heat pumps with solar water heaters, and grid-connected PV

**Mr. Bethuel Langa** earned both his BSc and BSc Honours from Sol Plaatje University in 2019 and 2023 respectively. He is currently pursuing an MSc in Chemistry at the University of Fort Hare, focusing on renewable energy and computational studies of photovoltaic devices. His present research investigates sustainable recovery processes of ionomer membranes from used MEAs.

**Dr. Azile Nqombolo** work investigates the ever-increasing global concern and need for sustainable and clean water resources. She is researching functional nanomaterials' synthesis and characterization for water remediation. She also has a keen research interest in renewable energy technologies, solar photovoltaic applications and devices, semiconductor materials, power electronics, and electrical, electrochemical studies and membranes and their recovery from end-of-life Fuel Cells. Her research has been supported by NRF Thuthuka and UFH Seed grants.

**Dr. Mojeed A. Agoro** research has been pivotal in advancing the field of nanostructures, solar cells, renewable energy, inorganic, analytical chemistry, recovery of membranes and PGM in end-of-life Fuel Cells. His work on nanostructured quantum dots led to a significant increase in conversion efficiency in solar cells, achieving a new benchmark of novel efficiency. As a supervisor, he has guided numerous students in their academic pursuits, fostering the next generation of scientists and researchers in chemistry.

**Dr. Nicholas Rono** is currently a Postdoctoral Research Fellow at the Fort Hare Institute of Technology, University of Fort Hare, South Africa. He works on optoelectronic materials for high performance solar cells from computational and experimental perspectives. Dr. Rono graduated with Bachelor of Education (Physics/Chemistry subjects), a Master of Science in Chemistry (Physical and Computational) from Egerton University, Kenya. He holds a PhD in Chemistry degree from the University of KwaZulu-Natal, South Africa.

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