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REVIEW
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Fullerene Reinforced Polymeric Nanocomposites for Energy Storage—Status and Prognoses

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This review deals with the progress in the field of polymer/fullerene nanocomposites particularly for the energy storage applications. Fullerene is a unique zero dimensional nanocarbon nanomaterial. Fullerene proposes several unique structural, optical, electrical, thermal, mechanical and other superior physical features to the polymeric nanocomposites. Consequently, the high performance polymer/fullerene nanocomposites result from the amalgamation of the unique characteristics of fullerene with the functional polymers. Here, the advancements in the polymer/fullerene nanocomposites regarding their processing and properties, especially the electrical conductivity, charge storage capacities, charge density, power density, charge-discharge, and cyclic performance have been discussed. Moreover, the future and challenging prospects have been summarized anticipating the progress in the field of polymer/fullerene-based energy storage technology.

Keywords: polymer, fullerene, nanocomposite, conductivity, supercapacitor

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1 INTRODUCTION

Fullerene is a unique allotropic form of nanocarbon (Thota et al., 2010). Like other nanocarbons, fullerene molecules have been reinforced in the polymers. Inclusion of fullerene in polymers has been known to enhance the facile processing and characteristics (Kim et al., 2017; Kanbur and Tayfun, 2019). Consequently, the polymer/fullerene nanomaterials have gained interest in the material science (Ravi et al., 2007). Fullerene nanofillers in polymers have amended the structural (Djahnit et al., 2019), morphology/crystallinity (Mohamadi and Sharifi-Sanjani, 2018), electrical conductivity (Yoshino et al., 1999), mechanical robustness (Zhao et al., 2021), and heat constancy (Djahnit et al., 2019) properties. Subsequently, polymer/fullerene nanomaterials have promising applications in the supercapacitor (Shen et al., 2021), solar cell (Armin et al., 2018), and biomedical (Moon et al., 2010) fields. Here, the poor fullerene miscibility in polymers may hinder the nanocomposite features (Prylutskyy et al., 2014). For the purpose, the fullerene molecules have been modified and introduced in polymers to developed the physical or chemical interactions. Recently, energy storage devices have adopted the use of polymer/fullerene nanocomposite (Issar and Arora, 2021; Shrestha et al., 2021). The capacitance properties and application in energy storage device have gained recent research interest (Lawes et al., 2015).

This review article presents progress in the arena of polymer and fullerene based nanocomposites. Inclusion of fullerene molecules in polymers has been employed to progress the electrical conductivity of the nanocomposites. Moreover, the specific capacitance, power density, charge density, charge-discharge, recyclability, and durability properties of the polymer/fullerene nanocomposites have been explored. However, novel polymer/fullerene nanocomposites have

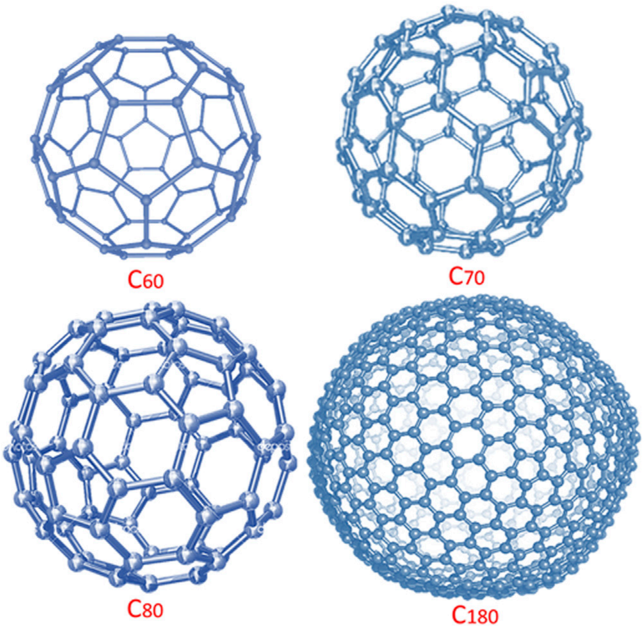

FIGURE 1 | Few fullerene analogues.

several challenges, which need to be overcome to attain high performance. In this regard, the mostly conducting polymers have been doped with fullerenes to attain the desired properties for the supercapacitors.

2 FULLERENE

Fullerene is a symmetrical form of nanocarbon (Jehoulet et al., 1992). Fullerene is a zero dimensional molecule with sp^2 hybrid carbon atoms (Chang, 2006). Fullerene is made up of polygones, i.e., pentagons and hexagons (**Figure 1**).

The discovery of fullerene is dated back to 1985 (Montellano López et al., 2011). Fullerene usually has inherent electrical, optical, magnetic, mechanical, thermal, and biomedical features (Withers et al., 1997; Akasaka et al., 2001; Giacalone and Martín, 2006). Fullerene molecules have been synthesized through the plasma method, arc discharge procedure, microwave synthesis, and chemical routes (Withers et al., 1997; Akasaka et al., 2001; Mojica et al., 2013). The methodological applications of fullerene have been perceived in solar cells (Chae et al., 2011), sensors (Modi et al., 2003), drug delivery (Gallo et al., 2007), etc. The solubility properties of fullerene molecules in various solvents have been focused (Nierengarten, 2004; Dmitruk, 2010). The chemical modification of C_{60} has found to improve the solubility of these molecules in water, chloroform, xylene, and organic solvents (Giacalone and Martín, 2006). Sometimes, the solubilizing agents or polymers such as poly(vinylpyrrolidone) have been used to improve the solubility of fullerene (Behera and Ram, 2016; Behera and Ram, 2017).

3 FULLERENE REINFORCED POLYMERIC NANOCOMPOSITES

Fullerene C_{60}, C_{70}, and higher fullerene analogues have been employed as reinforcement to form the polymeric nanocomposites (Geckeler and Samal, 1999). The polymer/fullerene nanocomposites have been premeditated for the optical, mechanical, thermal, charge transport, and energy storage characteristics (Kausar, 2017; Etxebarria et al., 2015; Harris, 2020). The dispersion and solubility of the fullerene molecules in polymers remain as major challenges during the nanocomposite formation (Mackay et al., 2006). Lack of solubility may lead to fullerene aggregation and mass formation. The agglomeration problem may lead to the decrease in the properties of the polymer/fullerene nanocomposites. In this way, the large-scale processing of the polymer/fullerene materials is quite challenge (Bartelt et al., 2014). The modification or functionalization of fullerenes have been focused (Song et al., 2018). Thus, the better physical or covalent contacts between the polymer and fullerene nanoparticles have revealed high performance nanocomposites. The polymer/fullerene nanocomposites have been researched for the improved electrical, magnetic, thermal, and mechanical features (Kausar, 2021). The thermoplastic and thermosetting polymers, such as epoxies, polystyrene, polyethylene, poly(methyl methacrylate), poly(ethylene glycol), and block copolymers, have been filled with the fullerenes. Then, the conducting polymers have been reinforced with the fullerene molecules (Bergbreiter et al., 1984). Among conducting polymers, polythiophene (PTh), polyaniline (PANI), polypyrrole (PPy), poly(3,4-ethylenedioxythiophene), etc. and derived polymers have been connected with the fullerene molecules to develop the nanocomposites (Umeda, 2005). Few conducting polymers covalently linked with the fullerene molecules are given in **Figure 2** (Martín, 2006).

The thermal stability properties of the polymer/C_{60} nanocomposite have been explored (**Table 1**) (Pereira et al., 2015). The addition of C_{60} contents improved the decomposition temperature of the nanocomposites (Alekseeva et al., 2018).

4 ELECTRICAL CONDUCTIVITY OF POLYMER/FULLERENE NANOCOMPOSITES

The polymer/fullerene nanocomposites have been considered for the supercapacitors (Schon et al., 2014), photovoltaics (Campoy-Quiles et al., 2008), electronics (McCamey et al., 2008), and other significant technical relevance. Tumbleston et al. (2014) produced the poly(3,4-ethylenedioxythiophene): polystyrene sulfonate acid and fullerene C_{60} based nanocomposite. The conjugated polymers behave as electron donor, whereas the fullerene molecules act as electron acceptors. Huang et al. (2011) formed poly(3-hexylthiophene) (P3HT) and fullerene C_{60} derived

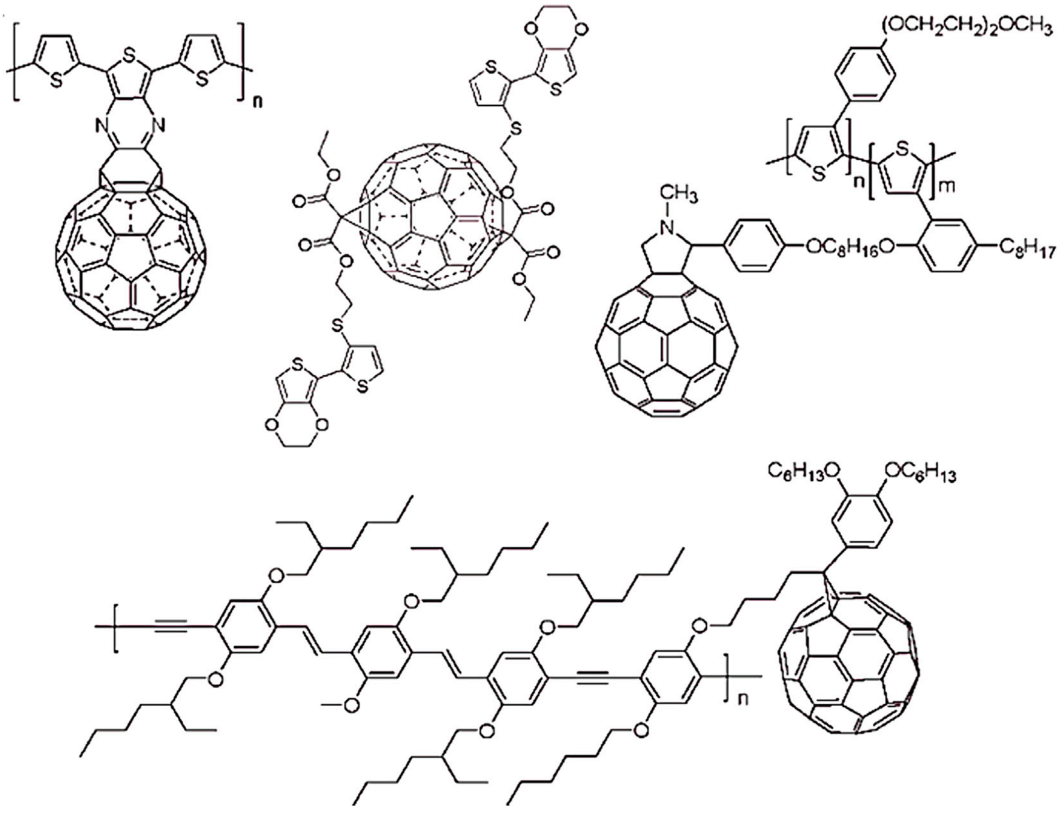

FIGURE 2 | Some polymers with covalently linked fullerenes (Martín, 2006). Reproduced with permission from RSC.

TABLE 1 | Thermo-oxidative stability of polymer and corresponding fullerene nanocomposites (Pereira et al., 2015).

Polymer (wt.%)	C_{60} (wt.%)	$T_{2\%}$ (°C)	T_{max} (°C)
100	0	280	348
99	1	302	381
97	3	303	375

TABLE 2 | Resistivity, Hall mobility, and carrier density of P3HT and nanocomposite thin film (Zabihi et al., 2016). P3HT = poly(3-hexylthiophene). Reproduced with permission from Elsevier.

Coating	P3HT	P3HT nanocomposite
Resistivity (Ωcm)	1.44×10^5	1.52×10^5
Carrier mobility (cm^2/Vs)	22.8	48.9
Carrier density (cm^{-3})	1.47×10^{12}	1.18×10^{12}

nanocomposites. Here again, the electron conduction was observed due to the electron donating polymer and electron accepting C_{60}. Chirvase et al. (2004) designed the P3HT and (Mohamadi and Sharifi-Sanjani, 2018Mohamadi and Sharifi-Sanjani, 2018)-phenyl-C61 butyric acid methyl ester (PCBM) based poly(3,4-ethylenedioxythiophene) (Mohamadi and Sharifi-Sanjani, 2018):-phenyl-C61 butyric acid methyl ester (P3HT:PCBM) nanocomposites. The P3HT:PCBM and fullerene based nanocomposites were explored for the conducting properties. The short circuit current density of 7.8 $mAcm^{-2}$ was observed. Zabihi et al. (Zabihi et al., 2016) also formed the P3HT:PCBM nanocomposites. The P3HT: PCBM and C_{60} based nanocomposites had high electrical conductivity, charge mobility, and current density properties. **Table 2** shows the conductivity properties of the P3HT and related nanocomposite. The carrier mobility and carrier density of the nanocomposite were found superior, relative to the neat P3HT. The notable augmentation in the electrical properties of the nanocomposite was due to the matrix-nanofiller interactions. Cheng et al. (Cheng et al., 2017) inspected PANI and polydivinylbenzene (PDVB) based PANI/PDVB nanocomposites with fullerene. **Figure 3** shows the enhancement in the electrical conductivity with the frequency. The electrical conductivity of the PANI/PDVB nanocomposite was enhanced from the 9×10^{-10} to 63.7 Sm^{-1}. In this way, various conjugated polymers and their blended combinations have been used with fullerenes to promote the electron conduction characteristics.

Polyazomethine is a conjugated polymer (Sobarzo et al., 2021). Bronnikov et al. (2017) reported polyazomethine/fullerene nanocomposites. Fullerene was loaded in 0.25–2.5 wt.% contents. The DC electrical conductivity of the polyazomethine/fullerene nanocomposites was

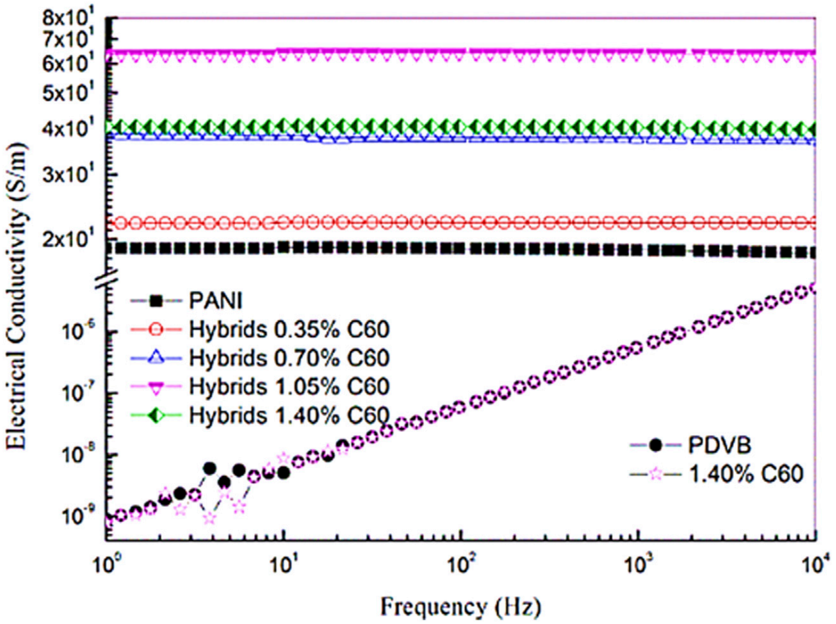

FIGURE 3 | Electrical conductivity of nanocomposite with different amount of C_{60} (Cheng et al., 2017). PANI, polyaniline; PDVB, polydivinylbenzene. Reproduced with permission Elsevier.

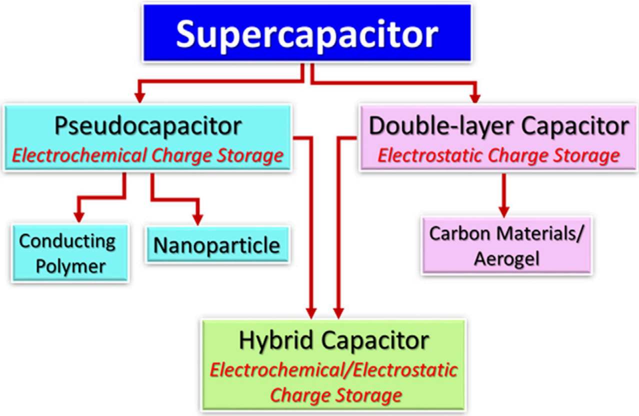

FIGURE 4 | Hierarchical cataloguing of supercapacitors.

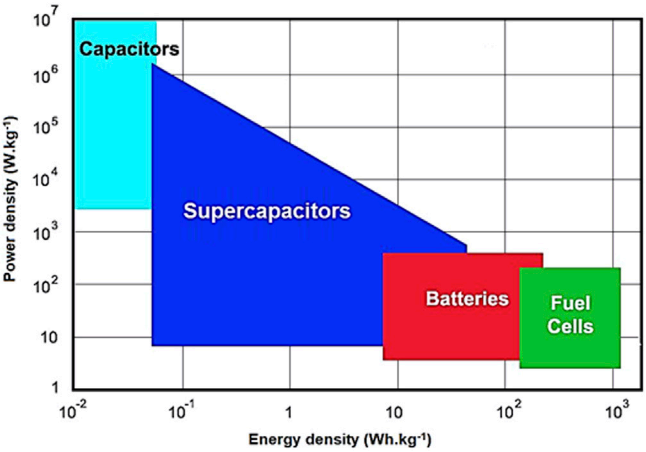

FIGURE 5 | Ragone chart of power density as a function of energy density for numerous energy devices (Libich et al., 2018). Reproduced with permission Elsevier.

augmented with the cumulative nanofiller content, as well as the rising temperature. The effect was observed due to the homogeneous dispersion of the fullerene molecules in the polyazomethine and the formation of percolation complex (Bronnikov et al., 2016). Ltaief (2004) amalgamated poly[2-methoxy-5-(2′-ethyl)hexoxy-1,4-phenylenevinylene] with fullerene. The aromatic stacking interactions between the polymer and fullerene has upsurges the electron conduction properties. The 40 vol.% fullerene developed conductive pathways and percolation threshold.

Mostly, conjugated polymers have generated better conducting trails with the fullerene molecules.

5 POLYMER/FULLERENE NANOCOMPOSITES FOR ENERGY STORAGE APPLICATIONS

Supercapacitors have been considered as capable energy storage devices for advanced electronic devices (Naoi et al., 2013; Raza et al., 2018). Among energy storage devices, the supercapacitors possess high specific capacitance, power density, and charge-discharge performance (Simon and Gogotsi, 2010a). The major types of the supercapacitors are portrayed in **Figure 4**.

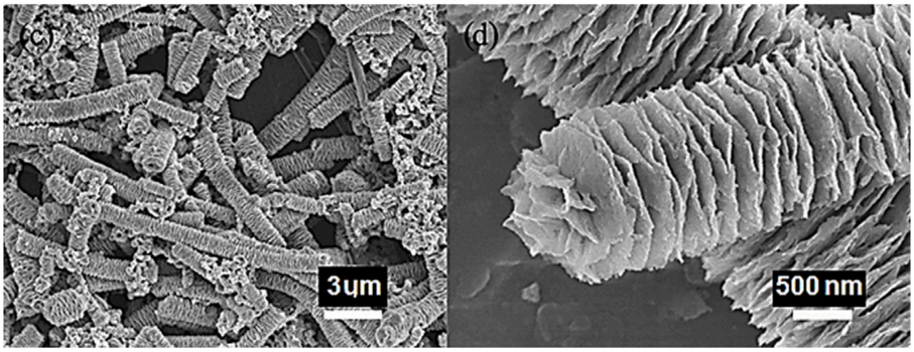

FIGURE 6 | SEM images PANI-EB/FW (Wang et al., 2017). PANI-EB/FW = polyaniline emeraldine base/fullerene C_{60} whisker. Reproduced with permission from Elsevier.

Supercapacitors have been mainly categorized as the pseudocapacitors having electrochemical charge storage, double-layer capacitors with electrostatic charge storage, and hybrid capacitors containing electrochemical/electrostatic charge storage. Among energy storage devices, supercapacitors have wide ranging energy density and power density performances (**Figure 5**) (Libich et al., 2018). The supercapacitor performances have been found in between the capacitors and batteries/fuel cells (Conway, 1999; Hu et al., 2006; Pothu, 2020).

Energy storage devices have been the current demand of the modern electronics and vehicle industry (Winter and Brodd, 2004; Simon and Gogotsi, 2010b; Larcher and Tarascon, 2015). The pseudocapacitors or electrochemical supercapacitors have been deliberated as talented energy storage devices (Mousty and Leroux, 2012; Chen, 2017). Supercapacitors revealed inexpensiveness and high power stowage (Bao et al., 2011; Chen et al., 2011). These devices have been known for the high resilience, high energy density, and high power density, compared with the conservative storage devices (Ryu et al., 2004; Kim et al., 2005; Sen et al., 2013). Consequently, supercapacitors have caused revolution in the field of next-generation energy storage devices (Liu et al., 2010; Arico et al., 2011). Supercapacitors have been successfully applied in the electronics, robotics, etc. (Wang et al., 2007; Smith et al., 2008; Chen et al., 2010). The supercapacitors have found the use of conjugated polymers such as polyaniline, polypyrrole, polythiophene, and poly(3,4-ethylenedioxythiophene) (PEDOT), etc. (White and Slade, 2004; Snook et al., 2007; Liu et al., 2008). These conjugated polymers possess high electron conduction and specific capacitance (Patake et al., 2009; Li et al., 2010; Poizot and Dolhem, 2011). However, drawback of using conducting polymers in supercapacitors may be the low charge-discharge, stability, and reversibility. Therefore, the conjugated polymers have been filled with the nanoparticles such as graphene, graphite, and inorganic nanoparticles (Stenger-Smith et al., 2002; Jayalakshmi and Balasubramanian, 2008; Zhang et al., 2010).

Fullerenes have been explored for the supercapacitor devices due to the intrinsic high surface area and electrical conductivity properties (Wang et al., 2017; Benzigar et al., 2019). The polymeric nanocomposites of fullerenes have been applied in the supercapacitor designs (Pech et al., 2010; Lin et al., 2013). Wang et al. (2017) prepared polyaniline emeraldine base (PANI-EB) and polyaniline emeraldine base/fullerene C_{60} whisker (PANI-EB/FW) nanocomposite. The PANI-EB/FW was used in the supercapacitor electrode. **Figure 6** shows the unique morphology of the PANI-EB/FW supercapacitor electrode. The unique fullerene nanowhisker with high surface area was observed. **Figure 7** demonstrates the specific capacitance of PANI-EB/FW as 813 Fg^{-1}, which was found considerably higher than the neat PANI-EB (248 Fg^{-1}). **Figure 8** displays the capacitance retention of 85.2% for the nanocomposite electrode, after 1,500 cycles. The excellent performance was due to the synergistic effects of the polymer and fullerene nanoparticles.

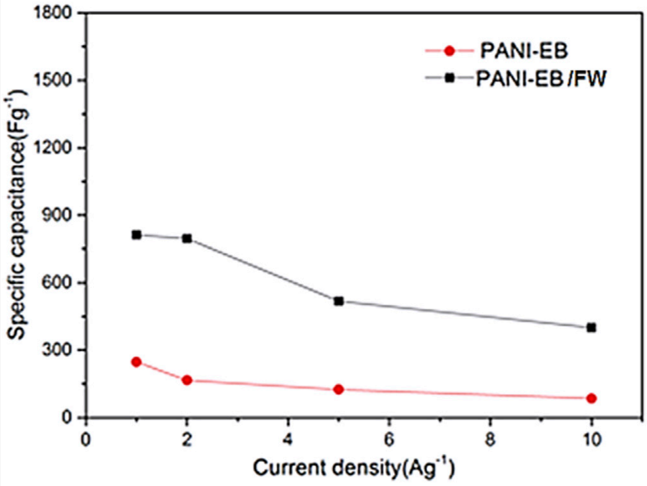

FIGURE 7 | Specific capacitance of PANI-EB and PANI-EB/FW at different current densities of 1, 2, 5, 10 Ag^{-1}, respectively. PANI-EB = polyaniline emeraldine base; PANI-EB/FW = polyaniline emeraldine base/fullerene C_{60} whisker (Wang et al., 2017). Reproduced with permission from Elsevier.

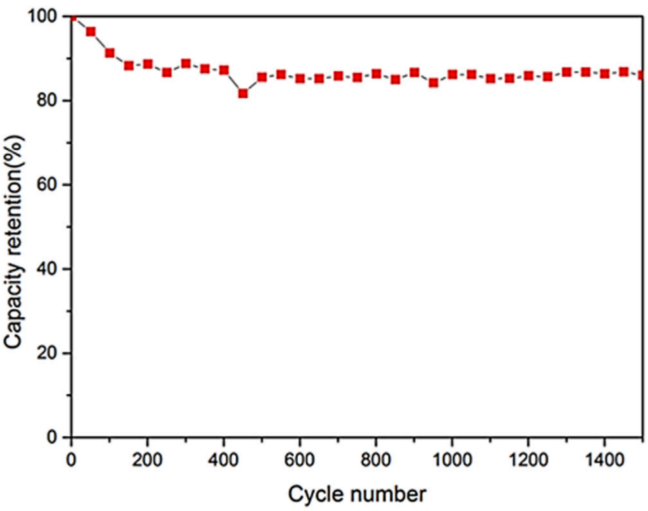

FIGURE 8 | Cycling stability of PANI-EB/FW nanocomposite at 10 Ag^{-1} for 1,500 cycles (Wang et al., 2017). PANI-EB/FW = polyaniline emeraldine base/fullerene C_{60} whisker. Reproduced with permission from Elsevier.

Xiong et al. (2012) also designed PANI-EB and fullerene C_{60} based supercapacitor electrode. **Figure 9** presents the synthesis course to the PANI-EB and C_{60} based nanocomposite. The C_{60} was linked with para-phenylenediamine (PPD) to form functional fullerene. Then, the PPD functional C_{60} was included to the *in situ* polymerization of aniline monomer. **Figure 10** depicts the increased specific capacitance of 776 Fg^{-1}, relative to the neat PANI-EB (492 Fg^{-1}) over 50 cycles. The capacitance retention of the fullerene based nanocomposite and neat PANI-EB were observed as 96.5% and 94.9%, respectively, after 500 cycle. The C_{60} has electron retreating efficiency towards the conjugated polymer and so enhancing the conductivity and capacitance properties (Xiong et al., 2010; Dou et al., 2013). Ramadan et al. (2020) produced the polyaniline (PANI)/phenyl-C_{60}-butyric acid methyl ester (PCBM) based PANI/PCBM nanocomposite. The PCBM of 2.5, 5 and 10 wt.% was added to the polyaniline. **Figure 11** shows that the specific capacitance of the neat polyaniline was 1,110 Fg^{-1}. The capacitance was considerably enhanced to 2,201 Fg^{-1} with wt.% nanofiller. The 96% capacitance retention was observed over 1,000 cycles. Such high values of specific capacitance were observed due to the better amalgamation of PANI and PCBM and high charge transport properties.

The poly(3-hexylthiophene) (P3HT) and PCBM derived P3HT:PCBM nanocomposites have also bene reported for the supercapacitors (Wang et al., 2012; Momodu et al., 2015). The current-voltage and capacitance-voltage techniques were used for the electrical conductivity and specific capacitance measurements (Amao, 2003). Such supercapacitors have been used for the optical/electronic devices. Hence, the approaching investigations must focus the better design combinations of the conjugated polymers and fullerenes with superior specific capacitance values (Peng et al., 2019).

6 FUTURE AND CHALLENGING ATTRIBUTES OF POLYMER/FULLERENE TOWARDS ENERGY STORAGE

Nowadays, the modern supercapacitor devices having superior resilience, capacitance, power density, and charge storage have been necessitated (Sen and De, 2010; Amoura et al., 2015; Senthilkumar, 2015). In this regard, novel design

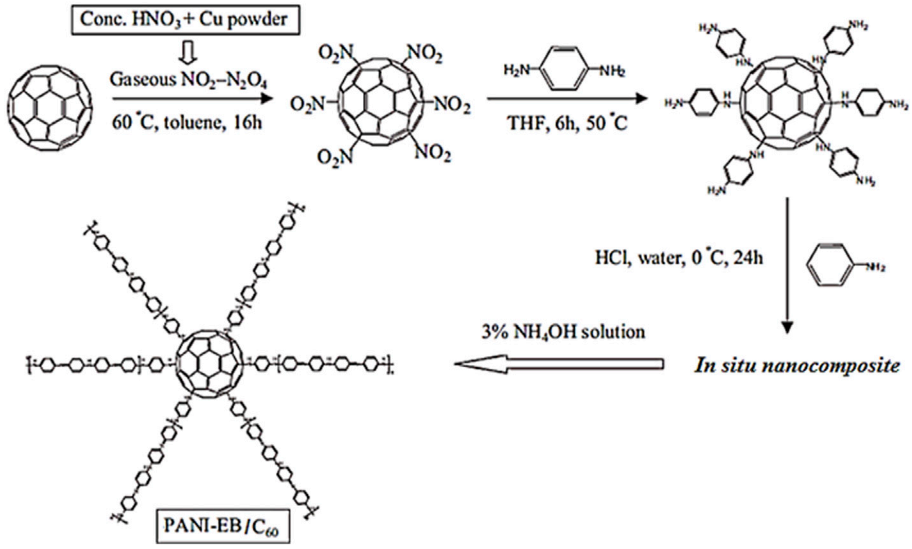

FIGURE 9 | Synthesis route to PANI-EB/C_{60} hybrid (Xiong et al., 2012). PANI-EB/C_{60} = polyaniline emeraldine base/fullerene C_{60}. Reproduced with permission from Elsevier.

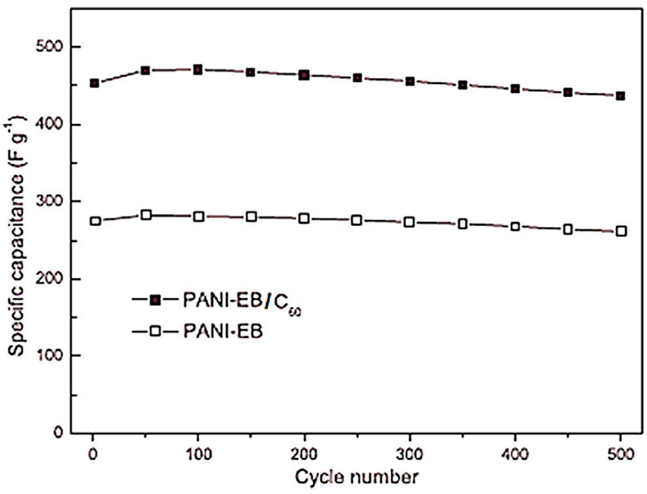

FIGURE 10 | Specific capacitance against the cycle number for PANI-EB and PANI-EB/C_{60} (Xiong et al., 2012). PANI-EB = polyaniline emeraldine base; PANI-EB/C_{60} = polyaniline emeraldine base/fullerene C_{60}. Reproduced with permission from Elsevier.

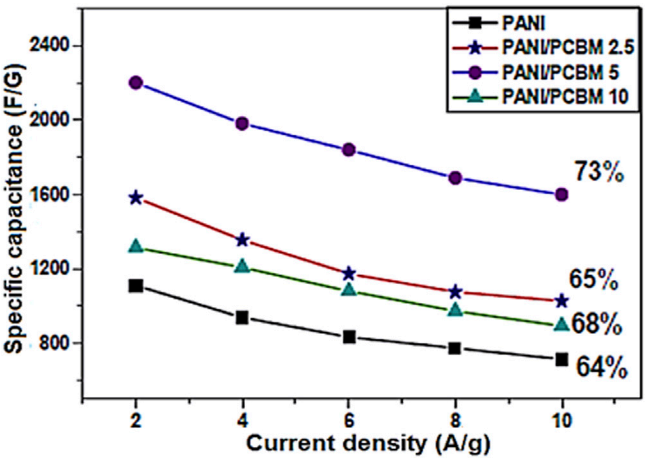

FIGURE 11 | The specific capacitance vs. current density for PANI/PCBM nanocomposite (Ramadan et al., 2020). PANI/PCBM = polyaniline/phenyl-C_{60}-butyric acid methyl ester. Reproduced with permission from Elsevier.

distinctions have been researched for the energy storage devices (Choudhary et al., 2020; Duran et al., 2020; Wang et al., 2020). Conducting polymers such as PANI, PTh, P3HT, PEDOT, etc. possess high electrical conductivity and capacitance to be applied in the supercapacitor electrodes (Wu, 2002; Jayalakshmi et al., 2007; Pandolfo, 2013). The supercapacitors having light weight, low cost, high capacitance, high power density, high constancy, recyclability, and charge-discharge recitals have been investigated (Villers et al., 2003; Cericola and Kötz, 2012; Abdelhamid et al., 2015). Use of fullerene molecules have further heightened the performance of the conjugated polymers in the supercapacitors. The polymer/fullerene nanocomposites with combinations of numerous polymers and fullerene molecules towards the structure, morphology, and energy storage properties have unveil the plethora of future opportunities. These devices have been pragmatic in electronics, digital devices, and electric vehicles. Nevertheless, the future supercapacitors with higher energy density than batteries have been desirable. The specific capacitance of the polymer/fullerene nanomaterials has been directly related to the electron conduction properties. Consequently, there is a lot of interest to augment the electrical conductivity of the nanocomposites. The fullerene aggregation may decrease the electron conductivity and capacitance characteristics. In this regard, the fullerenes demand apposite functionalization and dispersion to develop the percolation pathways in the polymers. Better interactions between polymer and fullerene nanofiller presented considerable development in the electrical conductivity and ensuing features for the high performance supercapacitors. Moreover, the future research may focus the use of phase change material in these energy storage materials (Yin et al., 2021). Especially the use of eco-friendly or bio-based materials may yield high performance fullerene based energy storage devices (Gong et al., 2021; Yin et al., 2022).

7 SUMMARY

Hence, this review abridges the indispensable aspects of the polymer/fullerene nanocomposites for the energy storage devices. This article will be pioneering in the field of polymer/fullerene based supercapacitors. The polymer/fullerene nanomaterials have enriched electrical conductivity properties in addition to the thermal stability and mechanical strength features. Mostly, the conducting or conjugated polymers have been explored with fullerenes in the supercapacitor application. The dispersion properties of the fullerene nanoparticles in the polymeric matrices have enhanced the electron transport through the system. The superior electrical conductivity has in turn promoted the specific capacitance, and other desirable supercapacitor properties.

AUTHOR CONTRIBUTIONS

AK: Writing of entire manuscript, outline, original draft, analysis, review, and editing.

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