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REVIEW PAPER

PVDF-based composites for electromagnetic shielding application: a review

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Abstract

With the rapid development of information technology, electromagnetic shielding materials are playing an increasingly significant role in electronic reliability, healthcare, and national defense security. Hence, developing high performance electromagnetic shielding materials with thin thickness, low density, wide bandwidth, and strong absorption has attracted great interests. Recently, polyvinylidene fluoride (PVDF) as high-performance electromagnetic shielding materials has grabbed considerable attention, owing to its low density, good flexibility, stable corrosion resistance and favorable shaping capability. In this review, we firstly introduce the theory of electromagnetic shielding. In the main part, the preparation and recent advances of PVDF-based electromagnetic shielding composites are summarized, including single-, binary-, and multi-component filler composites, microstructure design of composites, and the factors influencing the EMI SE performance. The key point to enhance the EMI SE performance is to modulate the electromagnetic and dielectric properties of the composites to create diversified loss mechanisms. Finally, the shortcomings, challenges, and prospects of PVDF-based electromagnetic shielding materials are also put forward, which will be helpful to people working in the related fields.

Keywords PVDF-based composites · Electromagnetic shielding materials · Electromagnetic shielding mechanism · Absorption · Reflection · Multiple reflection

Introduction

As science and technology progresses by leaps and bounds in modern times, various electronic and electrical equipment, such as cellular towers, wireless devices, smartphones, palm computer, a variety of household appliances and modern radar systems, have enabled more intimate connections between human within societies and brought great convenience for people's daily life [1–3]. Nevertheless, electromagnetic radiations generated by electronics and electrical equipment have become an unintended consequence as they create high-energy hot spots that can remarkably reduce the service life of electric components [4–7]. In addition, the rapid development of 5G industry makes the electromagnetic interference (EMI) between components and equipment more and more serious [8, 9]. Nowadays, EM waves have been identified as a new source of pollution, which not only disrupts normal communication, but could also pose a potential threat to human health [10, 11]. As electronics and electrical equipment become increasingly

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digitized, integrated, and moving toward lower power to meet the requirements of high-speed, lightweight and miniaturization, they become more susceptible to external EMI [8, 12]. Thus, eliminating or blocking of the undesirable electromagnetic radiations has become an inevitable topic.

Metals such as Cu, Al, Ag, and stainless steel have been widely used against electromagnetic pollution owing to their high conductivity [13, 14]. However, their high density, difficult processability, and high corrosion susceptibility have limited their applications in highly integrated modern mobile electronics [15, 16]. Compared with traditional metal-based materials, polymer composites with fillers (such as metal fillers [17, 18], carbon fillers [19, 20], conducting polymers [21, 22], and dielectric [23, 24]/magnetic [25, 26] materials) have become the focus of contemporary research, owing to their low density, good flexibility, stable corrosion resistance, favorable shaping capability, high electrical and thermal conductivity and excellent EMI shielding effectiveness (SE) performance [27–29].

The EMI SE performance of polymer composites depends on various factors such as the type [30, 31], morphology [32–34] and electromagnetic properties of fillers [35–37], the dispersion state of fillers in the polymer matrix [38–40], structure [41–44] and thickness [45–47] of composites. Therefore, high performance polymer electromagnetic shielding composites can be constructed by proper selection of filler types, preparation methods, and optimal structural designs.

polyvinylidene fluoride (PVDF), a semi-crystalline fluoropolymer with a repeating unit of CH_2-CF_2 [48–50], has been used in polymer sensors, heat exchangers, and housings for home appliances, due to its high mechanical strength, excellent chemical resistance, and good radiation resistance [51–54]. In recent works, PVDF composites filled with a wide range of nanoparticles were used for EMI shielding [55–58]. The results showed that the strong polar fluorine atoms in PVDF would contribute a better interaction with nano-fillers [59–61]. Based on its unique structure and properties, PVDF has excellent application prospects in the field of electromagnetic shielding [14, 62–64]. The design of efficient PVDF-based electromagnetic shielding composite has gradually become a research focus in the field of electromagnetic shielding technology [50, 61, 65, 66].

In this paper, we were reviewed the mechanism of electromagnetic shielding and the preparation method of PVDF-based electromagnetic shielding composites, on this basis, the research progress of lightweight flexible PVDF-based electromagnetic shielding materials are summarized from the proper selection of fillers to the factors influencing the EMI SE performances. Meanwhile, the future development prospects of lightweight flexible PVDF-based electromagnetic shielding materials are also prospected, which will be helpful to staff working in the related fields.

Mechanism of EMI shielding effectiveness

Various theories such as electromagnetic field and transmission line are usually used to explain the mechanism of electromagnetic shielding. In this paper, the transmission line theory with accurate and simple calculation is used to explain the mechanism of electromagnetic shielding [67]. In transmission line theory, electromagnetic shielding enclosure is usually regarded as a section of transmission line [68]. When the electromagnetic shielding enclosure is approached by electromagnetic wave, the electromagnetic shielding enclosure attenuates the electromagnetic wave through three processes [69, 70] (as shown in Fig. 1). First, the electromagnetic wave is transmitted to the surface of the electromagnetic wave shielding enclosure. Due to the impedance mismatch between the interface of the

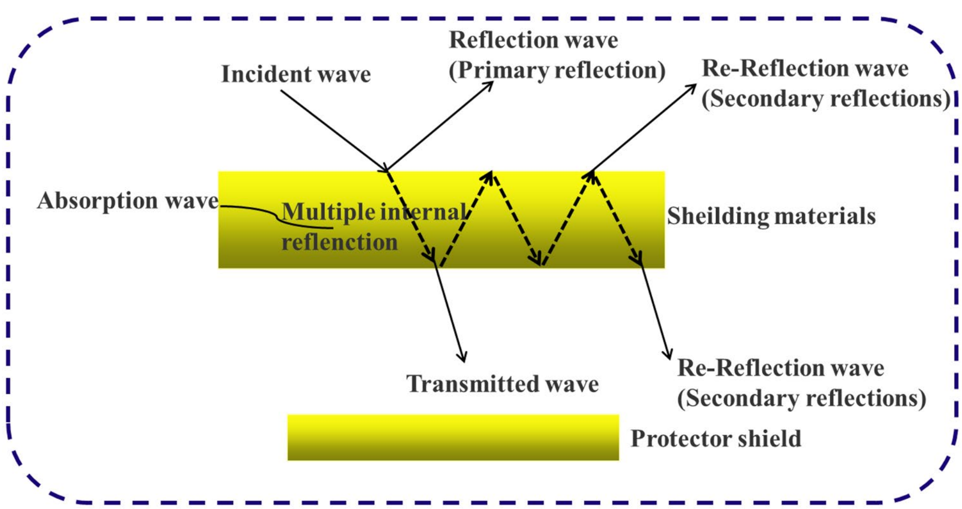

Fig. 1 Schematic illustration of electromagnetic wave strike process on protected device

electromagnetic wave shielding enclosure and the free interface of the air, part of the electromagnetic wave is reflected off the surface of the electromagnetic wave shielding enclosure, causing the reflection attenuation (SE_R) of the electromagnetic wave, and the electromagnetic wave entering the electromagnetic wave shielding enclosure will be relatively reduced. Second, the unreflected electromagnetic wave enters the electromagnetic wave shielding enclosure and transmits in the enclosure. As the energy of electromagnetic wave is absorbed (SE_A) by the electromagnetic wave shielding enclosure, so the electromagnetic wave attenuated again. Third, when the remaining electromagnetic wave is transmitted to the edge of the electromagnetic wave shielding enclosure, it is reflected back into the electromagnetic wave shielding enclosure again. After energy is further absorbed by the multiple reflection (SE_M) between the electromagnetic wave shielding enclosure interface and the free interface, the purpose of attenuating the transmitted electromagnetic wave is achieved, thus the protected components or environment are not subject to electromagnetic pollution. The shielding performance of the shielding materials is usually determined by the shielding efficiency (SE) [70, 71], which can be expressed by Eq. (1).

$$SE = SE_R + SE_A + SE_M \tag{1}$$

EMI SE is closely related to the charge, current and polarization phenomena induced on the surface of the shielding structure and inside of the shielding enclosure [69, 70]. When the electromagnetic wave is reflected and lost on the surface of the electromagnetic wave shield, there is a poor impedance matching effect between the surface of the shielding material and the free interface, and the electric charge can be induced in the magnetic field inside the shielding material, which requires the shielding material to have good conductivity [17, 72]; when the unreflected electromagnetic wave enters the electromagnetic shielding enclosure for absorption and attenuation, the shielding material has a large number of electric or magnetic dipole, causing dipole oriented polarization in the magnetic field, which requires the shielding material to have high permeability, high electromagnetic loss [26, 73] and suitable dielectric constant [23, 74]; when the remaining electromagnetic wave is transmitted to the transmission edge of the electromagnetic shielding enclosure for multiple reflection attenuation, the shielding materials with porous structure accompanied by a large number of interfaces, can improve the multiple reflection and multiple scattering times of electromagnetic wave, thus the SE of shielding materials can be effectively improved [42, 44, 75]. Therefore, a successful electromagnetic shielding materials should not only have good reflectivity, but also possess good electromagnetic wave absorption property [36, 59].

Preparation and electromagnetic shielding performance of PVDF-based composites

PVDF-based electromagnetic shielding composites are composed of electrically insulated PVDF [76, 77], conductive fillers [78–80] (such as metallic fillers, carbon materials, intrinsically conducting polymers) and/or magnetic fillers [30, 81](e.g., Fe_3O_4, Fe_2O_3, and barium ferrite (BF) and/or dielectric fillers [82, 83] (e.g., $BaTiO_3$, barium strontium titanate, ZnO, MnO_2, SiC, SiO_2). PVDF-based nanocomposite materials were fabricated by melt blending [84, 85], solution blending [17, 51], and/or followed by hot pressing [18, 63], and so on. These materials can significantly or permanently reducing electromagnetic radiation [73, 86]. The fillers were the main factors of electromagnetic shielding performance of electromagnetic shielding materials, and combined with proper preparation technologies and optimized structural designs, the shielding effect of PVDF nano composites can be further improved [61, 87]. Researches on PVDF-based electromagnetic shielding composites are summarized in Table 1.

PVDF/pure conductive-filler composites

As PVDF is an electrical insulator [84, 92], the electrical conductivity of fillers is one of the main factors affecting the EMI SE performance of shielding materials [63, 80, 99, 124]. Conductive fillers such as metals [18, 47], carbons [112, 114], conductive polymers [21, 22], and MXenes [124, 125] provides excellent electric conductivity. When these fillers blend with PVDF, they could make great contribution to the construction of a continuous conductive network and thus enhance the electrical conductivity and EMI SE performance of PVDF-based composites [63, 80, 106].

PVDF/metal composites

Take the advantage of the excellent conductivity, metals were initially used as electromagnetic shielding materials [84]. However, due to the high density [85] and price [88] of pure metal electromagnetic shielding materials, easy of oxidation or reaction with chemicals [89], which may reduce the conductivity over time [89], as well as their poor wear resistance and scratch resistance [91], pure metal materials have great limitations in the application of electromagnetic shielding [72]. Therefore, metals are also compounded with other materials to achieve better electromagnetic shielding performance [47]. For example, the light weight electromagnetic shielding material with better performance can be prepared by compounding metal fillers with polymers [17]. The composites formed by incorporating a small amount of metal

Table 1 EMI SE performance of PVDF-based composites

materials		thickness (mm)	EMI SE (dB)	Frequency	Ref.
Matrix	Filler loading				
PVDF	10 wt% Cu	-	40	3.06 EHz	[84]
PVDF	15 wt% Al	-	20	2 EHz	[85]
PVDF	0.5 wt% AgNWs	0.098	107.2	8–12 GHz	[79]
PVDF	50 vol% CIP	1.2	20	8–12 GHz	[88]
PVDF	40 vol%μ-Ni	1.95	23	8–12 GHz	[89]
PVDF	35 vol% n-Ni	1.95	42.87	8–12 GHz	[72]
PVDF	35 vol% n-Fe	1.93	40.21	8–12 GHz	[35]
PVDF	6 wt% Ni chains	0.5	35.4	18-26.5 GHz	[47]
PVDF	30 wt% Ni chains	4.5	35	12–18 GHz	[17]
PVDF	9 wt% NiNWs		43	8–12 GHz	[90]
PVDF	10 wt% Ni chains	2	26.8	8–12 GHz	[63]
PVDF	15 wt% NiO	0.3	12	8–12 GHz	[91]
PVDF	50 wt% LMPA	-	26.8	8–12 GHz	[18]
PVDF	10 wt% CF	0.9	12	0.1–1.6 GHz	[92]
PVDF/ PMMA	40 wt% CF	0.32	20	150–1200 GHz	[93]
PVDF	8 wt% CF		17.3	0.015–3 GHz	[94]
PVDF	8 wt% CF		10	0.015–3 GHz	[95]
PVDF	15 wt% FGS	2	45.6	8–12 GHz	[96]
PVDF/PVP	5 wt% MWCNTs	0.06	20	0.1–1.5 GHz	[97]
PVDF	40 wt% MWCNTs	6.8	30	8.2 GHz	[98]
PVDF	70 vol % graphite	2	93	8–26 GHz	[99]
PVDF	80 wt% carbonized charcoal	2.8	70.1	8–12 GHz	[100]
PVDF	80 wt% graphite	1.8	98.47	8–12 GHz	[101]
PVDF	50 wt% particulate nano carbon	9	50	8–12 GHz	[102]
PVDF	5 g/L graphene	0.652	20	1–8 GHz	[103]
PVDF	25 g/L graphene	0.598	31.2	100–3000 MHz	[104]
PVDF	10 wt% GPs	0.4	16	8–12 GHz	[51]
PVDF	15 wt% MWCNTs	0.02	47	8–12 GHz	[52]
PVDF	5 wt% graphene	-	20	8–12 GHz	[87]
PVDF	2 wt% MWCNTs	-	20	12–18 GHz	[105]
PVDF	0.5 wt% MWCNTs	0.3	98	1–18 GHz	[39]
PVDF	0.5 wt% MWCNTs	0.3	26.5	8–18 GHz	[40]
PVDF/ABS	3 wt% PMMA-MWCNTs	-	32	8–18 GHz	[106]
PVDF PS/HDPE	1.6 vol% MWCNTs	-	31	8–12 GHz	[20]
PVDF-HFP	40 wt% RGO	-	30	8–12 GHz	[107]
PVDF	2 wt% MWCNTs	-	25	12–18 GHz	[108]
PVDF/OBC	2.7 wt% MWCNTs	2	34	8–12 GHz	[109]
PVDF/PLA	0.25 wt% MWCNTs	1	7.86	1–6 GHz	[110]
PVDF/PS	3 wt% MWCNTs	1	19	8–12 GHz	[111]
PVDF/POK	2 vol% of MWCNTs	0.9	21	8–12 GHz	[112]
PVDF	3.5 wt% MWCNTs	1.1	17.7	8–12 GHz	[113]
PVDF	3.5 wt% MWCNTs	1.1	9.5	8–12 GHz	[33]
PVDF	2 wt% MWCNTs	0.05	7.77	8–12 GHz	[114]
PVDF	3.5 wt% MWCNs	1.1	20.3	8–12 GHz	[115]
PVDF	3.5 wt% MWCNTs	1.1	20.2	8–12 GHz	[34].
PVDF	3.5 wt% MWCNTs	1.1	16.7	8–12 GHz	[116]
PVDF	50 wt% Co@C	1	24.06	8–12 GHz	[77].
PVDF	50 wt% CoNi@C	1	27	8–12 GHz	[32]

Table 1 (continued)

materials		thickness (mm)	EMI SE (dB)	Frequency	Ref.
Matrix	Filler loading				
PVDF	50 wt%Fe_3C@C	1	21	8–12 GHz	[117]
PVDF	50 wt%Ni@C	1	20.5	8–12 GHz	[118]
PVDF	9 wt% MWCNTs	0.3	25	8–12 GHz	[119]
PVDF	15 wt% MWCNTs	2	56.72	8–12 GHz	[42]
PVDF	10 wt% graphene	1.5	37.4	26.5–40 GHz	[120]
PVDF	8 wt% MWCNTs	4	132.6	26.5–40 GHz	[19]
PVDF	4 wt% graphene	0.02	50	8–12 GHz	[121]
PVDF	7 wt% MWCNTs	0.6	30.89	8–12 GHz	[122]
PVDF-co-HFP	52 wt% PANI	1	6	8–12 GHz	[21]
PVDF/ PEDOT	18 wt%PEDOT	0.014	40	8–12 GHz	[22]
PVDF-co-HFP	30 wt% PANI	1	5	8–12 GHz	[123]
PVDF	30 wt% PANI	1	65	8–12 GHz	[80]
PVDF	90 wt% MXenes	0.017	42.9	8–12 GHz	[124]
PVDF/PS	12 wt% MXenes	1.35	55	8–12 GHz	[125]
PVDF	5 wt% $Ba_{0.2}Mg_{0.8}Fe_2O_4$	1.5	20	2–8 GHz	[126]
PVDF	30 wt% $CuFe_2O_4$	-	5	8–12 GHz	[30]
PVDF	20 wt% $BaFe_{12}O_{19}$	0.21	97.6	8–18 GHz	[26]
PVDF	20 wt% $Ba_4Co_2Fe_{36}O_{60}$	0.122	83	8–18 GHz	[81]
PVDF	90 wt% $BaFe_{11\cdot7}A_{l0\cdot3}O_{19}$	7	5.24	2.45 GHz	[25]
PVDF	50 wt% ZnO	0.13	8	8–12 GHz	[24]
PVDF	25 wt% h-BNNPs	5.6	11.2	8–12 GHz	[82]
PVDF	30 wt% CuS	-	44	2–18 GHz	[23]
PVDF	50 wt% $CuCo_2S_4$	2.5	20	8–18 GHz	[83]
PVDF	50 wt% $La_{0.8}Sr_{0.2}MnO_3$/La/Sr	1.75	20	8–18 GHz	[127]
PVDF/PVC	10 wt% $SrTiO_3$	-	12.5	12–18 GHz	[74]
PVDF	5 wt%Ag-10 wt% graphite	0.5	29.1	8–12 GHz	[128]
PVDF	3 wt%Au–3 wt% MWCNTs	0.5	26.7	8–12 GHz	[86]
PVDF	Fe@CF	0.6	54	12–18 GHz	[129]
PVDF	5 wt% MWCNTs/10 wt% CoNi	0.9	30	12–18 GHz	[130]
PVDF	3 wt% MWCNTs/50 wt% CoNi	-	41	12–18 GHz	[131]
PVDF	3 wt% MWCNTs/50 wt% NiFe	-	35	12–18 GHz	[132]
PVDF/SMA	5 wt% MWCNTs/30 wt% NiFe	-	23	8–18 GHz	[38]
PVDF/TPU	5 wt% MWCNTs@ NiFe	1.4	35.7	8–18 GHz	[133]
PVDF	6 wt% MWCNTs@Co chains	0.3	41	18-26.5 GHz	[134]
PVDF	3 wt% CB/20 wt% CI	2	27	8–18 GHz	[135]
PVDF	5 wt% graphene/8 wt% Ni chains	0.6	57.3	8–18 GHz	[45]
PVDF	1 wt% MWCNTs/6 wt% Ni chains	0.6	55.8	8–18 GHz	[45]
PVDF	10 wt% graphene/10 wt% Ni chains	0.7	51.4	8–12 GHz	[14]
PVDF	2.0 mg/cm^2 MWCNTs and1.9 mg/cm^2 AgNWs	0.74	34	8–12 GHz	[37]
PVDF	3 wt% MWCNTs/10 wt% NiNWs	1.1	22	8–12 GHz	[136]
PVDF	3 wt% MWCNTs/10 wt% AgNWs	1.1	27	8–12 GHz	[136]
PVDF	4 wt% MWCNTs/IL	1.8	46	12–18 GHz	[137]
PVDF	10 wt% MWCNTs/12 wt% Ni@MWCNTs	0.5	46.6	8–18 GHz	[138]
PVDF	50 wt% Ni@CMS	2	20	8–18 GHz	[139]
PVDF	15 wt% AgNPs@CB/graphite	4	35	12–18 GHz	[140]
PVDF	50 wt% FeC	4.3	23.9	8–18 GHz	[73]
PVDF	10 wt% MXenes/10 wt% Ni chains	0.36	34.4	8–12 GHz	[36]
PVDF	10 wt% MXenes/5 wt% AgNWs	0.6	41.26	8–12 GHz	[141]

Table 1 (continued)

materials		thickness (mm)	EMI SE (dB)	Frequency	Ref.
Matrix	Filler loading				
PVDF	0.5 mg/mL MXenes/1.28 wt % AgNWs	-	47.8	8–12 GHz	[142]
PVDF/PETG	5 wt% long CF/15 wt% CB	4	30	0.1–1500 MHz	[143]
PVDF	0.5 wt% MWCNTs /10 wt% graphite	0.25	36.46	8–12 GHz	[144]
PVDF	0.5 wt% MWCNTs /10 wt%	-	14.64	8–12 GHz	[145]
PVDF	2 wt% MWCNTs	1.1	11.6	8–12 GHz	[146]
PVDF	40 wt% graphene/NWF	2	48.5	0.03–1.5 GHz	[147]
PVDF	7.5 wt% CB/2 wt% graphite	1	16	8–12 GHz	[148]
PVDF/PC	3 wt% MWCNTs/30 wt% NF	-	20	8–18 GHz	[149]
PVDF/SAN	2 wt% MWCNTs-5 wt%NF	5	26	8–18 GHz	[150]
PVDF/ABS	2 wt% MWCNTs-5 wt% Fe_3O_4	-	26.5	2–18 GHz	[151]
PVDF/PC	3 wt% MWCNTs-Fe_3O_4	0.9	31	8–18 GHz	[59]
PVDF/ABS	1 wt% MWCNTs-3 wt%Fe_3O_4	-	23	8–18 GHz	[152]
PVDF/PC	3 wt% MWCNTs-Fe_3O_4	0.9	32	8–18 GHz	[60]
PVDF/PC/PMMA	2 wt% MWCNTs/5 wt% BF	1	37	8–18 GHz	[153]
PVDF/PC	3 wt% MWCNTs/3 wt% Fe_3O_4	1	38	8–18 GHz	[154]
PVDF/PC	3 wt% MWCNTs/3 wt% Fe_3O_4	1	31	12–18 GHz	[155]
PVDF/PC	5 wt%MWCNTs/10 wt% manganese ferrite	-	50	12–18 GHz	[156]
PVDF/PMMA	3 wt% MWCNTs/3 wt% $NiFe_2O_4$	-	27	12–18 GHz	[157]
PVDF	10 wt% RGO-$SrFe_{12}O_{19}$	3	33	8–12 GHz	[158]
PVDF	40 wt% RGO-$SrAl_4Fe_8O_{19}$	2	42	8–12 GHz	[159]
PVDF	40 wt% RGO-$CuAl_2Fe_{10}O_{19}$	1.5	60	8–18 GHz	[160]
PVDF	10 wt% RGO/10 wt% $BaCo_2Fe_{16}O_{27}$	0.2	35.94	8–12 GHz	[161]
PVDF	10 wt% RGO/ 10 wt% $BaZrFe_{11}O_{19}$	0.2	48.59	8–12 GHz	[162]
PVDF	8 wt% MWCNT/5 wt% Fe_3O_4	1.1	32.7	18–26 GHz	[163]
PVDF	8 wt% graphene/5 wt%Fe_3O_4	1.1	35.6	18–26 GHz	[163]
PVDF/HDPE/PS	1 vol% MWCNTs/1 vol% Fe_3O_4	2.7	25	8–12 GHz	[164]
PVDF/PE	10 wt% MWCNTs/5 wt% Fe_3O_4	2.6	26.3	18–26 GHz	[165]
PVDF	30 wt% CB/40 wt% Fe_3O_4	0.37	55.3	8–12 GHz	[166]
PVDF	3 wt% FLG/15 wt% NF	-	53	1–12 GHz	[167]
PVDF	10 wt% MWCNTs/5 wt% Fe_3O_4	0.9	53	8–12 GHz	[168]
PVDF	5 wt% exfoliated graphite /85 wt% $BaFe_{11.7}Al_{0.3}O_{19}$	7	50	1–7 GHz	[169]
PVDF	30 wt% CB/40 wt% $Sr_3YCo_4O_{10+\delta}$	2.5	50.2	8–18 GHz	[170]
PVDF	40 wt% BT	1.2	9	8–12 GHz	[31]
PVDF	20 vol% BT /10 vol% Ag	1.2	26	8–12 GHz	[31]
PVDF	5 wt% NiO/20 wt% $BaTiO_3$	-	11.5	12–18 GHz	[171]
PVDF	15 wt%NiO@SiO_2	0.3	20	8–12 GHz	[76]
PVDF	2 wt% MWCNTs/2 wt% MnO_2	-	20	8–12 GHz	[172]
PVDF	25 wt% RGO@MoS_2	5	27.9	2–18 GHz	[173]
PVDF	15 wt% RGO@hollow ZnS	-	40	8–18 GHz	[174]
PVDF	30 wt% RGO@ BT	5	8	2–18 GHz	[175]
PVDF	5 wt% MWCNTs@SiO_2	0.4	24	8–12 GHz	[176]
PVDF	3 g/L graphene-MXenes	0.191	53.8	8–12 GHz	[177]
PVDF	5 wt%MWCNTs/40 wt%BN	2	8.68	8–18 GHz	[43]
PVDF	9.5 wt% graphene-SiCNWs	1.2	32.5	8–12 GHz	[62]
PVDF	2 wt%CB/8 wt%Zeolite 13X	0.08	8.4	8–18 GHz	[48]
PVDF	5 wt% MWCNTs/2.5 wt% ZnONWs	1.1	27.3	8–18 GHz	[178]
PVDF	12.5 wt% PPy-MTT	0.25	5	8–12 GHz	[179]
PVDF	25 wt% MXenes-PANI	0.012	21	8–12 GHz	[180]

Table 1 (continued)

materials		thickness (mm)	EMI SE (dB)	Frequency	Ref.
Matrix	Filler loading				
PVDF	30 wt% C-Fe_3C	2	35	2–18 GHz	[181]
PVDF	30 wt% ferrite-C_3N_4	0.175	88	8–18 GHz	[182]
PVDF	30 wt% ferrite-C_3N_4	0.19	83	8–18 GHz	[183]
PVDF	3 wt% MWCNTs/5 wt% BT-GO	-	30	8–18 GHz	[184]
PVDF	3 wt% MWCNTs/2.2 vol % CoNWs	-	35	8–18 GHz	[184]
PVDF/PC	3 wt% MWCNTs/5 wt% RGO-ferrite	5.5	38	8–18 GHz	[185]
PVDF/PC	PANI-MWCNTs-Fe_3O_4/BT	5	37	8–18 GHz	[186]
PVDF/PC	3 wt% MWCNTs-MnO_2/5 wt% RGO@ Fe_3O_4	0.9	37	8–18 GHz	[187]
PVDF/PC	3 wt% MWCNTs/5 wt% RGO@BT@Fe_3O_4	0.9	35	8–18 GHz	[188]
PVDF	5 wt% CNS@Fe_3O_4@SiO_2	-	42	8–18 GHz	[189]
PVDF	10 wt%PANI@Fe_3O_4@SWCNTs	2	29.7	12–18 GHz	[75]
PVDF	25 wt% PANI@Fe_3O_4@RGO	2	28.18	12–18 GHz	[44]
PVDF	2 wt% G-D-GQDsAg	-	46	8–12 GHz	[78]
PVDF	1 wt% MWCNTs/1 wt% RGO/1 wt% Cu	0.5	28.5	8–12 GHz	[190]
PVDF	2.75 wt% graphene-2.75 wt% Ni-2.75 wt% MWCNTs	0.6	41.8	12–18 GHz	[46]
PVDF	10 wt% RGO-Fe_3O_4/3 wt% MWCNTs	0.48	26.3	8–26 GHz	[191]
PVDF	3 wt% MWCNTs/10 wt%RGO@CuS	1	25	12–18 GHz	[55]
PVDF	3 wt% MWCNTs/6 wt% graphene/8 wt%Ni	0.3	43.7	18–26 GHz	[192]
PVDF	2.5 wt% graphene/11.5 wt% TiO_2/1 wt% MMT	-	43.7	12–18 GHz	[193]
PVDF	5 wt% Ag-5 wt% Cu@MWCNTs	0.1	26	8–12 GHz	[194]
PVDF	5 wt% Ag-5 wt% Cu@RGO	0.1	29	8–12 GHz	[194]

and metal oxide particles (e.g. nickel (Ni) [90], copper (Cu) [84], iron (Fe) [88], nickel oxide (NiO) [91] into the electrically insulating PVDF matrix have good electrical conductivity and electromagnetic shielding performance [35].

Arranz-Andrés et al. prepared PVDF/Cu [84] and PVDF/aluminum (Al) composites [85] through melt blending and hot pressing methods. They demonstrated that the reinforcement effect for metal nanoparticles in the PVDF matrix had improved the EMI SE performance of PVDF/Cu and PVDF/Al composites. Silver nanowires (AgNWs) have high aspect ratio and high conductivity, making them a research hotspot for electromagnetic interference shielding materials in recent years. Qian et al. prepared PVDF/AgNWs composites by dip-coating and spray-coating method, the three-dimensional (3D) networks and a two-dimensional (2D) layer structures were formed, respectively (Fig. 2). They explored that the 3D AgNWs networks were beneficial to multiple reflections and interface scattering of EM waves, EMI SE of dip-coated

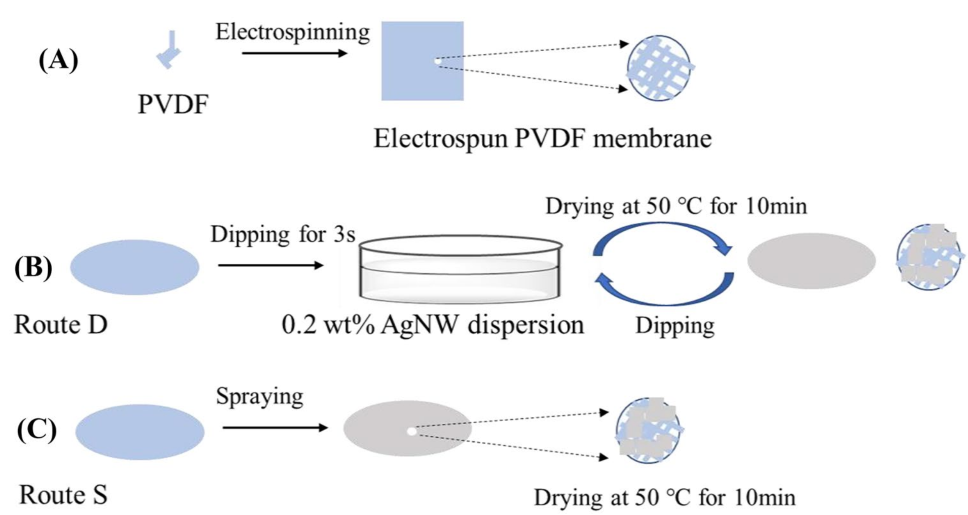

Fig. 2 Schematic process for the construction of 3D and 2D conductive networks in PVDF/AgNWs composites through dip-coating (Route D) and spray-coating (Route S) process. **A** PVDF membranes prepared by electrospinning. **B** Dip-coated PVDF/AgNWs hybrid membranes with different dip-coating cycles prepared by 0.2 wt% AgNWs dispersion. **C** Spray-coated PVDF/AgNWs hybrid membranes with the same area density of AgNWs with those prepared by dip-coating [79]

sample was higher than that of the spray-coated sample at the same area density of AgNWs [79].

It can be observed that magnetic metal particles like Fe, Co, Ni and their alloy exhibit high saturation magnetization, facilitating these particles as electromagnetic shielding materials in the gigahertz frequency region. Joseph et al. prepared PVDF/carbonyl iron (CIP) composites through solution blending and hot pressing methods. Due to conductive nature of metallic magnetic CIP, PVDF/CIP composites not only possess magnetic dipoles to produce magnetic loss and dielectric loss, but also has electrical conductivity to produce conductivity loss [88]. In Gargama's group work, they prepared PVDF/micro-size of Ni (μ-Ni) [89], PVDF/nanocrystalline Ni (n-Ni) [72] and PVDF/nanocrystalline iron (n-Fe) [35] composites through simple mechanical blending and hot pressing methods. They observed that the dielectric constant is substantially enhanced with the loading of magnetic metal particles in the PVDF matrix due to the interfacial polarization from the interface between the magnetic metal particles and PVDF, and absorption loss was the dominant mechanism of the EMI SE. What's more, Zhao et al. prepared PVDF/Ni chains composite films through the solution blending and hot pressing methods. They observed that three-dimensional conductive-magnetic Ni chains networks were formed after the addition of Ni chains in the PVDF matrix, which resulted a high EMI SE properties of composite films [47].

It is well known that usage of metal oxide nanoparticles can significantly improve the mechanical, electrical and magnetic properties of polymeric composite films without sacrificing the flexibility of the composite films. Dutta et al. fabricated PVDF/NiO composites by solution blending method. They reported that NiO nanoparticles in PVDF can improve the β phase fraction and dielectric properties, and the β phase fraction and dielectric properties of PVDF increased with the increase in NiO nanoparticles content [91]. Besides the pure metal particles, alloys also show excellent electrical conductivity and can be used as conductivity filler in PVDF composites. Low-melting-point alloys (LMPA) have attracted extensive attention due to their unique low melting point, high thermal conductivity coefficient, excellent thermal stability, and high electrical conductivity. Zhang et al. prepared PVDF/LMPA ($SnBi_{58}$) composites through solution blending and hot pressing methods. They claimed that the introduction of LMPA into PVDF resulted in a continuous LMPA network, which improved both the thermal conductivity and the EMI SE performance [18].

It can be concluded from the literatures that the composites of electrically insulated PVDF and electrically conductive metal can improve the EMI SE performance of the shielding material than that of pure metal in the high frequency band. The composites show better EMI SE performance in the whole tested electromagnetic wave frequency range, expanding the bandwidth of electromagnetic shielding, and reducing the cost of using pure metals or alloys. PVDF/metal composites can meet commercial and civil needs, and can even be used in the field of military industry. However, they also have some disadvantages. Generally, a higher content of metal fillers is always necessary to achieve good EMI SE performance, which will inevitably result in the reduction in the mechanical strength. In addition, the density of metal is high, so delamination or non-uniformity is easy to occur in the molding process, which may affect the stability of the composites.

PVDF/carbon composites

Recently, carbon materials (e.g. carbon nanofiber [92, 94, 95], carbon nanotubes [97, 105, 110, 119], graphite [101], graphene [96, 103, 104, 121], and carbon black [121] are popular fillers owing to their high aspect ratios and superior electrical properties. Therefore, carbon materials as the electrically conductive fillers added into the PVDF matrix could form a conductive network, leading to conductive loss. Interfacial polarization can be formed at the interface between the carbons and PVDF which will lead to dielectric loss. In addition, scattering and multiple reflections can also be formed due to the dielectric constant difference at the interfaces. Thus, the EMI shielding mechanism of PVDF/carbon composites is caused by the synergistic effects of conductive loss, dielectric loss, interfacial scattering and multiple scattering [112, 120].

Carbon nanofibers (CF) with high aspect ratio have advantages in conductivity and specific surface area, which facilitated well EMI SE at lower filler loading, and have attracted the attention of researchers. Lee et al. prepared PVDF/CF composites through solution blending method [92]. Naseer et al. cast PVDF/poly(methyl methacrylate) (PMMA)/CF blend solutions on the cellulosic substrates through air spray painting method [93] to obtain the composites. Yilmaz et al. prepared PVDF/CF composites through melt blending and hot pressing methods [94]. Yuksek et al. obtained PVDF/CF composite fibers through melt spinning processing method and produced conductive woven fabrics with composite fibers using handloom machines. They found that the PVDF-based composites exhibited high EMI SE performance due to good dispersion of conductive CF which has relatively large specific surface area and high aspect ratio, and the EMI SE of composites increased with increasing in CF content [92–95]. Multi-wall carbon nanotubes (MWCNTs) are regarded as prospective conductive filler for PVDF-based electromagnetic shielding materials, as their high aspect ratio, good mechanical strength and excellent electrical conductivity at low filler concentration [97]. Kim et al. prepared PVDF/poly(vinyl pyrrolidone) (PVP)/

MWCNTs composites through solution blending and coating methods [97]. Ram et al. prepared PVDF/MWCNTs composites by solution blending technique [98]. They observed that the electrical conductivity and EMI SE were significantly improved with increasing of the loading amount of MWCNTs due to its excellent conductivity and high aspect ratio [97, 98]. Graphite is also used as a conductive filler for PVDF matrix composites because it is highly conductive, economically viable, abundant in nature and light in weight [101]. Halder et al. prepared PVDF/carbonized charcoal [100] and PVDF/graphite [101] composites through solution blending and hot pressing methods. Carbon particles in the PVDF polymer layers formed a conductive network, thus exhibiting promising EMI shielding properties [100, 101]. Graphene nanosheets, as a new carbon-based nano material, has been widely studied due to its ultra-high specific surface area, excellent mechanical flexibility and electrical conductivity [103, 104]. Fan et al. prepared PVDF/graphene /nonwoven composites through a cyclic dipping-drying method. They reported that through the cyclic impregnation drying process, a large number of graphene were adsorbed on the nonwoven framework, forming an increasingly conductive network, thus the EMI SE performance was improved [103]. Additionally, they also fabricated PVDF/graphene /nonwoven composites by a coating-drying method. The coating agent with high graphene content provided good viscosity and formed a bridge between fibers, which facilitated to the formation of interconnected conductive networks, thus improving the EMI SE performance of composites. Additionally, the nonwoven which coated with PVDF/graphene twice can further improve the loading amount of graphene and a 2-side coated nonwoven had a better EMI SE than a 2-layer coated nonwoven [104].

The composites of PVDF and carbon fillers can improve the mechanical properties of pure carbon electromagnetic shielding materials. With a low loading of carbons, materials suitable for electromagnetic shielding applications can be obtained, and the cost will also be reduced compared with pure carbon nano shielding materials. Moreover, PVDF/carbon composites have good film-forming and processing properties, which is help to the practical applications. However, compared with PVDF/metal composites, exploitation of high-performance PVDF/carbon composites remains a great challenge.

PVDF/conductive polymer-filled composites

Conductive polymers (e.g., polyaniline (PANI), poly (3,4-ethylenedioxythiophene) (PEDOT), polypyrrole (PPy) show good conductive properties. However, the poor mechanical and processing properties have limited their applications in electromagnetic shielding. By blending conductive polymer materials with PVDF, electromagnetic shielding materials with excellent mechanical properties and EMI SE performance can be obtained, due to the effects of conductive loss of conductive polymer and high mechanical property of PVDF. Pontes et al. prepared PVDF/PANI composites by two procedures involving the in situ polymerization of aniline in the presence of PVDF and powder blending process using ball milling, followed by hot pressing method. They reported that the solvent-free powder blending approach using ball milling offered better application advantages and this process resulted a higher EMI SE than in situ polymerization [21]. Lee et al. prepared core-shell PVDF/PEDOT nanofiber composites with a conductive shell through the in situ polymerization and electrospunning method. They observed that the shell-conductive nanofiber exhibited high EMI SE performance and mechanical properties by incorporating low-density PEDOT. This was due to the enhanced conduction loss and multiple reflections of the incident EM waves caused by conductive shell and porous structure of the nanofiber composites, respectively, and absorption loss is the main shielding mechanism [22].

The PVDF/conductive polymer composites can not only enhance the electromagnetic shielding performance, but also improve the mechanical properties by interacting with each other to, to give shielding materials with good comprehensive performance. However, the PVDF/conductive polymer-filled composites with high loading content will increase the cost. Thus, it is necessary to modify the conductive filler and develop a new process to reduce the amount of filler while maintaining the conductive performance of composite materials.

PVDF/other conductive-filled composites

MXenes are the family of two-dimensional (2D) transition metal carbides and/or nitrides with a formula of $M_{n+1}X_nT_x$ [5, 6, 195], where M, X and T_x represent transition metal, carbon and/or nitrogen and the functional surface terminations (such as -O, -F, -OH), respectively [196, 197]. In recent years, MXenes have attracted extensive attention from researchers in the field of electromagnetic shielding for their excellent conductivity, abundant surface heteroatoms and specific 2D characteristics like graphene [8, 198]. However, as the poor mechanical properties and oxidation capacity of pure MXenes would significantly limit their applications in wearable electromagnetic shielding [71, 199], blending MXenes with polymer such as PVDF to prepare polymer-based conductive composites is a promising strategy to obtain better EMI shielding materials [124, 125].

$Ti_3C_2T_x$ [6, 16, 200] is a representative type of MXenes, which can be used to prepared conductive composites with good EMI SE performance by blending it with PVDF. Li et al. prepared the flexible and durable PVDF/$Ti_3C_2T_x$ MXenes composites with compact hierarchical brick-and-mortar structure through solution blending and blade coating methods. They claimed that the highly aligned MXenes nanosheets incorporated in PVDF matrix significantly improved the electrical conductivity and EMI SE performance of composites (Fig. 3). Also, due to the surface plasmon resonance and high electrical conductivity, the composites showed photo/electro-thermal heating abilities with rapid response to time, high stability and controllability, enabling their application in special conditions [124]. Wang et al. prepared co-continuous PVDF/polystyrene (PS)/$Ti_3C_2T_x$ composites through solution blending or hot pressing method. They found that PVDF/PS/$Ti_3C_2T_x$ MXenes obtained by solution blending method had layered double-percolated structures which were beneficial to form conductive network and imporved the multiple radiation of electromagnetic wave inside the composites to elevate EMI SE performance. Additionally, due to the layered double-percolated structures, the composites showed better EMI SE performance under low filler content [125].

The combination of PVDF and MXenes with excellent electrical conductivity can give high-performance shielding materials with good mechanical properties. However, due to the high price of MXenes, though the composites with high loading of MXenes (> 50%) have good shielding performance, the high cost would limit their production and application. Therefore, it is necessary to explore better preparation process and develop PVDF/MXenes composites with high performance and low cost [125].

PVDF/pure magnetic-filler composites

It is well known that magnetic fillers such as ferrites [126] (e.g., Fe_3O_4 [163, 164], M-type barium hexaferrite [26], U-type hexaferrite [81]) have good ferromagnetic properties,

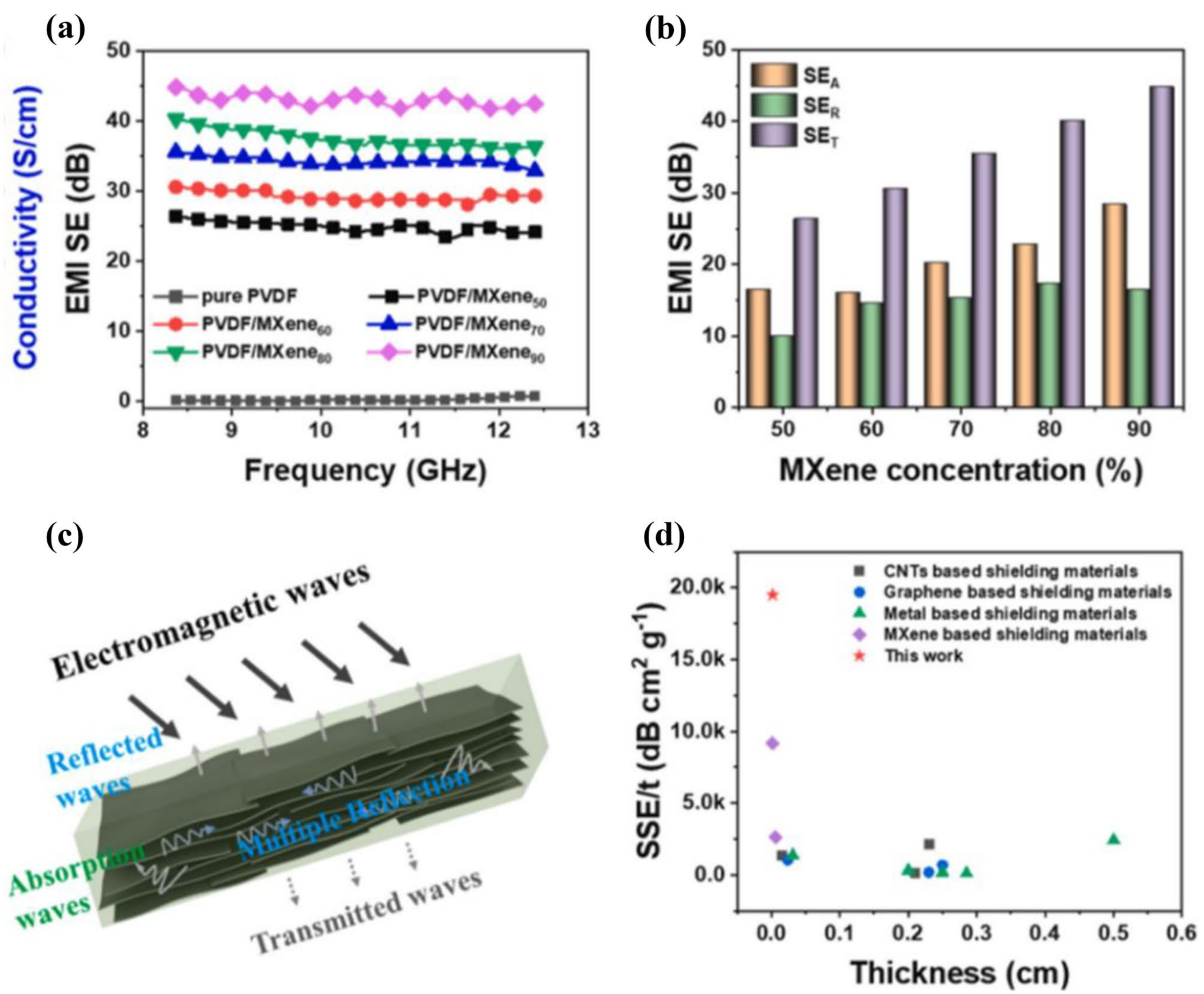

Fig. 3 **a** EMI shielding performance at X band, **b** average SE_T, SE_A and SE_R. **c** Schematic illustration of EM wave transfer in PVDF/MXenes composites. **d** Comparison of SSE/t as a function of thickness with previous reports [124]

and possess high permeability and a certain permittivity, enabling them suitable for design high-performance EMI SE materials. Therefore, introducing ferrite particles fillers into PVDF matrix could help to increase the EMI SE performance of PVDF-based composites, which is attributed to the good dielectric and magnetic loss ability of ferrite particles. Hence, PVDF-based magnetic composites with high EMI SE performance have been widely reported in recent years.

Revathi et al. prepared PVDF/barium-substituted magnesium ferrite ($Ba_{0.2}Mg_{0.8}Fe_2O_4$) composite fibers through electrospinning method. Barium magnesium ferrites were added to PVDF matrix to significantly improve the magnetic property of the PVDF-based composites, and electrospinning could improve the ferroelectric properties of PVDF, which resulted in a good EMI SE performance due to the dielectric loss and magnetic loss [126]. In Sutradhar's work, they reported the preparation of PVDF/M-type barium hexaferrite ($BaFe_{12}O_{19}$) (BaH) nanocomposites [26] and PVDF/Co_2U-type hexaferrite ($Ba_4Co_2Fe_{36}O_{60}$) nanocomposites [81] through solution blending method. Incorporation of hexaferrite in PVDF matrix have significantly improved the relative permeability and permittivity, and enhanced electroactive β phase of PVDF. This was beneficial to improve the dielectric loss of nanocomposites, resulting in a higher EMI SE performance [26, 81]. In addition, Darwish et al. prepared PVDF/$BaFe_{11.7}Al_{0.3}O_{19}$ (HF) composites [25] through mechanical blending and hot pressing method; Halder et al. prepared PVDF/copper ferrite ($CuFe_2O_4$) composites through resolution blending method [30]. They also observed that these PVDF/ferrite composites could control the EMI SE efficiency by tuning their magnetic properties without considering the conductivity values, so as to suppress electromagnetic interference [25, 30].

PVDF/pure dielectric-filler composites

Beside conductive and magnetic materials, some dielectric materials (such as barium carbonate ($BaTiO_3$), strontium titanate ($SrTiO_3$) [74], silicon carbide nanowires (SiC) [62], boron nitride (BN) [43, 82], copper sulfide (CuS) [23], zinc oxide (ZnO) [24]) possess high dielectric constant and high dielectric loss, are promising candidates for electromagnetic shielding materials. Many researchers have been studied on the development of PVDF-based dielectric-filler composites with high EMI SE performance. Dielectric properties of dielectric-filler and the interfacial polarization between the PVDF and fillers are thought to contribute significantly to the EMI SE performances. Studies have also shown that these composites can be used under harsh conditions because of their high corrosion resistance, electrical insulation, and superior thermal stability.

Aepuru et al. prepared PVDF/Flower-like radial ZnO (RZnO) composites through solution blending method. Introduction of RZnO into PVDF matrix significantly improved the dielectric property of the PVDF/RZnO nanocomposites and the dielectric constant increased with increasing of RZnO content, resulting in the improvement in the EMI SE performance of composites. The domination shielding mechanism was absorption loss [24]. Sankaran et al. obtained PVDF/hexagonal BN nanoparticles (h-BNNPs) composites through solution blending method. They observed that h-BNNPs was uniformly distributed in the PVDF matrix leading to improvement in the dc electrical conductivity and EMI SE performance. The dc electrical conductivity increased with increasing in h-BNNPs loadings and the domination shielding mechanism was absorption loss [82]. Biswas et al. reported the PVDF/'wool-ball' like hollow copper sulfide (CuS) composites obtained through solution blending and hot pressing methods. They claimed that PVDF/CuS composites possessed a high EMI SE performance due to dielectric heating and polarization loss. The composites also exhibited good thermal energy dissipation at a certain time frame due to the good thermal conductivity of CuS [23]. What's more, Peymanfar et al. prepared PVDF/$CuCo_2S_4$ composites through solution blending and hot pressing methods. They found that the increase in size of $CuCo_2S_4$ enhanced the complex permittivity and magnetic loss, resulting in improvement in EMI SE performance of composites [83]. Joseph et al. obtained PVDF/polyvinyl chloride (PVC)/strontium titanate ($SrTiO_3$) nanocomposite films through solution blending method. They reported $SrTiO_3$ uniformly distributed in the matrix which enhanced the dielectric constant and dielectric loss of composites[[[[74]].

PVDF/multiple-component filler composites

As we all know, the excellent EMI SE performance of shielding materials is usually determined by the high conductive loss, dielectric loss and magnetic loss [184, 187]. In spite of extensive research efforts, single-component fillers such as conductive, dielectric or magnetic materials, with high EMI SE performance under low loading of fillers and minimum thickness of films still remains a challenge [86, 130]. This is mainly because of the difficulty in obtaining high EMI SE performance by a single shielding mechanism. Without losing processability, the shielding performance can be significantly improved by the introduction of multiple-component fillers. This involves the design and formation of shielding interfaces of conductive, magnetic and/or dielectric hybrid materials to alleviate harmful impact of EM waves [187, 188]. The system includes binary-component and multi-component fillers, i.e. Conductive filler, dielectric filler or magnetic filler are added to the PVDF matrix in coupled or multiple combinations, to form a heterogeneous system that maintains its essential properties, which forming a large interface that generates multi-interfacial polarization and improves the dielectric loss, thus further enhancing the EMI SE performance [60, 76, 117, 138, 180, 182].

PVDF/binary conductive-filler composites

The conductivity of PVDF can be improved by adding large amounts of conductive fillers, however, higher loadings degrade the flexibility, increase the product weight and result in poor processability. The designed binary conductive fillers not only promote the uniform dispersion of each fillers in PVDF matrix and lower percolation threshold [144, 201], but also is more effective in improving the electrical conductivity of PVDF-based composites than a single conductive-filler [14, 38, 147].

PVDF/metal-carbon composites In Dinakaran's group, they prepared PVDF/Ag–Graphite[[[[128] composites and PVDF/Au-MWCNTs [86] composites through solution blending method. They observed that compared to single carbon fillers, the presence of metal fillers significantly elevated the dielectric constant and lowers the dielectric loss of composites [86, 128]. Sang et al. introduced MWCNTs and AgNWs into PVDF casted commercial nonwoven fabrics (NWF) to obtain PVDF/MWCNTs/AgNWs/NWF composites (Fig. 4). They observed that the synergetic effects between the AgNWs and MWCNTs networks yielded the ideal electrical conductivity and mechanical strength [37]. Lakshmi et al. synthesized graphene quantum dots decorated graphene (G-D-GQDs) and G-D-GQDs that decorated with conducting Ag nanoparticles (G-D-GQDsAg) by a solvothermal method, and prepared PVDF/G-D-GQDs and PVDF/G-D-GQDsAg composites. They found that the latter showed better EMI SE performance than that of the former, due to the presence of Ag nanoparticles that increased the electrical conductivity [78]. Ertekin et al. added Ag nanoparticles into PVDF/carbon black (CB) and PVDF/graphite composites. They also found that the EMI SE performance was significantly improved due to the synergetic effects of metal particles and carbons [140].

Materials with high permeability and high electrical conductivity always exhibit good EMI shielding properties. Therefore, high-magnetic metal particles together with high-electrical conductivity carbon materials being added to PVDF matrix can yield good electromagnetic shielding materials. In Bose's group, they prepared PVDF/Fe@CF [129], PVDF/MWCNTs/CoNi [130, 131], PVDF/MWCNTs/NiFe [132], PVDF/styrene maleic anhydride (SMA)/MWCNTs/NiFe [38], and PVDF/thermoplastic polyurethane (TPU)/ MWCNTs@NiFe [133] composites through solution blending and hot pressing method. They concluded that the incorporation of magnetic metal particles or alloys in PVDF/MWCNTs composites not only improved the electrical conductivity, but also enhanced the magnetic and dielectric properties, resulting in significantly improvement in absorption-dominated EMI SE performance of composites. What's more, Li et al. reported the preparation of anisotropy-shaped Co (MWCNTs@Co) decorated PVDF/MWCNTs composites (PVDF/MWCNTs@Co) through solution blending and hot pressing methods. They claimed that the presence of MWCNTs@Co significantly improved the EMI SE performance due to the high conductivity (from MWCNTs and Co), magnetic loss (from Co), interfacial polarization and multiple reflections and scattering of EM waves (from MWCNTs, Co and PVDF) [134]. Wang et al. prepared PVDF/CB/carbonyl iron (CI) composites [135], Kumar et al. prepared PVDF/FeC composites[[[[73], Zhao et al. prepared PVDF/carbon (MWCNTs or graphene)/Ni chains composites [45] and Liang et al. prepared PVDF/graphene/Ni chains composites [14] through solution blending and hot pressing

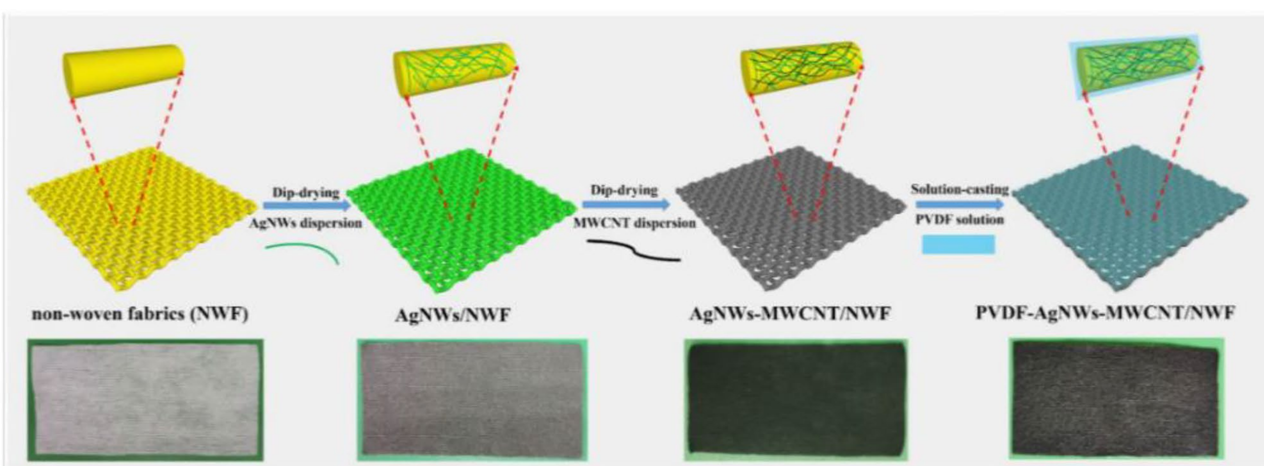

Fig. 4 Schematic illustration of fabrication steps of the as-prepared PVDF/ MWCNTs/AgNWs /NWF composites [37]

methods. Peymanfar et al. synthesized wrinkled Ni@grapy carbon microsphere nanosheet (CMS) particles (Ni@CMS) via co-precipitation and hydrothermal complementary method, and PVDF/Ni@CMS composites through solution blending and hot pressing method [139]. Both works identified that combining carbon materials with magnetic metal particles resulted in good absorption-dominated EMI SE performance [14, 45, 73, 135, 139].

PVDF/binary carbon-filled composites Song et al. prepared PVDF/poly(ethylene terephthalateco-1,4-cylclohexylenedimethylene terephthalate) (PETG)/CF/CB composites [143], Halder et al. prepared PVDF/MWCNTs/graphite composites [145], and Leão et al. prepared clean polyvinylidene fluoride scrap (PVDF)/CB/expanded graphite composites [148] through melt blending and hot pressing methods. Zhao et al. prepared PVDF/MWCNTs/graphene composites through solution blending and hot pressing methods [144]. They found that compared to single-component carbon fillers, the addition of binary carbon-fillers in the PVDF matrix exhibited higher conductivity and EMI SE performance due to the synergistic effect between the binary carbon-fillers, and an increase in EMI SE performance with increasing the amount of carbon fillers was observed [143–145, 148]. Additionally, Mei et al. prepared a flexible NWF which consisted of CF and polypropylene/polyethylene (PP/PE) core/sheath bicomponent fibers, and immersed them into the PVDF/ graphene solution to fabricate PVDF/graphene/NWF composites. The PVDF/graphene/NWF composites exhibited high EMI SE performance due to the formation of 3D conducting network in the present of a high percentage of graphene, and the synergistic effect between NWF and graphene [147].

PVDF/conductive filler-MXenes composites PVDF/MXenes composites exhibit high electrical conductivities and EMI SE performances under a high MXenes loading (≥ 50 wt%) [124]. However, the exorbitant loading may cause the poor mechanical and processing properties, which would seriously hinder their application in flexible devices. Some studies have shown that combination of MXenes with conductive fillers can improve the high EMI SE performance under lower loading of Mxenes.

Wang et al. and Cheng et al. prepared PVDF/MXenes/Ni chains composites [36] and PVDF/MXenes/AgNWs composites [141], respectively, through solution blending method. They observed that compare to those only using MXenes or metal particle fillers, PVDF/MXenes/metal composites exhibited higher EMI SE. A low loading amount of MXenes or metal particles (not exceeding 10%) gave maximum EMI SE values in X band, due to the synergistic effects between high conductivity of MXenes and high conductivity and aspect ratio of metal particles. Additionally, Yang et al. prepared sandwich-sturctured PVDF/MXenes/AgNWs composites by hot pressing method, with the electrospun PVDF film as the battom layer, vacuum-filtrated AgNWs as a conductive inner layer, and filtrated MXenes as top layer. They observed that except for the establishment of conductive AgNWs pathways, the introduction of MXenes further improved the electrical conductivity and multiple reflection between neighboring conductive layers, resulting in an outstanding EMI SE performance at a low fillers content [142].

Besides metal particles, carbons or polymer conductive fillers combining with MXenes could also improve the EMI SE performance of PVDF-based composites. Raagulan et al. perpared PVDF/graphene-MXenes composites through low-cost spray coating and solvent blending methods. They found that PVDF/graphene-MXenes composites showed high conductive properties and excellent EMI SE performance [177]. Xu et al. synthesized MXenes-PANI composites by in situ polymerization and added them into the PVDF matrix to obtained a porous structure PVDF/MXenes-PANI composites through solution blending method. Then Au was evaporated on the surface of PVDF/MXenes-PANI to gave PVDF/MXenes-PANI/Au (MPPA) composites, and MPPA composites as the top electrode to prepared of the functional device. Due to the highly efficient conductivity of porous structure MPPA composites, the device exhibited a superior EMI SE performance [180].

PVDF/conductive–magnetic filler composites

As mentioned above, PVDF/magnetic-filler composites have poor conductivity loss due to the relatively poor electrical conductivity performances of magnetic ferrite particles [73], thus it is difficult to obtain high electromagnetic shielding performance. On the other hand, PVDF/conductive-filler composites with good conductivity [79, 120] makes the dielectric constant different on the interface between the composites and free space, resulting in the poor impedance matching, which easily leads to the reflection of the incoming electromagnetic wave on the surface of composites and cause a secondary pollution in free space [202]. In order to address the above problems, combination of conductive-filler with magnetic-filler is an effective strategy to improve the absorption-dominated EMI SE performance [60]. Actually, incorporation of conductive-filler and magnetic-filler in PVDF-based composites can induce multi-interfacial polarization to improve the dielectric loss as well as the conductivity and magnetic properties [163].

Acharya et al. synthesized reduced graphene oxide (RGO)-strontium ferrite ($SrFe_{12}O_{19}$) (SF), RGO-strontium aluminium ferrite ($SrAl_4Fe_8O_{19}$) (SAF) and copper aluminum ferrite ($CuAl_2Fe_{10}O_{19}$) (CFA) particles by one pot chemical reduction method, and prepared PVDF/RGO-SF [158], PVDF/RGO-SAF [159] and PVDF/RGO-CAF

[160] composites through solution blending and hot pressing method. Cheng et al. prepared PVDF/carbon (MWCNTs or graphene)/Fe_3O_4 composites [163], Lalan et al. prepared PVDF/CB/Fe_3O_4 composites [166], Ramazanov et al. prepared PVDF/MWCNTs/Fe_3O_4 composites [168], and Darwish et al. prepared PVDF/exfoliated graphite/HF composites [169] through solution blending and hot pressing method. Anand et al. prepared PVDF/RGO/barium hexaferrite ($BaCo_2Fe_{16}O_{27}$) [161] and PVDF/RGO/Zr doped barium hexaferrite ($BaZrFe_{11}O_{19}$) composites [162], Ahmed et al. prepared PVDF/few layered graphene (FLG)/CF composites [167] through solution blending method. They reported that carbon and ferrite in PVDF matrix formed network structure through their effective interaction which increased the conductive loss and magnetic loss. In addition, a large number of capacitive regions are accumulated on the interfaces formed by carbon and ferrite, resulting in high dielectric loss, thus endowing the excellent absorption-dominated EMI SE performance of the composites [158–163, 166–169]. Li et al. prepared PVDF/high density polyethylene (HDPE)/PS/MWCNTs/Fe_3O_4 composites by melt blending and hot pressing methods, in which HDPE and PS displayed a core-shell structure. By changing the order of adding materials during melt blending, can tailor Fe_3O_4 in PVDF matrix or HDPE core, and led MWCNTs selectively localized in PS phase. They found that with the incorporation of Fe_3O_4 and MWCNTs, the EMI SE performance was enhanced, while by incorporating Fe_3O_4 in the HDPE core, this composites exhibited higher EMI SE performance [164]. Zhao et al. prepared PVDF/polyethylene (PE)/MWCNTs/Fe_3O_4 composites through melt blending, hot pressing and supercritical CO_2 foaming methods. The EMI SE of composites was significantly enhanced by virtue of the selective localization of MWCNTs and Fe_3O_4 as well as directionally arrangement of MWCNTs in the cell wall during foaming process. The composites possessed a high electric conductivity and magnetic permeability, and the special honeycomb internal structure caused multiple reflections [165]. Lalan et al. prepared PVDF/CB/room-temperature ferromagnetic $Sr_3YCo_4O_{10+\delta}$(SYCO) composites through solution blending and coagulation methods. They observed that the addition of CB and SYCO facilitated the improvement of electrical conductivity and increased the β phase of PVDF, which also significantly enhanced the permittivity and permeability of composites, resulting in the highest absorption-dominated EMI SE performance reported so far [170]. What's more, Gao et al. synthesized a two-dimensional C-Fe_3C nanoparticles which possess dielectric and magnetic properties, and obtained PVDF/C-Fe_3C composites through solution blending and hot pressing methods. They reported that integrating C-Fe_3C nanoparticles into PVDF matrix effectively improves the dielectric and magnetic properties of the composites resulting in excellent EMI SE performance [181].

PVDF/conductive-dielectric filler composites

Although dielectric fillers such as ceramics have a high dielectric constant, the effective dielectric constants of PVDF/ferroelectric ceramic composites remain low [74, 82]. This is mainly caused by the low dielectric constant of PVDF, and the difficulty in obtaining composites with more than 50% ceramics. In this regard, incorporation of both conductive and dielectric fillers into PVDF matrix may be a promising method to improve EMI SE performance, because the present of both conductive and dielectric fillers in PVDF matrix not only improve the conductivity and dielectric properties, but may also enhance the β phase of PVDF which could lead to the better dielectric properties of the composite [31, 171].

PVDF/metal–ceramic composites Joseph et al. added Ag particles into PVDF/$BaTiO_3$ (BT) composites and obtained PVDF/Ag/BT composites through solution blending and hot pressing methods. Compare to BT single filler, the incorporation of a small amount the Ag significantly improved the conductivity of PVDF/Ag/BT composites, leading to a better EMI SE properties [31]. Muzaffar et al. prepared PVDF/NiO/BT composites through solution blending method. They found that NiO and BT particles in PVDF matrix formed a conductive network and interacting surfaces which enhanced the EMI SE properties of the composites [171]. Dutta et al. synthesized NiO@SiO_2 particles and added them into PVDF matrix to obtain PVDF/NiO@SiO_2 composites through solution blending method. They claimed that the presence of NiO@SiO_2 particles in PVDF matrix led to the increment of the effective surface area and the interfaces, resulting in enhancement in the dielectric and conductivity properties of the composites [76].

PVDF/carbon–ceramic composites Eswaraiah et al. prepared PVDF/MWCNTs/MnO_2 composites [172], Guo et al. obtained PVDF/RGO@BT composites [175], Zeraati et al. prepared PVDF/MWCNTs/ZnO nanowire composites [178], respectively, through solution blending and hot pressing methods. The preparation of PVDF/RGO@MoS_2 composites [173], PVDF/RGO@hollow ZnS composites [174], PVDF/MWCNTs@SiO_2 composites [176], PVDF/CB/Zeolite 13X composites [48] through a solution blending method were also reported. They found that carbons and ceramics in the PVDF matrix contributed to the improvement in EMI SE performance of composites due to the interfacial polarization (from carbons, ceramics and PVDF), high conductivity (from carbons) and good dielectric (from ceramics) properties. Additionally, special geometry structures of carbons or ceramics would further improve the EMI SE performance of composites. For example, the hollow ZnS particles and ZnO nanowire with high aspect ratios are helpful to enhance multiple reflections of the electromagnetic wave and interfacial polarization, leading to high EMI SE performance of composites. Zhang et al. synthesized PVDF@MWCNTs microspheres and blended them with BN particles to give segregated PVDF@MWCNTs/BN composites through melt blending and hot pressing methods. They reported that MWCNTs in the segregated composites formed an electrical conductive micro-network, and BN micro-network provided thermal conductivity and dielectric properties, leading to the improvement of permittivity of composites [43]. Liang et al. synthesized poly (diallyldimethylammonium chloride) (PDDA) modified SiC nanowires (SiCNWs) and mixed them with PVDF/graphene solutions via electrostatic assembly followed by hot-pressing to obtain PVDF/graphene-SiCNWs composites. They observed that SiCNWs was located between the graphene nanosheets by self-assembly. This was beneficial to the dispersion of graphene, interfacial polarization as well as multiple reflections, resulting in the improvement of dielectric properties and EMI SE performance of composites [62].

PVDF/conductive polymer–ceramic composites It has reported that introduction of layered inorganic guest materials into conductive polymers result in high electrical conductivity, large surface areas and improved dielectric properties of the composites [179]. Schiefferdecker et al. using in situ polymerization synthesized polypyrrole (PPy)-montmorillonite (MTT) particles and added into the PVDF matrix to obtain PVDF/PPy-MTT composites through electrospun method. They reported that the formation of a conducting network (from PPy) and dielectric properties (from MTT) of composites endowing superior EMI SE performance. The maximum EMI SE of 5 dB was achieved in X band for the PVDF/12.5 wt% PPy-MTT composites [179].

PVDF/ferrite–ceramic composites

In this direction, many articles have been published in recent time where magnetic fillers and dielectric materials have been considered inside the matrix of PVDF for the co-modulation of magnetic and dielectric properties of PVDF-based composites [182]. In Sutradhar's group, they synthesized X-type hexaferrite ($Ba_2Co_2Fe_{28}O_{46}$)-C_3N_4 and Ni-Zn-Cu-ferrite ($Ni_{0.50}Zn_{0.30}Cu_{0.20}Fe_2O_4$)-$C_3N_4$ binary filler by the solid-state reaction method, and obtained PVDF/X-type hexaferrite-C_3N_4 [182] and PVDF/Ni-Zn-Cu-ferrite-C_3N_4 [183] composites through solution blending method. They found that the loading of magnetic–ceramic binary fillers endowing the enhancement of interfacial polarization and the dielectric properties (from C_3N_4) and the magnetic properties (from ferrite) of composits [182, 183].

PVDF/multi-component filler composites

As mentioned, the efficiency of EMI shielding materials are determined by their high electrical conductivity, large dielectric and magnetic loss. Thus, some studies have shown that a careful selection of multi-component fillers, which with conductivity, dielectric or magnetic properties are helpful to design enhanced EMI SE materials.

Sharma et al. obtained PVDF/MWCNTs/RGO@CuS flower composites through melt blending and hot pressing methods [55]. Zeng et al. prepared PVDF/MWCNTs/Ni@MWCNTs composites through solution blending and hot pressing methods [138]. The preparation of PVDF/MWCNTs/RGO/metals (Ag, Au, Cu) composites [190], PVDF/Ag-Cu@MWCNT or RGO composites [194] and PVDF/MWCNTs/graphene/Ni composites through solution blending method [192]. They observed that the incorporation of mutli-component conductive fillers in the PVDF matrix contributed to the improvement in EMI SE performance of composites, resulting from the synergistic between the multiple phase conductive fillers [138, 167, 190, 192, 194]. Moreover, The preparation of PVDF/MWCNTs/BT-GO composites [184], PVDF/MWCNTs/RGO-$MnFe_2O_4$ composites [185], PVDF/PANI@Fe_3O_4@SWCNTs [75], PVDF/PANI@Fe_3O_4@RGO composites [44], and PVDF/HFP/MWCNTs/Fe_3O_4/ionic liquid (IL) composites [137] through solution blending method. Peymanfar et al. obtained PVDF/$La_{0.8}Sr_{0.2}MnO_3$/La/Sr nanocomposites through solution blending and hot pressing methods [127]. Both works identified that the binary component conductive fillers combined with either dielectric fillers or magnetic fillers can produce higher EMI SE preformance [44, 75, 127, 184, 185]. Kumar et al. prepared PVDF/graphene/TiO_2/MTT composites through solution blending method. They reported that conductive filler combined with binary-component dielectric fillers are embedded into the PVDF matrix, which will be helpful to obtain an good EMI shielding materials [193].

What's more, Biswas et al. prepared PVDF/MWCNTs/RGO@BT@Fe_3O through melt blending method [189], Bhattacharjee et al. prepared PVDF/MWCNTs/carbon nanosphere (CNS)@Fe_3O_4@SiO_2 composites through solution blending and hot pressing methods. They also found that in addition to excellent conductivity (from MWCNTs, RGO, CNS), dielectric (from BT, SiO_2) and magnetic property (from Fe_3O), RGO@BT@Fe_3O_4 and CNS@Fe_3O_4@SiO_2 particles possess good heterogeneous boundaries with multiple scattering and interfacial polarization, resulting in exhibiting excellent EMI SE performance of composites.

Influence factors on the EMI SE performance of PVDF-based composites

According to theory of electromagnetic wave shielding effectiveness, many factors influence the shielding effectiveness of the material, containing electrical conductivity, magnetic conductivity, dielectric constant, thickness and structure of the material, etc. Thus, the factors affecting the shielding performance of PVDF-based composites mainly containing filler (such as content, type, morphology, size, synthesis conditions, dispersion state in the PVDF matrix), the β phase fraction and content of PVDF, thickness and structure of composites.

The filler factors

Type and content of filler

As an electrical insulator, PVDF has a poor EMI SE performance, incorporating conductive, dielectric or magnetic fillers in the PVDF matrix will obtains different EMI SE performance. Sabira et al. obtained PVDF/graphene composites, the maximum EMI SE of 47 dB in X band [52]. Meher et al. prepared PVDF/PANI composites, the maximum EMI SE of 65 dB in X band [80]. Sutradhar et al. prepared PVDF/Co_2U-type hexaferrite composites, the maximum EMI SE of 60 dB in X band [81]. Zhang et al. prepared PVDF/Ni chains composite, the maximum EMI SE of 26.8 dB in X band [63]. Aepuru et al. prepared PVDF/RZnO composites, the maximum EMI SE of 8 dB in X band [24].

In addition, some studies show that the addition of more component fillers in PVDF matrix exhibited higher EMI SE performance. Liang et al. prepared PVDF/graphene/Ni chains composites, the maximum EMI SE of 51.4 dB in X band [14]. Zeng et al. obtained PVDF/MWCNTs/Ni@MWCNTs composites, the maximum EMI SE of 51.4 dB in X band [138]. What's more, Biswas et al. studies the effect of the addition of different magnetic phase (here inverse-spinel ferrites, MFe_2O_4 (M = Fe, Co, Ni)) on EMI SE performance of PVDF/PC/MWCNTs composites. They found that Fe_3O_4 particles possess high magnetic properties than $CoFe_2O_4$ and $NiFe_2O_4$, have more helpful to improve the absorption-dominated EMI SE performance of composites [155]. This indicated that the careful selective of fillers type plays a crucial role in determining product performance.

It's worth noting that the content of fillers also affects the EMI SE performance of PVDF-based composites. Zhao et al. prepared PVDF/Ni chains composites, they found that when the Ni content increased from 3 wt% to 6 wt%, the σ_{DC} electrical conductivity was increased by 9 orders of magnitude resulting from the conductive network of Ni chains had formed within the PVDF matrix with the 6 wt% Ni content, and with a further increase in the Ni chains content (12 wt%), the composite's electrical conductivity of correspondingly was marginally increased. Moreover, the increase in the interface areas as the increased Ni chains content, led to the formation of greater amounts of equivalent micro-capacitors in the PVDF composites, resulting in a higher permittivity. Thus, with a high concentration of Ni (12 wt%), the higher EMI SE performance emerged [47]. Sutradhar et al. prepared PVDF/BH composites, they observed that 20 wt% of BH loaded PVDF-based composite shows better EMI SE performance than 10 wt% of BH, due to higher concentration of magnetic fillers leads to excellent permeability, permittivity, and dielectric loss [26]. Aepuru et al. prepared PVDF/RZnO composites, they also reported that as the increase in RZnO percentage, leads to higher permittivity, resulting in improvement on the EMI SE efficiency of PVDF/RZnO composites [24]. Therefore, in practical application, it is necessary to comprehensively consider the conductivity, mechanical properties and EMI SE performance of shielding materials, to determine the type and optimal dosage of fillers.

Morphology and size of fillers

The EMI SE performance of the composites also depending on the morphology and size of the introduced fillers. Arief et al. prepared PVDF/MWCNTs composites with micro flowers rods, and microspheres structure Co-Ni particles, they found that due to the higher surface roughness of Co-Ni micro flower have good dispersibility and uniformity in the polymer matrix, PVDF/MWCNTs/micro flower structure Co-Ni composites shown the best EMI SE performance[[[[79]. Li et al. prepared PVDF/ MWCNTs@Co flowers or Co chains composites, they observed that with the same Co content, the EMI SE performance of PVDF/MWCNTs@Co chains is superior to that of PVDF/MWCNTs@Co flowers, resulting from the unique dimensional chain-like structure of Co chains, which was beneficial for improves conductive interface between MWCNTs and PVDF and orientation polarization [134]. Zeraati et al. prepared PVDF/MWCNTs/metal nanoparticles composites, they investigated the effect of geometry of metal nanoparticles such as nickel nanowires (NiNWs), AgNWs, nickel nanoparticles (NiNPs), and silver nanoparticles (AgNPs) on EMI SE performance of composites [136]. They found that the metal nanoparticles added into the PVDF/MWCNTs blends leads to the improvement of dispersions state of MWCNTs, and the high aspect ratio of nanowires of metal nanoparticles was beneficial to formed a strong interconnected conductive network, resulting in higher EMI SE performance. Carbons with high aspect ratio have advantages in both electrical conductivity and specific surface area, so the influence of the aspect ratio of carbon on EMI SE performance cannot be ignored. Lee et al. prepared PVDF/CF composites, and they reported that the EMI SE performance decreased with the decrease in the filler's aspect ratio [92]. Song et al. prepared

PVDF/poly(ethylene terephthalateco-1,4-cylclohexylenedimethylene terephthalate) (PETG)/CF/CB composites, they observed that PVDF/PETG/long CF/CB composites exhibited better EMI SE performance than PVDF/PETG/short CF/CB composites, due to the better synergistic effect between long CF and CB [143].

Additionally, the size of fillers also could can affect the EMI SE performance of PVDF-based composites. Biswas et al. prepared PVDF/PC/MWCNTs/Fe_3O_4 composites, and studied the effect of shape (like spherical, cubic, cluster, flower) and size (15 nm, 25 nm,75 nm, 100 nm, 150 nm) of the Fe_3O_4 nanoparticles on the EMI SE performance of composites. They reported that PVDF/PC/MWCNTs composites with spherical shaped Fe_3O_4 (15 nm) nanoparticles exhibits excellent EMI SE, due to the smaller size spherical shaped Fe_3O_4 nanoparticles provides excellent dispersibility in PVDF matrix [154]. Gargama et al. prepared PVDF/μ-Ni composites [89] and PVDF/n-Ni composites [35], they found that the maximum EMI SE of 23 dB and 42.87 dB was achieved in X band for the PVDF/40 vol% μ-Ni and PVDF/35 vol% n- Ni composites respectively, while the thickness of composites was 1.95 mm. Ram et al. obtained PVDF/particulate nano carbon composites, they reported that particulate nano carbon filler which with smallest particle size and the highest surface area incorporated into PVDF matrix, resulting in highest conductivity and EMI SE performance [102]. Moreover, Joseph et al. prepared PVDF/micron sized BT composites and PVDF/nano sized BT composites, they found that the PVDF/ nano sized BT composites exhibits better dielectric properties and EMI SE performance, due to the small size of nanoparticles was beneficial to the increase of the number of dipoles and the effective dispersion of BT in the PVDF matrix [31]. Interesting, Peymanfar et al. prepared PVDF/$CuCo_2S_4$ composites, they found that the largest particle size of $CuCo_2S_4$ showed better EMI SE performance, due to its high complex permittivity, magnetic loss, and impedance [83]. The above research indicated that the fillers with different morphologies can exhibit exciting behavior, resulting from their distinct properties associated to various size and shape anisotropies.

Synthesis conditions of filler

As we all known, the morphology and performance of fillers is governed by that of synthesis conditions. In Sundararaj's group work, they synthesized nitrogen-doped nanotubes(N-MWCNTs) and undoped MWCNTs by chemical vapor deposition technique, then prepared PVDF/N-MWCNTs composites and PVDF/MWCNTs, they reported that the types of the N-MWCNTs which synthesized by Co catalyst shows superior electrical and EMI SE performance, resulting from its high synthesis yield, high aspect ratio, low nitrogen content, numerous polarizable centers of N-MWCNTs and good dispersion of N-MWCNTs [33]. In addition, the synthesis temperature has an effect on the and EMI SE performance of PVDF-based composites [32, 118]. In addition, they found that the electrical conductivity of MWCNTs could optimized by varying the synthesis temperature (550 ^{o}C, 650 ^{o}C, 750 ^{o}C, 850 ^{o}C and 950 ^{o}C), the relatively low synthesis temperature of MWCNTs was benefit to obtained a good electrical conductivity and EMI SE performance [34, 115, 116]. Moreover, Choudhary et al. prepared PVDF/MWCNTs composites, they observed that the lower synthesis temperature (800 ^{o}C) of MWCNTs leads to superior electrical conductivity and EMI shielding behavior [32]. The results indicated that the lower synthesis temperature of carbons would causing better shielding performance. On the contrary, for PVDF/carbon-metal composites, the higher carbon-metal synthesis temperature was more help to obtain good EMI SE performance. In Sahoo's group work, they obtained PVDF/Co@C composites [77], PVDF/Ni@C composites [118] and PVDF/Fe_3C@C composites [117], they found that the higher synthesis temperature(1000 ^{o}C) would leads to higher amounts of graphitic carbon and high saturation magnetization, which corresponding to produce higher electrical conductivity and magnetic permeability, resulting in better EMI SE performance [77, 117, 118].

State of dispersion of fillers

Some studies were showed that higher loadings of fillers tend to agglomerate thereby leading to detrimental effects on the structural and EMI SE performance of the PVDF-based composites [40]. Therefore, achieving higher EMI SE at lower fractions of fillers is the current challenge as this also ensures the structural integrity of the nanocomposites. To address this challenge, taking some measures including processing method of composite, orientation distribution, surface functionalization, the designing of co-continuous structures and the selective localization of fillers, to facilitate better dispersion of fillers in the PVDF matrix.

The orientation distribution fillers had remarkable influences on the percolation threshold, dielectric and electrical properties of PVDF-based composites [17, 40, 87, 105]. Xu et al. fabricated PVDF/oriented Ni chains nanocomposites, they reported that rotational orientation method was beneficial for improve the dispersion of magnetic materials in the polymer matrix, thus enhancing in both microwave absorption and EMI SE performance [17]. Kumar et al. prepared PVDF-hexafluoropropylene (HFP)/aligned asymmetric conducting RGO composites, they reported that due to the good dispersion of RGO in PVDF-HFP matrix and the strong specific interaction between RGO and PVDF-HFP matrix, the RGO was oriented and asymmetric distributed in PVDF-HFP matrix, and this asymmetric highly electrically conducting composites exhibited high EMI SE profermance

[107]. Gebrekrstos et al. prepared PVDF/oriented MWCNTs composites, they also found that the alignment of MWCNTs was beneficial to improve the electrical conductivity and EMI SE performance [108]. The results demonstrated that ordered arrangement of fillers in the PVDF matrix can be used to design an efficient EMI shielding materials.

The surface functionalization of fillers was help to facilitate better dispersion of fillers in the PVDF matrix. Eswaraiah et al. prepared PVDF/acid-functionalized graphene composites [87], Kumar et al. prepared PVDF/acid functionalization of MWCNTs composites [39, 40], Sharma et al. prepared PVDF/MWCNTs modified with IL composites [105]. The results observed that the surface functionalization of carbons obtains a good dispersion, resulting in an efficient EMI SE with lower loadings of the fillers [39, 40, 87, 105]. Besides, solution blending composites showed better EMI SE properties than melt blending composites [39, 40]. This indicated that a good dispersion of fillers and selects the appropriate processing method would ensure good EMI shielding of PVDF-based composites to design flexible EMI shielding materials which can find use in commercial applications.

Moreover, dispersing the fillers in the interface of binary polymer blends, the electromagnetic shielding materials with excellent EMI SE performance can be obtained [111]. Sultana et al. prepared co-continuous structure PVDF/PS/MWCNTs composites, they observed that the MWCNTs mainly selective locates in PVDF phase resulting in increased EMI SE of composite [111]. Since most of the polymers are immiscible with each other, selective localization of fillers takes place depending on relative affinity of fillers towards different polymer phases [112, 187]. Yang et al. prepared co-continuous structure PVDF/polyketone (POK)/MWCNTs nanocomposites, they reported that MWCNTs mainly selective locates in POK phase due to MWCNTs has the better compatibility with POK, resulting in low percolation threshold and high EMI SE performance [112]. Biswas et al. prepared PVDF/PC/MWCNTs-MnO_2/RGO@Fe_3O_4 composites, they found that all particles were selectively localized in PVDF phase, resulting in higher adsorption-dominated EMI SE peofermance [187]. These results indicated that the selective localization of fillers in one phase, which facilitate well EMI SE at lower loadings. In addition, the thermodynamic and kinetic factors could influence selective localization of fillers [110]. Salehiyan et al. prepared PVDF/PLA blends by MWCNTs interfacial localization, they observed that MWCNTs localized at the interface of PVDF/PLA blends by kinetic driving [110]. Besides binary polymer blends, the designs of ternary continuous structures could also help to improve the EMI shielding performance of composites. Dou et al. prepared tri-continuous structure PVDF/PS/HDPE ternary blends containing MWCNTs, they found that MWCNTs can be selective located in interfacial PS phase by tuning the thermodynamic and kinetics conditions which leading to an ultralow percolation threshold of 0.022 vol%, and obtained an excellent EMI SE performance at lower content of MWCNTs [20]. Zha et al. obtained PVDF/ethylene-α-octene block copolymer (OBC)/MWCNTs composites, they reported that due to the affinity between OBC and virgin MWCNTs, the filler of MWCNTs was distributed in the interface of PVDF/OBC blends [109].

Furthermore, the combination of surface functionalization of fillers and continuous structures not only improves the dispersion of fillers in the polymer matrix, but also could help to selective localization of fillers in the target position, resulting in high EMI SE performance at lower content of fillers. Kar et al. synthesized PMMA wrapped MWCNTs (PMMA-MWCNTs) via in situ polymerization and prepared PVDF/acrylonitrile butadiene styrene (ABS)/PMMA-MWCNTs to constructing ternary continuous, they reported that PMMA wrapped MWNTs not only improved the dispersion, but also can be localized at the interface of the PVDF/ABS blends to increase the local concentration of MWCNTs, resulting in significantly improvement in EMI SE performance [106]. Additionally, In Bose's group work, they modified MWCNTs with pyrenebutyric acid (PBA), 3,4,9,10-perylenetetracarboxylic dianhydride (PTCD), hydrazono methyl phenol (AHB) or IL via π–π stacking to facilitates its better dispersion in PVDF matrix, the surface of magnetic particles (such as /nickel ferrite (NF), cobalt ferrites (CFs), or Fe_3O_4) were introduced amine-terminal groups to chemically grafted onto modified-MWCNTs (MWCNTs-magnetic particles), and obtained PVDF/PC/MWCNTs/NF composites [149], PVDF/poly(styrene-co-acrylonitrile) (SAN)/MWCNTs-NF or CFs composites [150], PVDF/ABS/PTCD-MWCNTs-Fe_3O_4 composites [151], PVDF/PC/MWCNTs-Fe_3O_4 [59], PVDF/ABS/MWCNTs-Fe_3O_4 composites [152] and multi-layered PVDF/PC/MWCNTs- Fe_3O_4 composites [60]. They found that the MWCNTs-magnetic particles selectively localized in the PVDF phase, the modification of MWCNTs can improves its dispersion and connectivity between the MWCNTs and synergy with co-continuous structures which further resulting in enhanced electrical conductivity and low percolation value, which leading to ideal EMI SE performance. These results indicated that modification of MWCNTs and selectively localizing through co-continuous blends could be significantly enhances the EMI SE performance [59, 149–151].

In addition, the fillers were selectively localized in different phases, the EMI SE performance was many folds higher than when they were localized in the same phases [38, 149, 153, 186, 188]. Thus, it is also a noteworthy method that modify the surface of fillers of different components and distribute them in the different target polymer phase. In Bose's group work, they prepared PVDF/PC/

PANI-MWCNTs-Fe_3O_4/BT composites which BT posited in PC phase and PANI-MWCNTs-Fe_3O_4 selectively localized in PVDF phase [186], and PVDF/SMA/MWCNTs/NiFe composites [38] which MWCNTs was amine functionalized on the surface to posited in the SMA phase (due to amine–anhydride coupling), NiFe particles localized on the PVDF due to thermodynamics, they obtained PVDF/PC/MWCNTs/RGO@BT@Fe_3O_4 composites [188], PVDF/PC/MWCNTs/NF composites [149] and PVDF/PC/PMMA/MWCNTs/barium ferrite (BF) composites [153] which MWCNTs selectively localized in PVDF phase, and RGO@BT@Fe_3O_4, NF or BF posited in PC phase. They observed that the dispersion of conductive fillers and magnetic fillers in different polymer phases generates a large number of interfaces leading to improve dielectric properties, which facilitated to improve the EMI shielding performance of the composites, except for the high electrical conductivity (from conductive fillers) and magnetic properties (from magnetic particles). The result showed that the designs of continuous structures and the selective localization of fillers will be helpful to the improvement in EMI SE performance at lower loadings of the nanofiller [149, 153, 186, 188].

β Phase fraction and content of PVDF

Many research showed that the β phase of PVDF exhibited a better piezoelectric, ferroelectric and pyroelectric properties, which was beneficial to improve dielectric properties and EMI SE performance. As a consequence, there many researchers are more tend choose the suitable fillers to improve the β phase content in PVDF matrix. Kar et al. fabricated PVDF/exfoliated graphite submicron platelets (GPs) composites, they reported that 0.5 wt% GPs in the PVDF matrix significantly improving the β phase of PVDF resulting in the improvement on the dielectric properties, which is responsible for the good EMI SE performance [51]. Sabira et al. obtained PVDF/graphene composites, they observed that the filler of graphene can improve the β phase of PVDF, resulting in a high EMI SE performance [52]. What's more, Meher et al. prepared PVDF/PANI/IL composites, they reported that the presence of IL resulting in the improvement of β phase of PVDF, and the dielectric properties and EMI SE performance of the composites have been increased with the development of β phase of PVDF [80]. Additionally, Soares et al. added IL to PVDF-co-HFP/PANI composites, they found that the addition of PANI in the amount of 30 wt% and the presence of IL could improves the β phase of PVDF originating a good dielectric properties, resulting in an excellent EMI SE performance [123].

In addition, the content of PVDF could affects the mechanical properties and EMI SE performance of composites. Lee obtained core-shell PVDF/PEDOT nanofiber composites [22], they found that the relatively poor fillers doping when the PVDF with lower content, while the high resistance measured in the higher content of PVDF, and the nanofiber composites composed of 16 wt% PVDF, resulting in the highest SE and superior mechanical properties. The results indicated that the EMI SE performance of composites can be adjusted by changing the PVDF content.

Thickness and structure of composites

According to the mechanism of EMI shielding effectiveness, the entering power was reflected, and it had been scattered and absorbed several times at the polymer and the filler's interface. A larger interface area and fillers content would produce higher EMI SE [41, 203]. Thus, for the PVDF-based composites, the EMI SE performance were effectively tuned by controlling the composite's thicknesses [45]. Zhao et al. prepared PVDF/Ni chains composites, they observed that the EMI shielding performance of composites was increased with increasing thickness, and the average EMI performance of the PVDF/6 wt% Ni chains composites at thicknesses of 0.2 mm, 0.3 mm, 0.4 mm, and 0.5 mm were 15.4 dB, 24.7 dB, 30.3 dB, and 35.4 dB, respectively [47]. What's more, they prepared PVDF/carbon (MWCNTs or graphene)/Ni chains composites, they reported that increasing the thickness of composites can significantly improves the EMI SE performance, due to the increase of composites thickness, the concentration of conductive fillers and the interface area between fillers and PVDF was increased, causing the improvement in the reflection and absorption effect of composites on electromagnetic wave, and the total shielding of the PVDF/graphene/6 wt% Ni chains composites increased from 23.6 to 57.3 dB and, as their thicknesses were increased from 0.3 to 0.6 mm [45]. In addition, Bhaskara Rao et al. prepared PVDF/MWCNTs composites and design multilayer structure by stacking techniques [119]. They reported that the multilayer stacking can increase the EMI SE of composites, due to the introduction of multilayer structure not only increases the thickness of composites and fillers content, but also can obtain a large the interface area between each layer leading to multiple reflections and dielectric loss, which was beneficial to the improvement in EMI SE performance of composites. The maximum EMI SE of 25 dB was obtained for 1 layer sample of 0.3 mm thickness and further improving to 32 dB by 3 layers stack it (0.9 mm thickness). This result indicated that apart from adjusting thickness of composites, an effective structural design also can further improve EMI SE performance of composites.

Studies have shown that different structure (multilayer structure, sandwich structure, porous structure, segregated structure) of PVDF-based composites have different EMI

SE performance. Sandwich structures is a kind of multilayer structure which has different outer and inner layers, and individual performance of composites of each layer can be tailored to achieve the right combination of performance. Qi et al. prepared PVDF/graphene, PVDF/MWCNTs and PVDF/Ni composite layers and make a 3-layered sandwich structure composites [46]. They found that the overall shielding performance could be further improved by increasing the thickness of the EMI shielding layer and the number of layers, and the novel 3-layered sandwich structure composites possess a high EMI SE performance, especially 3-layered sandwich structure composites which with a unique order of graphene-Ni-MWCNTs. Yang et al. prepared sandwich structure PVDF/MXenes/AgNWs composites [142]. They observed that the multiple reflection between neighboring conductive layers, resulting in an outstanding EMI SE of sandwich structure composites. Sushmita et al. obtained multilayered sandwich structure composites, which blending individually with unique order of PVDF/10 wt% RGO-Fe_3O_4 or PVDF/10 wt% MoS_2-Fe_3O_4 composites-PMMA composites-PC/3 wt% MWCNTs composites. They found that this unique arrangement of a multilayered assembly suppressed EMI primarily by absorption [191]. In Bose's group work, they obtained sandwich-structured composite films by layer-wise assembly, which the PVDF/Fe@CF composites in the top and bottom, PVDF/MWCNTs composites in the middle layer, they also observed that the sandwich structured composites showed a significant improvement in EMI SE performance, resulting from the synergistic effect of each individual layer [129]. In particular, they prepared PVDF/MWCNTs composites and designed sandwich-structured composites via stacking method, they found that careful control of different layers in this layer-wise assembly, facilitate significantly improves EMI SE performance [130]. And, they obtained multilayered thin polymer composites through stacking method [156], the PVDF/PC/MWCNTs/manganese ferrite composites as the outer layers, the porous structure PVDF/PC/MWCNTs composites as the inner layers. They reported that the composites possess excellent EMI absorption, sandwich structure and the middle porous layers also could exhibit multiple reflections. The result indicated that the sandwich structure composites can be fabricated with tailor-made composites through design different inner structures (sandwich structure or porous structure) and multilayer assembly, sandwich structure or porous structure could improve dielectric properties, resulting in a good EMI SE performance [46, 129, 130, 142, 156, 191]. Thus, porous construction composites potential to be used as lightweight high EMI shielding materials [2, 5, 15, 204].

Sharma et al. obtained porous structure PVDF/PMMA/MWCNTs composites which selectively etching PMMA phases from composites, and then prepared porous structures PVDF/MWCNTs/$NiFe_2O_4$ composites through vacuum filtration method [157]. They found that the porous structure of composites was beneficial to improve the interfacial polarization and multiple reflections, resulting in a significant improvement in the absorption-dominated EMI SE. As the same processing, Wang et al. prepared 3D network porous PVDF/MWCNTs [42], due to the unique 3D construction, the porous PVDF/MWCNTs composites exhibit high EMI SE performance by reflecting and scattering waves many times in the interior of the composites, and absorption loss was the dominant contribution to the shielding mechanism. These results indicated that porous structure was helpful to enhance the multiple-reflection effect and interfacial polarization, leading to improvement in the shielding performance [42, 157]. What's more, in Khatua's group work, they prepared PVDF/PANI@Fe_3O_4/carbon [75] and PVDF PANI@Fe_3O_4/RGO [44] composites with porous structure by leaching out of the NaCl out from the composites. They found that the porous structure combined with continuous conductive network and magnetic particles resulting in an excellent EMI SE performance at low loading of fillers [44, 75].

Apart from the selective etching or salt leached method, physical foaming such as injection molding foaming, batch foaming and supercritical CO_2 foaming method has been demonstrated to be the most efficient way to prepare porous composites. Zhao et al. prepared PVDF/graphene nanoplatelet (graphene) [120] and PVDF/MWCNT [19] nanocomposites with microcellular structure through batching foaming. They reported that the microcellular structure in the PVDF foams could cause multiple reflection and scattering, which facilitated to improve the absorption-dominated EMI SE performance [19, 120]. Jia et al. prepared microcellular PVDF beads through supercritical CO_2 foaming method, and fabricated microcellular PVDF/carbon-based filler (carbon black or graphene) composites by induced phase separation technique [121]. They reported that due to multiple reflections which resulting from the large interfacial area and the microporous structure of composites, the composites possess high EMI SE performance. Moreover, Zhang et al. prepared PVDF/Ni chains composites by foaming method [63]. They reported that due to the foaming would induce a more efficient segregated structure to build the conductive pathway, and the microcellular structure was not only immensely lower the percolation threshold, but also promoted multiple reflections, resulting in good absorption-dominated EMI SE performance. Wang et al. prepared segregated PVDF/MWCNTs composites [122], they observed that the electrical MWCNTs was only lie on the interfaces of PVDF grains to formed the segregated structure, which forming the conductive networks and introducing numerous interfaces with the PVDF matrix, leading to the improvement in the conductive loss, and multiple reflections. Thus, the segregated structure is a very promising processing method to facilitated composites possess an excellent EMI SE performance at low loading, resulting from the fillers was mainly lie on the interfaces of polymer grains in segregated composites.

Conclusion and outlook

Based on the above generalization of the PVDF-based electromagnetic shielding materials, it can be concluded that the application of PVDF-based composites in the field of electromagnetic shielding has been more and more extensive, and the research of the PVDF-based electromagnetic shielding materials is indeed a very attractive work. This review highlighted the recent developments related to PVDF-based composites as promising electromagnetic shielding materials. Some studies show that the single fillers such as conductivity, magnetic or dielectric materials was incorporated in the PVDF matrix could obtain effective electromagnetic shielding performance. There also have been more tremendous interests in PVDF-based composites with multiple-component fillers which including binary fillers and multiple fillers, for electromagnetic shielding applications. The integration of binary fillers in the PVDF matrix, could generally create obvious synergetic effects and complementary behaviors, as well as more loss mechanisms (e.g., various polarization loss), which contribute to the electromagnetic shielding performance greatly. To further upgrade the shielding for incident EM waves, the EMI SE performance of PVDF-based composites could be optimized by incorporating multiple fillers (ternary or quaternary fillers). Therefore, PVDF-based composites with ternary or quaternary fillers are making breakthroughs and are becoming a popular approach for high-performance electromagnetic shielding materials. Additionally, there are many factors influence the shielding effectiveness of the PVDF-based composites containing fillers (such as the content, type, morphology, size, synthesis conditions, dispersion state of fillers in the PVDF matrix), the β phase fraction of PVDF, thickness and structure of composites and so on. In spite of extensive research efforts, the PVDF-based composites with high EMI SE performance under lower loading of fillers and minimum thickness still remains challenging. Significantly, designing of binary or ternary continuous structures and the selective localization of fillers facilitated the improvement of EMI shielding performance at low loadings. Moreover, designing the multilayer structure, sandwich structure, porous structure or segregated structure of PVDF-based composites could obtain excellent adsorption-dominated EMI SE performance. Therefore, in the future, the research trend of PVDF-based electromagnetic shielding materials tends to the preparation of PVDF-based with multiple fillers, plays the multi factors synergy, and make it have better shielding performance by reasonable preparation methods and optimal structural designs. In addition, functional particles can also be introduced to endows the PVDF-based electromagnetic shielding composites with special functions, such as flame resistance, superhydrophobicity, antiultraviolet aging, and so on, so as to makes them develop in the direction of higher performance, more intelligence and more environmental protection.

Nowadays, a large part of achievements has been made with regard to the PVDF-based composite shielding materials. However, more and more research interest has been paid to the design of new structure to obtain satisfactory electromagnetic shielding performance, and it is still highly necessary to study the related fundamental scientific issues. For instance, it is important to understand the shielding mechanisms of the structure and to investigate the physical/chemical properties of interfaces as well as their effects on the shielding performance. Thereafter, we can reveal the interactions between the PVDF and other component fillers in composite materials and the interrelations of the electromagnetic shielding effects from several component fillers, and thus prepare the novel PVDF-based composite shielding materials with meeting the requirements. It is believed that novel PVDF-based composites with ideal compositions and optimal microstructures will present a bright future in the field of microwave shielding.

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Declarations

Competing interests The authors declare no potential conflict of interest.

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