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# A Succinct Review on the PVDF/Imidazolium-Based Ionic Liquid Blends and Composites: Preparations, Properties, and Applications

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**Abstract:** Poly(vinylidene fluoride) (PVDF) is a versatile thermoplastic fluoropolymer with intriguing characteristics, which is receiving considerable attention from researchers in many areas. Recently, PVDF and its copolymer, such as poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) have been blended with ionic liquids to produce blend and composite materials for target applications. In this succinct review, two types of ionic liquids that are utilized for the preparation of PVDF and PVDF-HFP blends and composites, namely, hydrophilic and hydrophobic imidazolium-based ionic liquids, are reviewed. In addition, the effect of the ionic liquids on the physicochemical properties of the PVDF and PVDF-HFP blends and composites, is described as well. On top of that, a multitude of applications of the blends and composites are also succinctly reviewed. This review may give inspirations to the polymer blend and composite researchers in diversifying the applications of thermoplastic fluoropolymers through the utilization of ionic liquids.

**Keywords:** poly(vinylidene fluoride); poly(vinylidene fluoride-co-hexafluoropropylene); ionic liquid; blend; composite

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## 1. Introduction

These days, polymers can be tailor-made for specific applications, it can be done by chemically modifying the polymer functional groups and side chains [1]. On the other hand, some polymers can physically be modified by utilizing ionic compounds, such as ionic liquids for target applications. Lately, thermoplastic fluoropolymer, for example, poly(vinylidene fluoride) (PVDF) is blended with ionic liquids to produce blend and composite materials for a multitude of applications. PVDF is made from vinylidene fluoride monomer through a free-radical polymerization process. Figure 1 shows the schematic of the polymerization of vinylidene fluoride to make PVDF. In addition, PVDF copolymer, such as poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) is also blended with ionic liquids to produce blends and composites for a wide variety of applications. PVDF-HFP is made via copolymerization process by means of two different monomers [2]. Figure 2 indicates the schematic of the copolymerization of vinylidene fluoride with hexafluoropropylene to make PVDF-HFP. The significant advantages of PVDF and PVDF-HFP are their solubility in organic solvents and their capability to use in electronic and electric applications.

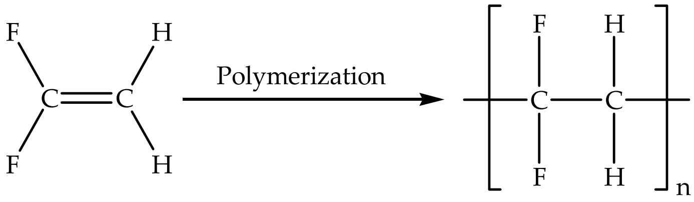

**Figure 1.** Schematic of the polymerization of vinylidene fluoride to make PVDF.

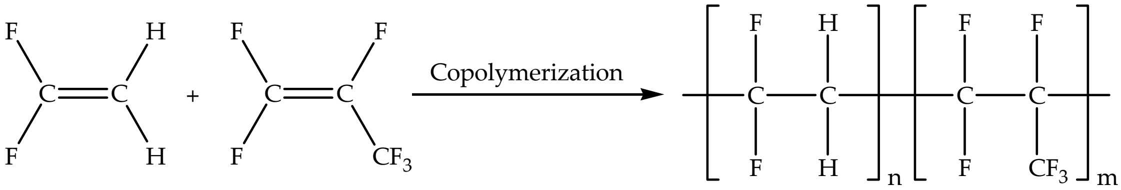

**Figure 2.** Schematic of the copolymerization of vinylidene fluoride with hexafluoropropylene to make PVDF-HFP.

Ionic liquids are ionic compounds that have liquid property below 100 °C. They are non-volatile since they have a low vapor pressure. Ionic liquids are also regarded as an environmentally benign solvent because they can be recycled [3]. Moreover, ionic liquids have interesting solvent properties, for instance, good solubility with countless organic solvents, high thermal stability, good electrical conductivity, high polarity, and non-flammability [4]. They also possess the aptitude to dissolve most organic materials, including biopolymers and some inorganic materials. In addition, ionic liquids can be created according to the necessities of the usage [5]. The utilization of ionic liquids on the PVDF and PVDF-HFP is a practical approach for the preparation of blends and composites with multiple functions. In numerous types of ionic liquids, imidazolium-based ionic liquids are frequently utilized for that purpose. This is because of the efficiency and unique properties of the ionic liquids [6]. Nonetheless, many imidazolium-based ionic liquids have similar cations but distinct counter anions that commonly possess different hydrophobicity. Therefore, in this succinct review, they have been classified into two types, specifically hydrophilic and hydrophobic imidazolium-based ionic liquids.

Table 1 demonstrates examples of hydrophilic imidazolium-based ionic liquids utilized for preparation of PVDF and PVDF-HFP blends and composites. It can be observed that the hydrophilic ionic liquids with halide counter anions ($Cl^-$, $Br^-$, and $I^-$) have repeatedly been utilized, followed by tetrafluoroborate counter anion ($BF_4^-$). This is probably because of their easier availability and cost-effectiveness compared to other hydrophilic ionic liquids. On the other hand, $OAc^-$, $DCA^-$, $H_2PO_4^-$, $EtSO_4^-$, $NO_3^-$, $SCN^-$, and $CF_3SO_3^-$ counter anions are rarely utilized, but they can play a potential role in the blends and composites. Figure 3 exhibits the chemical structures of hydrophilic imidazolium-based ionic liquids collected in Table 1. Meanwhile, Table 2 exhibits examples of hydrophobic imidazolium-based ionic liquids utilized for preparation of PVDF and PVDF-HFP blends and composites. It is seen that the counter anions, such as $NTf_2^-$, $TFSA^-$, $FSI^-$, $FAP^-$, $PF6^-$, and $B(CN)_4^-$ contribute to the hydrophobic property of the ionic liquids regardless of the type of the imidazolium cations. However, $BF_4^-$ counter anion can exhibit both hydrophilic and hydrophobic properties. This phenomenon is influenced by the length of the cation alkyl chain [6]. That is why [Hmim][$BF_4$] is hydrophobic, whereas [Emim][$BF_4$], [Bmim][$BF_4$], and [Veim][$BF_4$] are hydrophilic. Figure 4 demonstrates the chemical structures of hydrophobic imidazolium-based ionic liquids collected in Table 2.

**Table 1.** Examples of hydrophilic imidazolium-based ionic liquids utilized for preparation of PVDF and PVDF-HFP blends and composites.

| Hydrophilic Imidazolium-Based Ionic Liquid | Abbreviation | References |
|---|---|---|
| 1-Methyl-3-carboxymethylimidazolium chloride | [Mcmim][Cl] | [7] |
| 1-Ethyl-3-methylimidazolium chloride | [Emim][Cl] | [8,9] |
| 1-Butyl-3-methylimidazolium chloride | [Bmim][Cl] | [10] |
| 1-Hexyl-3-methylimidazolium chloride | [Hmim][Cl] | [8,9,11] |
| 1-Decyl-3-methylimidazolium chloride | [Dmim][Cl] | [8,9] |
| 1-Vinyl-3-butylimidazolium chloride | [Vbim][Cl] | [12,13] |
| 1-(2-Amino-ethyl)-3-methylimidazolium bromide | [Aemim][Br] | [14] |
| 1-Butyl-3-methylimidazolium bromide | [Bmim][Br] | [15,16] |
| 1-Hexadecyl-3-methylimidazolium bromide | [Hdmim][Br] | [17] |
| 3-Aminoethylimidazolium bromide | [Aeim][Br] | [18] |
| 1,3-Dimethylimidazolium iodide | [Mmim][I] | [19] |
| 1-Propyl-3-methylimidazolium iodide | [Pmim][I] | [19] |
| 1-Butyl-3-methylimidazolium iodide | [Bmim][I] | [19,20] |
| 1-Hexyl-3-methylimidazolium iodide | [Hmim][I] | [19] |
| 1-Ethyl-3-methylimidazolium acetate | [Emim][OAc] | [21] |
| 1-Ethyl-3-methylimidazolium dicyanamide | [Emim][DCA] | [22] |
| 1-Butyl-3-methylimidazolium dihydrogenphosphate | [Bmim][$H_2PO_4$] | [23] |
| 1-Ethyl-3-methylimidazolium ethylsulfate | [Emim][$EtSO_4$] | [21] |
| 1-Ethyl-3-methylimidazolium nitrate | [Emim][$NO_3$] | [24,25] |
| 1-Ethyl-3-methylimidazolium thiocyanate | [Emim][SCN] | [26] |
| 1-Ethyl-3-methylimidazolium tetrafluoroborate | [Emim][$BF_4$] | [27–32] |
| 1-Butyl-3-methylimidazolium tetrafluoroborate | [Bmim][$BF_4$] | [29,33–37] |
| 1-Vinyl-3-ethylimidazolium tetrafluoroborate | [Veim][$BF_4$] | [38,39] |
| 1-Ethyl-3-methylimidazolium trifluoromethanesulfonate | [Emim][$CF_3SO_3$] | [40] |
| 1-Butyl-3-methylimidazolium trifluoromethanesulfonate | [Bmim][$CF_3SO_3$] | [27] |

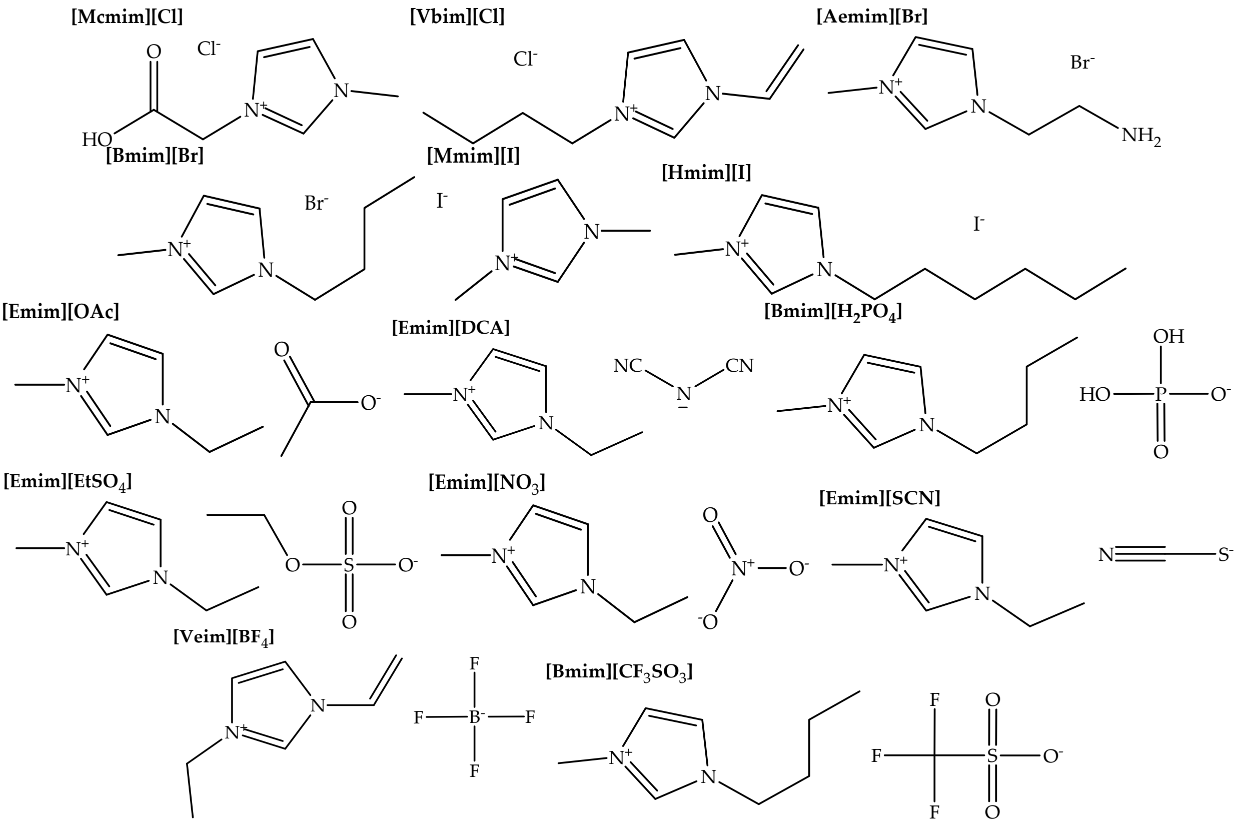

**Figure 3.** Chemical structures of hydrophilic imidazolium-based ionic liquids and the abbreviations are explained in Table 1.

**Table 2.** Examples of hydrophobic imidazolium-based ionic liquids utilized for preparation of PVDF and PVDF-HFP blends and composites.

| Hydrophobic Imidazolium-Based Ionic Liquid | Abbreviation | References |
|---|---|---|
| 1-Ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide | [Emim][NTf$_2$] | [8,9,27,41–48] |
| 1-Ethyl-3-dimethylimidazolium bis(trifluoromethylsulfonyl)imide | [Emmim][NTf$_2$] | [41] |
| 1-Propyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide | [Pmim][NTf$_2$] | [41] |
| 1-Butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide | [Bmim][NTf$_2$] | [49] |
| 1-Hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide | [Hmim][NTf$_2$] | [8,9,11] |
| 1-Decyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide | [Dmim][NTf$_2$] | [8,9] |
| 1,2-Dimethyl-3-propylimidazolium bis(trifluoromethylsulfonyl)imide | [Mmpim][NTf$_2$] | [50] |
| 1-Ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)amide | [Emim][TFSA] | [51] |
| 1-Ethyl-3-methylimidazolium bis(fluorosulfonyl)imide | [Emim][FSI] | [52] |
| 1-Ethyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate | [Emim][FAP] | [53] |
| 1-Hexyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate | [Hmim][FAP] | [27] |
| 1-Butyl-3-methylimidazolium hexafluorophosphate | [Bmim][PF$_6$] | [23,49,54–58] |
| 1-Benzyl-3-methylimidazolium hexafluorophosphate | [Bzmim][PF$_6$] | [59] |
| 1-Ethyl-3-methylimidazolium tetracyanoborate | [Emim][B(CN)$_4$] | [60,61] |
| 1-Hexyl-3-methylimidazolium tetrafluoroborate | [Hmim][BF$_4$] | [29] |

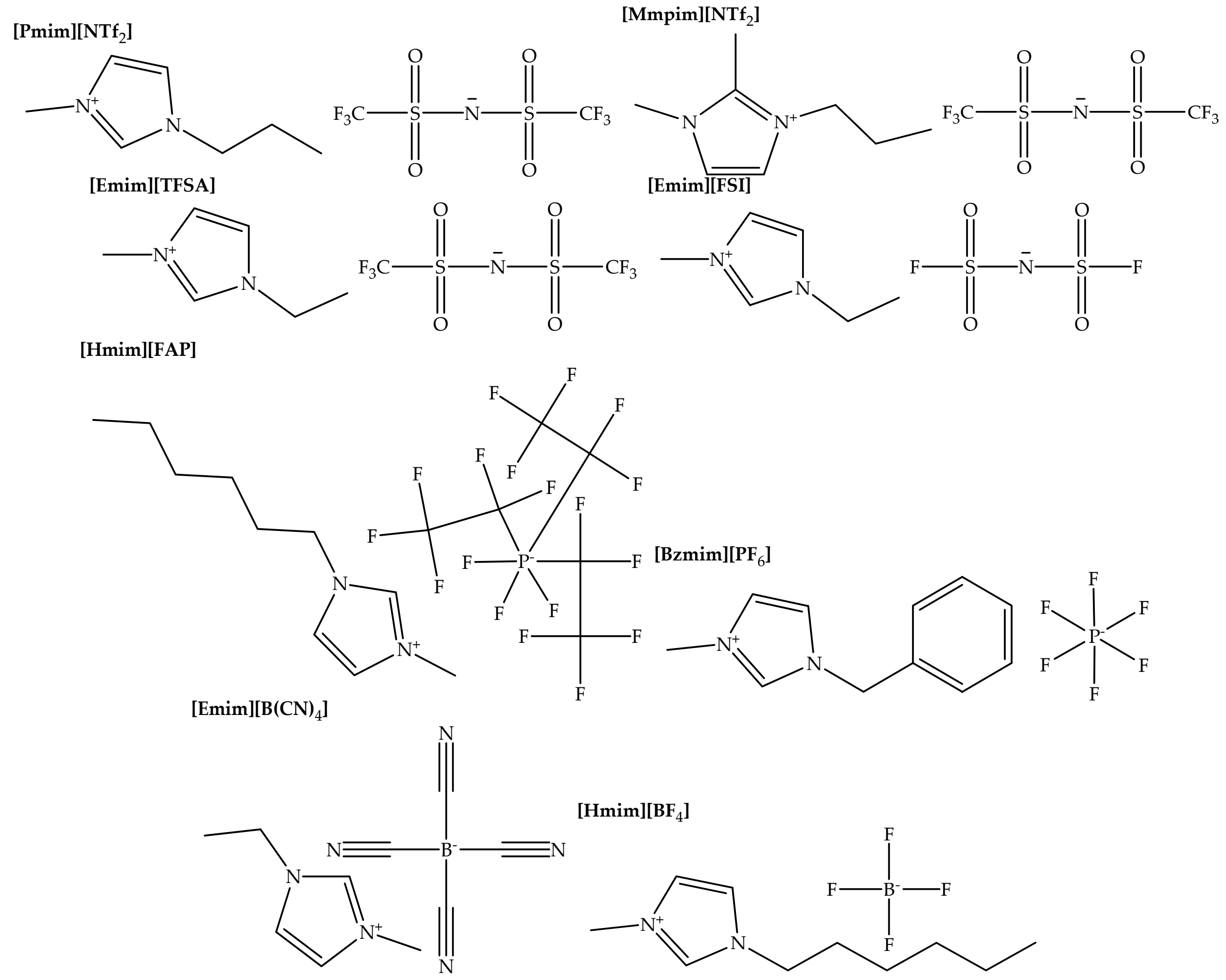

**Figure 4.** Chemical structures of hydrophobic imidazolium-based ionic liquids and the abbreviations are explained in Table 2.

Table 3 displays examples of fillers modified with imidazolium-based ionic liquids for preparation of PVDF and PVDF-HFP composites. It can be perceived that the allotropes of carbon, such as graphene, graphene oxide, and carbon nanotubes can be modified with the ionic liquids [17,32,39]. Clay mineral like montmorillonite can not only be modified with surfactants [62,63] but also with the ionic liquids [10,15]. Additionally, metal oxides, for example, titanium dioxide and silicon dioxide have been modified with the ionic liquids [36,52]. On top of that, organic materials, for instance, polyvinyl pyrrolidone grains and succinonitrile have also been modified with the ionic liquids [31,37]. The utilization of the ionic liquids as modifiers can provide an advantage because of their exceptional chemical structures that are able to interact with PVDF and PVDF-HFP, as well as inorganic and organic fillers. This consequently improves the interfacial bonding between the polymer matrix and filler. In addition, to the best knowledge of the authors, no succinct review has been conceived encompassing the study on the preparations, properties, and applications of PVDF/and PVDF-HFP/imidazolium-based ionic liquid blends and composites. That is the rationale of conceiving an organized review in this paper.

**Table 3.** Examples of fillers modified with imidazolium-based ionic liquids for preparation of PVDF and PVDF-HFP composites.

| Filler | Abbreviation | References |
|---|---|---|
| Graphene | Gr | [17,58] |
| Graphene oxide | GO | [32,38] |
| Carboxylated graphene oxide | GO-COOH | [14] |
| Multi-walled carbon nanotubes | MWCNTs | [20,39,48,57] |
| Carboxylated multi-walled carbon nanotubes | MWCNT-COOH | [18,56] |
| Sodium montmorillonite | NaMMT | [10] |
| Octadecylamine modified montmorillonite | OMMT | [15,16] |
| Silicon dioxide | $SiO_2$ | [52] |
| Titanium dioxide | $TiO_2$ | [7,36] |
| Polyvinyl pyrrolidone grains | PVP grains | [31] |
| Succinonitrile | SN | [37,40] |

## 2. Preparations of PVDF and PVDF-HFP Blends and Composites

### *2.1. PVDF/and PVDF-HFP/Hydrophilic Imidazolium-Based Ionic Liquid Blends*

Table 4 shows the types of polymer matrices, hydrophilic imidazolium-based ionic liquids, and preparation processes of PVDF and PVDF-HFP blends. It can be observed that the PVDF is more often used as a polymer matrix in comparison to PVDF-HFP. This is possibly due to PVDF-HFP lack of attention in the preparation of polymer/ionic liquid blends. On top of that, the blending process between the ionic liquids and PVDF can be conducted via solution blending in polar aprotic solvents, such as dimethylformamide [8,9], dimethylacetamide [23], and 1-methyl-2-pyrrolidone [27]. In addition, PVDF can also be blended with the ionic liquids through solventless ways, namely liquid impregnation and melt blending. Liquid impregnation can be done by impregnating PVDF in an ionic liquid at room temperature for 48 h [21], whilst melt blending can be carried out at 190 °C for 0.12 h [12,13]. Thus, the fastest way to blend between the ionic liquids and PVDF is through melt blending, but it requires a specific blending machine and a hot-pressing machine to shape the blends. Meanwhile, as for PVDF-HFP, it can be blended with the ionic liquids via soaking in an ionic liquid at room temperature for 24 h [19], and solution blending in low boiling point polar aprotic solvent like acetone [34,35].

**Table 4.** Types of polymer matrices, hydrophilic imidazolium-based ionic liquids, and preparation processes of PVDF and PVDF-HFP blends.

| Polymer Matrix | Hydrophilic Ionic Liquid | Blending Process | Blending Temp. (°C) | Time (Hour) | Final Process | Final Temp. (°C) | Time (Hour) | References |
|---|---|---|---|---|---|---|---|---|
| PVDF | [Emim][Cl] | SB | R | 3 | SC | 210 | 0.17 | [8,9] |
| PVDF | [Hmim][Cl] | SB | R | 3 | SC | 200 | 0.17 | [8,9,11] |
| PVDF | [Dmim][Cl] | SB | R | 3 | SC | 210 | 0.17 | [8,9] |
| PVDF | [Vbim][Cl] | MB | 190 | 0.12 | HP | 210 | 0.17 | [12,13] |
| PVDF | [Emim][OAc] | LI | R | 48 | U | U | U | [21] |
| PVDF | [Emim][EtSO$_4$] | LI | R | 48 | U | U | U | [21] |
| PVDF | [Emim][NO$_3$] | SB | R | U | S-C | 100 | 24 | [24,25] |
| PVDF | [Bmim][H$_2$PO$_4$] | SB | 50 | 6 | SC | 120 | 24 | [23] |
| PVDF | [Bmim][CF$_3$SO$_3$] | SB | 80 | U | SC | 25 | 24 | [27] |
| PVDF | [Emim][BF$_4$] | SB | R | U | MG | R | U | [27–30] |
| PVDF | [Bmim][BF$_4$] | SB | 60 | U | SC | 60 | U | [29,33] |
| PVDF-HFP | [Mmim][I] | S | R | 24 | U | U | U | [19] |
| PVDF-HFP | [Pmim][I] | S | R | 24 | U | U | U | [19] |
| PVDF-HFP | [Bmim][I] | S | R | 24 | U | U | U | [19] |
| PVDF-HFP | [Hmim][I] | S | R | 24 | U | U | U | [19] |
| PVDF-HFP | [Emim][DCA] | SB | 60 | U | SC | R | U | [22] |
| PVDF-HFP | [Emim][SCN] | SB | R | 12 | SC | R | U | [26] |
| PVDF-HFP | [Bmim][BF$_4$] | SB | 50 | 6 | SC | R | U | [34,35] |

SB = solution blending, MB = melt blending, LI = liquid impregnation, S = soaking, SC = solution casting, HP = hot pressing, S-C = spin-coating, MG = microfluidic generation, R = room, and U = unstated.

### 2.2. PVDF/and PVDF-HFP/Hydrophobic Imidazolium-Based Ionic Liquid Blends

Table 5 indicates the types of polymer matrices, hydrophobic imidazolium-based ionic liquids, and preparation processes of PVDF and PVDF-HFP blends. It is seen that the PVDF is also preferred for the preparation of polymer/ionic liquid blends than the PVDF-HFP. In addition, the solution blending is mainly done as well in preparation of the blends. However, liquid impregnation can be performed in a hydrophobic ionic liquid at room temperature for 0.5 h only [49], which has less impregnation time compared to the hydrophilic ionic liquid. On the other hand, the solution blending is typically conducted at room temperature regardless of the type of the imidazolium-based ionic liquids. However, elevated temperatures are also used for shortening the blending process. Lastly, the final process is essentially important for molding the blends prior to physicochemical characterization and actual application. The final process for the solution blending can be carried out through solution casting by pouring the solution on leveled glass plates and evaporating solvent at high or room temperature for certain or 24 h. The process has normally been applied for the production of the free-standing film [64]. Nevertheless, hot pressing can also be done to obtain the desired shape of the blends [59]. Additionally, the spin-coating can be operated as well after the solution blending [51].

### 2.3. PVDF/and PVDF-HFP/Ionic Liquid-Modified Filler Composites

Table 6 exhibits the types of polymer matrices, fillers, imidazolium-based ionic liquids, and preparation processes of PVDF and PVDF-HFP composites. Most of the fillers have been modified with the ionic liquids before the mixing process with the polymer matrices. In addition, the modification of the fillers with the imidazolium-based ionic liquids can be carried out through ultrasonication [20], stirring [7,10,14,17,18], and grinding [38,39,48,56–58] processes. The mixing process between the polymer matrices and modified fillers can be conducted via melt blending, solution blending, ultrasonication, and sonication. PVDF and the modified fillers can be processed via melt blending at a temperature range from 190 to 220 °C for less than 0.34 h. On top of that, solution blending between the polymer matrices and modified fillers can be done in polar aprotic solvents at room or elevated temperatures. Meanwhile, for ultrasonication [14,15,17] and sonication [18,36,52], they are usually performed in the solvents at room temperature, but require less time compared to the solution blending. The final processes, such as hot pressing, solution casting, compression molding, and soaking have also been employed to obtain characterizable and appliable PVDF and PVDF-HFP composites.

**Table 5.** Types of polymer matrices, hydrophobic imidazolium-based ionic liquids, and preparation processes of PVDF and PVDF-HFP blends.

| Polymer Matrix | Hydrophobic Ionic Liquid | Blending Process | Blending Temp. (°C) | Time (Hour) | Final Process | Final Temp. (°C) | Time (Hour) | References |
|---|---|---|---|---|---|---|---|---|
| PVDF | [Emim][NTf$_2$] | SB | R | 3 | SC | 210 | 0.17 | [8,9,27,41–44] |
| PVDF | [Emmim][NTf$_2$] | SB | R | U | SC | 210 | 0.17 | [41] |
| PVDF | [Pmim][NTf$_2$] | SB | R | U | SC | 210 | 0.17 | [41] |
| PVDF | [Bmim][NTf$_2$] | LI | R | 0.5 | U | U | U | [49] |
| PVDF | [Hmim][NTf$_2$] | SB | R | 3 | SC | 210 | 0.17 | [8,9,11] |
| PVDF | [Dmim][NTf$_2$] | SB | R | 3 | SC | 210 | 0.17 | [8,9] |
| PVDF | [Emim][B(CN)$_4$] | SB | R | 2 | SC | 50 | 24 | [60] |
| PVDF | [Bmim][PF$_6$] | SB | 50 | 6 | SC | 120 | 24 | [23,49,54,55] |
| PVDF | [Bzmim][PF$_6$] | SB | R | U | HP | 175 | U | [59] |
| PVDF | [Hmim][FAP] | SB | 80 | U | SC | 25 | 24 | [27] |
| PVDF-HFP | [Emim][NTf$_2$] | SB | 50 | 12 | SC | R | U | [45–47] |
| PVDF-HFP | [Mmpim][NTf$_2$] | SB | R | 4 | SC | R | U | [50] |
| PVDF-HFP | [Emim][TFSA] | SB | R | U | S-C | R | 0.02 | [51] |
| PVDF-HFP | [Emim][FAP] | SB | R | 12 | SC | R | U | [53] |
| PVDF-HFP | [Emim][B(CN)$_4$] | SB | R | 12 | SC | R | U | [61] |

SB = solution blending, LI = liquid impregnation, SC = solution casting, HP = hot pressing, S-C = spin-coating, R = room, and U = unstated.

**Table 6.** Types of polymer matrices, fillers, imidazolium-based ionic liquids, and preparation processes of PVDF and PVDF-HFP composites.

| Polymer Matrix | Filler | Ionic Liquid | Mixing Process | Mixing Temp. (°C) | Time (Hour) | Final Process | Final Temp. (°C) | Time (Hour) | References |
|---|---|---|---|---|---|---|---|---|---|
| PVDF | Gr | [Hdmim][Br] | Ul | R | 0.17 | HP | 190 | U | [17] |
| PVDF | Gr | [Bmim][PF$_6$] | MB | 200 | U | U | U | U | [58] |
| PVDF | GO | [Veim][BF$_4$] | SB | R | U | HP | 200 | 0.05 | [38] |
| PVDF | GO-COOH | [Aemim][Br] | Ul | R | U | SC | 60 | 48 | [14] |
| PVDF | MWCNT-COOH | [Aeim][Br] | So | R | 0.17 | SC | 70 | 72 | [18] |
| PVDF | MWCNTs | [Veim][BF$_4$] | MB | 190 | U | HP | 190 | U | [39] |
| PVDF | MWCNTs | [Emim][NTf$_2$] | MB | 220 | 0.33 | CM | U | U | [48] |
| PVDF | MWCNTs | [Bmim][PF$_6$] | MB | 190 | 0.08 | HP | 200 | U | [57] |
| PVDF | MWCNT-COOH | [Bmim][PF$_6$] | MB | 210 | 0.25 | CM | 220 | U | [56] |
| PVDF | NaMMT | [Bmim][Cl] | SB | R | 24 | SC | 100 | 2 | [10] |
| PVDF | SiO$_2$ | [Emim][FSI] | So | R | 2 | SC | 130 | 24 | [52] |
| PVDF | TiO$_2$ | [Mcmim][Cl] | SB | 200 | 4 | HP | 200 | U | [7] |
| PVDF | PVP grains | [Emim][BF$_4$] | SB | R | U | S | R | U | [31] |
| PVDF-HFP | GO | [Emim][BF$_4$] | SB | 50 | U | SC | R | 12 | [32] |
| PVDF-HFP | MWCNTs | [Bmim][I] | SB | 60 | 2 | SC | 80 | 12 | [20] |
| PVDF-HFP | OMMT | [Bmim][Br] | Ul | R | 0.5 | S | R | 5 | [15,16] |
| PVDF-HFP | TiO$_2$ | [Bmim][BF$_4$] | So | R | 2 | SC | 90 | 24 | [36] |
| PVDF-HFP | SN | [Bmim][BF$_4$] | SB | R | 10 | SC | R | U | [37] |
| PVDF-HFP | SN | [Emim][CF$_3$SO$_3$] | SB | R | 24 | SC | R | U | [40] |

Ul = ultrasonication, MB = melt blending, SB = solution blending, So = sonication, HP = hot pressing, SC = solution casting, CM = compression molding, S = soaking, R = room, and U = unstated.

## 3. Effect of Imidazolium-Based Ionic Liquids on the Physicochemical Properties of the Blends and Composites

### 3.1. Effect of Hydrophilic Imidazolium-Based Ionic Liquids

Table 7 shows the physicochemical properties of PVDF/and PVDF-HFP/hydrophilic imidazolium-based ionic liquid blends and composites. PVDF was blended with [Emim][Cl] ionic liquid for the preparation of a PVDF/[Emim][Cl] blend [9]. The crystalline, mechanical, thermal, and chemical properties of the blend were characterized by means of differential scanning calorimeter, universal testing machine, and FTIR spectrometer. The crystalline properties, such as the crystallinity degree of the blend decreased, which was attributed to the existence of interactions between the ionic liquid and PVDF matrix. Moreover, the mechanical properties, such as the elastic modulus of the blend decreased because of the presence of [Emim][Cl], which acted as a plasticizer. In addition, the thermal properties, such as the melting temperature of the blend decreased owing to the strong electrostatic interactions between the polymer chains and the ionic liquid, which possessed a high nucleation effect. On the other hand, the chemical properties, such as the characteristic absorption bands of the α-phase, of the blend are completely disappeared, and β-phase absorption bands appeared very intensively compared to the neat PVDF. This was due to the existence of strong local dipole moments within the blend [9]. Therefore, it can be concluded that PVDF blended with [Emim][Cl] ionic liquid provides low crystallinity degree and elastic modulus blend, as well as low melting temperature, but its components have good interactions.

**Table 7.** Physicochemical properties of PVDF/and PVDF-HFP/hydrophilic imidazolium-based ionic liquid blends and composites.

| **Blend ǀ Composite** | **Hydrophilic Ionic Liquid** | **Physicochemical Properties *** | | | | **References** |
|---|---|---|---|---|---|---|
| | | **Crystalline** | **Mechanical** | **Thermal** | **Chemical** | |
| PVDF | [Emim][Cl] | ▼ | ▼ | ▼ | ▲ | [9] |
| PVDF-HFP | [Bmim][$BF_4$] | ▼ | n/a | ▼ | ▲ | [35] |
| PVDF/MWCNT-COOH | [Aeim][Br] | ▼ | ▲<br>▲ | ▲ | ▲ | [18] |
| PVDF/PVP grains | [Emim][$BF_4$] | ▼ | ▼ | ▼ | ▲ | [31] |
| PVDF-HFP/OMMT | [Bmim][Br] | ▼ | n/a | ▼ | ▲ | [15] |
| PVDF-HFP/SN | [Emim][$CF_3SO_3$] | ▼ | n/a | ▲<br>▲ | ▲ | [40] |

* The symbol '▲' corresponds to an increase in the properties and '▼' a decrease in the properties while 'n/a' describes not available.

Meanwhile, PVDF-HFP was blended with different amounts of [Bmim][$BF_4$] ionic liquid for the preparation of PVDF-HFP/[Bmim][$BF_4$] blends [35]. The crystalline, thermal, and chemical properties of the blends were characterized by using X-ray diffractometer, differential scanning calorimeter, and FTIR spectrometer. The crystalline properties, such as the degree of crystallinity of the blends decreased, which was caused by the inclusion of ionic liquid in the polymer matrix. Additionally, the thermal properties, such as the melting temperature of the blends decreased because of the plasticization effect of [Bmim][$BF_4$], as well as due to its complexation with the PVDF-HFP backbone. On top of that, the chemical properties, such as the absorption bands of the C−H stretching vibrations, of the blends shifted to lower wavenumber regions in comparison to the C−H stretching vibrations of the neat PVDF-HFP. This was attributed to the substantial conformational changes that occurred in the PVDF-HFP matrix after complexation with ionic liquid [35]. Thus, it can be inferred that PVDF-HFP blended with [Bmim][$BF_4$] ionic liquid gives low crystallinity degree and melting temperature blends. However, their components have a good interaction.

In addition, MWCNT-COOH modified with [Aeim][Br] ionic liquid was used for the preparation of PVDF/MWCNT-COOH-[Aeim][Br] composites [18]. The crystalline, mechanical, thermal, and chemical properties of the composites were characterized by means of X-ray diffractometer, universal testing machine, differential scanning calorimeter, and FTIR spectrometer. The crystalline properties, such as the crystallization peaks of the composite (1.0 wt.%) decreased because of the formation and stabilization of β-phase in the composite. Nevertheless, the mechanical properties, such as the Young's modulus, tensile strength, elongation at break, and toughness, of the composite (0.5 wt.%) significantly increased by up to 55%, 328%, 39%, and 160%, respectively, compared to the neat PVDF. This was caused by the reinforcing and toughening effects of [Aeim][Br]-modified MWCNT-COOH on the composite. Furthermore, the thermal properties, such as the melting temperature of the composite (1.0 wt.%), increased by up to 6.1% in comparison to the neat PVDF. This was because of the homogeneous dispersion of modified MWCNT-COOH, which caused a compact and higher melting crystal. On the other hand, the chemical properties, such as the absorption bands of the CF−CH−CF bending vibrations, of the composites shifted to higher wavenumber regions compared to the CF−CH−CF bending vibration of the neat PVDF. This was due to the dipole-dipole interaction between the >$CF_2$ dipoles of PVDF and imidazolium cations of modified MWCNT-COOH [18]. Hence, it can be deduced that PVDF mixed with MWCNT-COOH-[Aeim][Br] grants high tensile strength and toughness composites, as well as high melting temperature with good interaction of their components.

PVDF, PVP grains, and [Emim][$BF_4$] ionic liquid were used for the preparation of a PVDF/PVP/[Emim][$BF_4$] composite [31]. The crystalline, mechanical, thermal, and chemical properties of the composite were characterized by using X-ray diffractometer, nanoindentation, differential scanning calorimeter, and FTIR spectrometer. The crystalline properties, such as the $\alpha$-phase peaks intensity of the composite decreased, which was attributed to the introduction of PVP and ionic liquid into the PVDF matrix, which also increased the β-phase. Moreover, the mechanical properties, such as the Young's modulus and tensile strength of the composite decreased because of the plasticizing effect of [Emim][$BF_4$]. In addition, the thermal properties, such as the melting temperature of the composite decreased owing to the existence of interactions among PVDF, PVP, and ionic liquid. On top of that, the chemical properties, such as the characteristic absorption bands of PVDF, PVP, and [Emim][$BF_4$] are present in the composite. This was due to the strong electrostatic interactions of PVP and ionic liquid with the active hydrogen group on PVDF [31]. Therefore, it can be concluded that PVDF mixed with PVP grains and [Emim][$BF_4$] ionic liquid provides low Young's modulus and tensile strength composite, as well as low melting temperature, but its components have good interactions.

Meanwhile, PVDF-HFP, OMMT, and [Bmim][Br] ionic liquid were used for the preparation of PVDF-HFP/OMMT/[Bmim][Br] composites [15]. The crystalline, thermal, and chemical properties of the composites were characterized by means of X-ray diffractometer, thermogravimetric analyzer, and FTIR spectrometer. The crystalline properties, such as the crystallinity pattern of the composites decreased, which was caused by the formation of highly amorphous composites. Additionally, the thermal properties, such as the decomposition temperature of the composites decreased because of the complexation of [Bmim]$^+$ cations of ionic liquid with the PVDF-HFP, which destabilized the C−H bonds of the polymer matrix. On the other hand, the chemical properties, such as the absorption bands of the Si−O bond, of the composites shifted to lower wavenumber regions, and their intensity decreased sharply in comparison to the Si−O bond of the OMMT. This was attributed to the successful intercalation of PVDF-HFP into the galleries of the OMMT [15]. Thus, it can be inferred that PVDF-HFP mixed with OMMT and [Bmim][Br] ionic liquid gives low crystallinity and decomposition temperature composites. However, their components have a good interaction.

In addition, PVDF-HFP, SN, and [Emim][$CF_3SO_3$] ionic liquid were used for the preparation of PVDF-HFP/SN/[Emim][$CF_3SO_3$] composites [40]. The crystalline, thermal, and chemical properties of the composites were characterized by using X-ray diffractometer, differential scanning calorimeter, and FTIR spectrometer. The crystalline properties, such as the crystallization peaks of the composites decreased because of the predominant amorphous nature of the composites. Nevertheless, the thermal properties, such as the melting temperature of the composite (10 wt.%) significantly increased by up to 21% compared to the pure PVDF-HFP. This was caused by the phase transition of the composite that occurred from the ordered phase to the disordered phase. On top of that, the chemical properties, such as the absorption band of the amorphous phase, of the composite became considerably narrower in comparison to the amorphous phase of the pure PVDF-HFP. This was due to the interaction between the nitrile group of SN and the PVDF-HFP backbone [40]. Hence, it can be deduced that PVDF-HFP mixed with SN and [Emim][$CF_3SO_3$] ionic liquid grants low crystallinity and high melting temperature composites with good interaction of their components.

### 3.2. Effect of Hydrophobic Imidazolium-Based Ionic Liquids

Table 8 displays the physicochemical properties of PVDF/and PVDF-HFP/hydrophobic imidazolium-based ionic liquid blends and composites. PVDF was blended with different contents of [Emim][NTf$_2$] ionic liquid for the preparation of PVDF/[Emim][NTf$_2$] blends [43]. The crystalline, mechanical, thermal, and chemical properties of the blends were characterized by means of differential scanning calorimeter, autograph universal testing machine, and FTIR spectrometer. The crystalline properties, such as the degree of crystallinity of the blends decreased, which was attributed to the existence of interactions of the ionic liquid with the polymer matrix. Furthermore, the mechanical properties, such as the elastic modulus and yielding stress of the blends decreased because of the presence of [Emim][NTf$_2$], which acted as a plasticizer for PVDF. Additionally, the thermal properties, such as the melting temperature of the blends decreased owing to the destabilization of the crystalline phase. On the other hand, the chemical properties, such as the characteristic absorption bands of the polar β-phase are present in all the blends compared to the neat PVDF, and their intensity increased for the larger ionic liquid contents. This was due to the strong electrostatic interactions of the ionic liquid with the dipoles of the polymer chains, which gave rise to the nucleation of the β-phase of the polymer [43]. Therefore, it can be concluded that PVDF blended with [Emim][NTf$_2$] ionic liquid provides low elastic modulus and yielding stress blends, as well as low melting temperature, but their components have good interactions.

**Table 8.** Physicochemical properties of PVDF/and PVDF-HFP/hydrophobic imidazolium-based ionic liquid blends and composites.

| **Blend ǀ Composite** | **Hydrophobic Ionic Liquid** | **Physicochemical Properties *** | | | | **References** |
|---|---|---|---|---|---|---|
| | | **Crystalline** | **Mechanical** | **Thermal** | **Chemical** | |
| PVDF | [Emim][NTf$_2$] | ▼ | ▼ | ▼ | ▲ | [43] |
| PVDF | [Bmim][PF$_6$] | ▼ | ▲<br>▲ | ▲<br>▲ | ▲ | [55] |
| PVDF-HFP | [Emim][NTf$_2$] | ▼ | ▼ | ▼ | ▲ | [47] |
| PVDF/MWCNTs | [Emim][NTf$_2$] | ▼ | ▲ | ▲ | ▲ | [48] |
| PVDF/MWCNTs | [Bmim][PF$_6$] | ▼ | n/a | ▲<br>▲ | ▲ | [57] |
| PVDF/MWCNT-COOH | [Bmim][PF$_6$] | ▼ | ▲<br>▲ | n/a | ▲ | [56] |

* The symbol '▲' corresponds to an increase in the properties and '▼' a decrease in the properties while 'n/a' describes not available.

Meanwhile, PVDF was blended with different amounts of [Bmim][PF$_6$] ionic liquid for the preparation of PVDF/[Bmim][PF$_6$] blends [55]. The crystalline, mechanical, thermal, and chemical properties of the blends were characterized by using X-ray diffractometer, universal testing machine, differential scanning calorimeter, and FTIR spectrometer. The crystalline properties, such as the α characteristic peaks of the blends disappeared, which was due to the decrease of α-phase content in the blends. Nonetheless, the mechanical properties, such as the tensile strength and elongation at break of the blend (100/20) significantly increased by up to 77% and 173%, respectively, compared to the neat PVDF. This was caused by the improvement of the ductility of the blend, which enhanced stretchability and strength at break simultaneously. Moreover, the thermal properties, such as the melting temperature of the blend (100/2), considerably increased by up to 5.3% in comparison to the neat PVDF. This was because of the transformation of the nonpolar α-phase for PVDF to the polar γ-phase with the introduction of a small amount of ionic liquid. On top of that, the chemical properties, such as the absorption bands of the $CF_2-CH_2$ bending vibrations, of the blends shifted to higher wavenumber regions compared to the $CF_2-CH_2$ bending vibration of the neat PVDF. This was due to the $>CF_2$ groups in the PVDF chains have specific interactions with the imidazolium ring in the ionic liquid [55]. Thus, it can be inferred that PVDF blended with [Bmim][$PF_6$] ionic liquid gives high tensile strength and elongation blends, as well as high melting temperature with good interactions of their components.

In addition, PVDF-HFP was blended with different contents of [Emim][$NTf_2$] ionic liquid for the preparation of PVDF-HFP/[Emim][$NTf_2$] blends [47]. The crystalline, mechanical, thermal, and chemical properties of the blends were characterized by means of universal testing machine, differential scanning calorimeter, and FTIR spectrometer. The crystalline properties, such as the degree of crystallinity of the blends decreased, which was attributed to the strong interaction of the ionic liquid with the polar units. Moreover, the mechanical properties, such as the tensile stress and Young's modulus of the blends decreased because of the introduction of [Emim][$NTf_2$] into the polymer matrix, which induced a plasticizing behavior of the blends. In addition, the thermal properties, such as the melting temperature of the blends decreased owing to the plasticization effect of the ionic liquid and its complexation with the PVDF-HFP backbone. On the other hand, the chemical properties, such as the characteristic absorption bands of the β-phase content of the blends increased, which was due to the nucleation of the β-phase through the electrostatic interactions between the imidazolium cations of the ionic liquid and the negatively polarized $CF_2$ groups of PVDF-HFP [47]. Hence, it can be deduced that PVDF-HFP blended with [Emim][$NTf_2$] ionic liquid grants low tensile stress and Young's modulus blends, as well as low melting temperature. However, their components have good interactions.

MWCNTs modified with [Emim][$NTf_2$] ionic liquid was used for the preparation of PVDF/MWCNTs-[Emim][$NTf_2$] composites [48]. The crystalline, mechanical, thermal, and chemical properties of the composites were characterized by using X-ray diffractometer, micro universal testing machine, differential scanning calorimeter, and FTIR spectrometer. The crystalline properties, such as the $\alpha$-phase peaks intensity of the composites decreased, which was attributed to the presence of the [Emim][$NTf_2$]-modified MWCNTs in the composites, which also induced the formation of the $\gamma$-phase from $\alpha$-phase. Nonetheless, the mechanical properties, such as the tensile strength, elongation at break, and Young's modulus of the composite (1.0 wt.%) increased by up to 14%, 20%, and 3.1%, respectively, compared to the neat PVDF. This was because of the improvement of the strength and ductility of the composite. In addition, the thermal properties, such as the melting temperature of the composite (1.0 wt.%) increased by up to 4.5% in comparison to the neat PVDF. This was owing to the existence of polar $\gamma$-phase in the composite, whereby its melting temperature was higher than the nonpolar $\alpha$-phase. On top of that, the chemical properties, such as the absorption bands of the $CH-CF-CH$ stretching vibrations, of the composites shifted to higher wavenumber regions compared to the $CH-CF-CH$ stretching vibration of the neat PVDF. This was due to the electrostatic interaction between the anion of the ionic liquid and PVDF [48]. Therefore, it can be concluded that PVDF mixed with MWCNTs-[Emim][$NTf_2$] provides high tensile strength and elongation composites, as well as high melting temperature with good interaction of their components.

Meanwhile, MWCNTs modified with [Bmim][$PF_6$] ionic liquid was used for the preparation of PVDF/MWCNTs-[Bmim][$PF_6$] composites [57]. The crystalline, thermal, and chemical properties of the composites were characterized by means of X-ray diffractometer, differential scanning calorimeter, and FTIR spectrometer. The crystalline properties, such as the $\alpha$ characteristic diffraction peaks intensity of the composites decreased, which was caused by the increase of polar phases amount formed in the composites. Additionally, the thermal properties, such as the melting temperature of the composite (1:1) significantly increased by up to 4.8% in comparison to the neat PVDF. This was because the polar phases in the composite possess a higher melting temperature than the nonpolar phase in the polymer. On the other hand, the chemical properties, such as the absorption bands of the $CF_2-CH_2$ bending vibrations, of the composites shifted to higher wavenumber regions compared to the $CF_2-CH_2$ bending vibration of the neat PVDF. This was attributed to the specific interactions of the $>CF_2$ groups in the polymer chains with the imidazolium ring in the ionic liquid-modified MWCNTs [57]. Thus, it can be inferred that PVDF mixed with MWCNTs-[Bmim][$PF_6$] gives low crystallinity and high melting temperature composites with good interactions of their components.

In addition, MWCNT-COOH modified with [Bmim][$PF_6$] ionic liquid was used for the preparation of PVDF/MWCNT-COOH-[Bmim][$PF_6$] composites [56]. The crystalline, mechanical, and chemical properties of the composites were characterized by using tensile machine and FTIR spectrometer. The crystalline properties, such as the absorption peaks of the nonpolar $\alpha$-phase crystals of the composites disappeared, which was attributed to the formation of polar $\beta$- and $\gamma$-phase crystals in the composites, which also resulted from the interfacial interactions between the PVDF chains and [Bmim][$PF_6$]-modified MWCNT-COOH. Nevertheless, the mechanical properties, such as the elongation at break of the composite (10:2) significantly increased by up to 326% compared to the neat PVDF. This was because of the presence of modified MWCNT-COOH, which induced toughness improvement of the composite. On top of that, the chemical properties, such as the absorption bands of the $-CF_2$ stretching vibrations, of the composites shifted to lower wavenumber regions in comparison to the $-CF_2$ stretching vibration of the neat PVDF. This was due to the interactions between the $-CF_2$ and imidazolium rings on ionic liquid-modified MWCNT-COOH [56]. Hence, it can be deduced that PVDF mixed with MWCNT-COOH-[Bmim][$PF_6$] grants low nonpolar crystals and high elongation at break composites with good interactions of their components.

## 4. Applications of PVDF and PVDF-HFP Blends and Composites

### *4.1. PVDF/and PVDF-HFP/Hydrophilic Imidazolium-Based Ionic Liquid Blends and Composites*

Table 9 shows the applications of PVDF/and PVDF-HFP/hydrophilic imidazolium-based ionic liquid blends and composites. It can be observed that the hydrophilic ionic liquids are frequently utilized in PVDF and PVDF-HFP blends and composites, this is probably due to their low cost and ready availability. On the other hand, the blends and composites are predominantly employed in electronic and electric applications, for instance, capacitors [20,22,26], electrolyte membranes [34,35], dielectric composites [38,39], and actuators [9,11]. Furthermore, they are also applied in gas processing, for example, carbon dioxide capture [21], as well as in water and wastewater treatment as antifouling membranes [7]. The utilization of [Emim][DCA] exhibits double advantages in PVDF-HFP blends, specifically it provided additional charge carriers, and it also reduced the crystallinity of polymer matrix, which consequently increased the ionic conductivity of the fabricated capacitors [22]. Moreover, the value of ionic conductivity of PVDF-HFP blends increases with an increase in the interaction of [Bmim][I] with the polymer matrix, which facilitated the ionic motion of electrolyte membranes [19]. Additionally, [Veim][$BF_4$] was utilized in PVDF/MWCNTs composites increased the dispersion of filler in the polymer matrix and induced the polar $\beta$-phase of the composites, which improved their dielectric constant by up to three times than the neat PVDF [38]. Furthermore, the bending movement of the PVDF blends increases with increasing [Hmim][Cl] content, which ascribed to their low degree of crystallinity and elastic modulus, and high AC electrical conductivity value of the prepared actuators [11].

**Table 9.** Applications of PVDF/and PVDF-HFP/hydrophilic imidazolium-based ionic liquid blends and composites.

| Blend I Composite | Application | References |
|---|---|---|
| PVDF/[Hmim][Cl] | Electromechanical actuators | [9,11] |
| PVDF/[Vbim][Cl] | Superthin dielectric capacitors | [12,13] |
| PVDF/[Emim][OAc] | Carbon dioxide capture fibers | [21] |
| PVDF/[Emim][$EtSO_4$] | Carbon dioxide capture fibers | [21] |
| PVDF/[Bmim][$H_2PO_4$] | Proton exchange membranes | [23] |
| PVDF/[Emim][$BF_4$] | Flexible electronics | [30] |
| PVDF-HFP/[Bmim][I] | Polymer electrolyte membranes | [19] |
| PVDF-HFP/[Emim][DCA] | Electrochemical double-layer capacitors | [22] |
| PVDF-HFP/[Emim][SCN] | Electrochemical double-layer capacitors | [26] |
| PVDF-HFP/[Bmim][$BF_4$] | Polymer electrolyte membranes | [34,35] |
| PVDF/Gr/[Hdmim][Br] | High-charge storage capacitors | [17] |
| PVDF/GO/[Veim][$BF_4$] | Polymeric dielectric composites | [38] |
| PVDF/MWCNTs/[Veim][$BF_4$] | Flexible dielectric materials | [39] |
| PVDF/$TiO_2$/[Mcmim][Cl] | Antifouling membranes | [7] |
| PVDF/PVP grains/[Emim][$BF_4$] | Flexible actuators | [31] |
| PVDF-HFP/GO/[Emim][$BF_4$] | Flexible solid-state supercapacitors | [32] |
| PVDF-HFP/MWCNTs/[Bmim][I] | All-solid-state supercapacitors | [20] |
| PVDF-HFP/OMMT/[Bmim][Br] | Composite electrolyte membranes | [15,16] |
| PVDF-HFP/$TiO_2$/[Bmim][$BF_4$] | Electrochemical double-layer capacitors | [36] |
| PVDF-HFP/SN/[Bmim][$BF_4$] | Solid-state supercapacitors | [37] |
| PVDF-HFP/SN/[Emim][$CF_3SO_3$] | Electrochemical double-layer capacitors | [40] |

### *4.2. PVDF/and PVDF-HFP/Hydrophobic Imidazolium-Based Ionic Liquid Blends and Composites*

Table 10 indicates the applications of PVDF/and PVDF-HFP/hydrophobic imidazolium-based ionic liquid blends and composites. It is seen that the blends and composites are typically utilized for application in electronic and electric, for example, capacitors [46,52], actuators [8,9], polymer electrolytes [50,59], sensor [27], anti-electrostatic films [55], flexible organic memory [51], flexible dielectric [58], and electromagnetic interference shield [48]. In addition, they can also be applied in gas processing as membranes for the separation of carbon dioxide [49,60]. The utilization of PVDF-HFP/[Emim][$NTf_2$] blend in the fabricated capacitor increased the specific capacitance at high current density, which exhibited superior performances [46]. Meanwhile, the bending response of prepared actuators also depends on the content of [Emim][$NTf_2$] in PVDF, which increased with increasing ionic liquid content and the blend thickness [8]. On the other hand, the polymer electrolyte prepared from the PVDF/[Bzmim][$PF_6$] blends possesses high values of the DC conductivity, which was induced by the increase of polar phase, ion mobility, and ion concentration [59]. The gas sensor based on PVDF/[Emim][$NTf_2$] blend owned high sensitivity, fast response/recovery times, and low sensor response hysteresis [27]. Additionally, the good interaction between PVDF and [Bmim][$PF_6$] provides permanent antistatic properties, which suitable for the production of anti-electrostatic films [55]. Moreover, the presence of [Emim][$NTf_2$] improved the dispersion of MWCNTs in the PVDF matrix, which enhanced the electrical conductivity of the composites, and resulted in better electromagnetic interference shielding properties [48].

**Table 10.** Applications of PVDF/and PVDF-HFP/hydrophobic imidazolium-based ionic liquid blends and composites.

| Blend \| Composite | Application | References |
|---|---|---|
| PVDF/[Emim][NTf$_2$] | Electromechanical actuators | [8,9] |
| PVDF/[Emim][NTf$_2$] | Nitrogen dioxide sensor | [27] |
| PVDF/[Pmim][NTf$_2$] | Electromechanical actuators | [41] |
| PVDF/[Bmim][NTf$_2$] | Carbon dioxide separation membranes | [49] |
| PVDF/[Emim][B(CN)$_4$] | Carbon dioxide separation membranes | [60] |
| PVDF/[Bmim][PF$_6$] | Carbon dioxide separation membranes | [49] |
| PVDF/[Bmim][PF$_6$] | Transparent anti-electrostatic films | [55] |
| PVDF/[Bzmim][PF$_6$] | Solid polymer-based electrolytes | [59] |
| PVDF/[Hmim][FAP] | Nitrogen dioxide sensor | [27] |
| PVDF-HFP/[Emim][NTf$_2$] | Flexible supercapacitors | [46] |
| PVDF-HFP/[Emim][NTf$_2$] | Composite bending actuators | [47] |
| PVDF-HFP/[Mmpim][NTf$_2$] | Polymer electrolyte films | [50] |
| PVDF-HFP/[Emim][TFSA] | Flexible organic memory | [51] |
| PVDF-HFP/[Emim][FAP] | Electrochemical double-layer capacitors | [53] |
| PVDF-HFP/[Emim][B(CN)$_4$] | Electrochemical double-layer capacitors | [61] |
| PVDF/Gr/[Bmim][PF$_6$] | Flexible dielectric materials | [58] |
| PVDF/MWCNTs/[Emim][NTf$_2$] | Electromagnetic interference shielding materials | [48] |
| PVDF/SiO$_2$/[Emim][FSI] | Solid supercapacitors | [52] |

## 5. Conclusions

Two types of ionic liquids, blending processes of PVDF and PVDF-HFP blends, and mixing processes of PVDF and PVDF-HFP composites have been succinctly reviewed in this paper. The physicochemical properties, for instance, crystalline, mechanical, thermal, and chemical of the blends and composites have also been revisited in this succinct review. Hydrophilic and hydrophobic imidazolium-based ionic liquids are the two most significant ionic liquids utilized for the preparation of the blends and composites. The utilization of hydrophilic imidazolium-based ionic liquids can efficiently decrease the crystalline, mechanical, and thermal properties of the PVDF and PVDF-HFP blends and composites. On top of that, the utilization of hydrophobic imidazolium-based ionic liquids can effectively increase the mechanical and thermal properties of the blends and composites. In addition, the ionic liquids can form specific interactions with polymer matrices and fillers. The blends and composites can be applied in electronic and electric appliances, such as capacitors, polymer electrolytes, actuators, dielectric materials, sensor, etc. This succinct review may be useful for the preparation of PVDF and PVDF-HFP blends and composites by utilizing ionic liquids in the diversification of thermoplastic fluoropolymers functions for a multiplicity of applications.

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