Chapter 2 Review of Literature

Chapter 2

Review of Literature

2.1 Introduction

In this section, a summary of the literature survey during the research has been presented. The information obtained from this survey helps to select the objectives and gives a thorough idea about the pathways followed to achieve the objectives. This chapter includes the review of available research reports of

- Bismaleimide composites
- Epoxy composites
- Bismaleimide Epoxy composites
- Polymer matrix composites with barium titanate (BT) fillers
- Polymer matrix composites with Rochelle salt crystals (RS) as fillers
- Polymer matrix composites with surface modified barium titanate as fillers.
- Polymer nanocomposites with MWCNT fillers

2.2. Bismaleimide (BMI) composites

Fabrication of BMI composites with different fillers and reinforcements such as carbon fiber, glass fiber etc., their properties and applications were discussed and described in detail in this literature review.

Chemical change of BMI matrix during thermo-oxidative ageing and its effect on mechanical properties of carbon–fiber reinforced BMI composites were investigated by Xinying Lv et al. (2012). It can be inferred from the experimental data that ageing time had a more negligible effect on the mechanical properties at 25°C than that at 150°C. As a result of decomposition of BMI resin on the surface of composites at 150°C, there exists a slight decrease in flexural strength and ILSS along with less chemical changes of BMI matrix resin¹.

R. Biju developed elastic memory composites (EMCs) and C. P. Raghunadhan Nair (2013) by the co-reaction of bismaleimide-end capped-poly(tetramethylene oxide) (BMI-PTMO), 2,2'-diallyl bisphenol–A (DABA) and 2,2-bis-4 [(4-maleimido phenoxy) phenyl]-propane (BMIP) and further reinforcement are done with carbon fabric. With the increase in PTMO content, shape recovery increases with a decrease in recovery time. In order to avoid sudden deployment, slow recovery is recommended which can be obtained from the appropriate composition of the blends².

Effects of length, diameter and number of walls on thermal, electrical and mechanical properties of carbon nanotube (CNT) -BMI composites were described and discussed in detail by Xin Wang et al. (2013). Composites with a large diameter of longer and multiwalled carbon nanotubes exhibited higher electrical and thermal conductivity. Bismaleimide composites consisting of CNT with increased length and diameter do not show any appreciable change in mechanical properties³.

The effect of isothermal ageing on flexural properties, weight loss and microstructures, surface shrinkages and cracks in carbon fiber reinforced (IM7)-BMI composites were studied in detail by Mohammad H. Haque et al. (2014) after exposing the composites to 260°C for 3000h in air⁴.

MWCNT-polyetherimide (PEI)-2,2'-diallyl bisphenol A modified 4,4'-bismaleimido diphenylmethane BDM (BD) composites were prepared by Yicheng Jiao et al. (2014) using melt mixing technique. With 0.4 weight % loading of MWCNTs, three phase structures such as sea-island phase, co-continuous phase and inverse phase were exhibited by MWCNT/PEI/BD composites with an increase in PEI content. 0.4 MWCNT/10 PEI/BD composite with sea-island structure exhibited a dielectric constant as high as 600 with low dielectric loss⁵.

Nanya Li et al. (2014) made attempts to reduce in cure-induced strains and deformation of carbon fiber reinforced BMI composites by an innovative microwave curing. One strategy that was attempted earlier was conventional thermal curing with disadvantages of fiber/matrix coefficient of thermal expansion (CTE) mismatch, heat transfer and convection. Nearly reduction in 95% of cure-induced strains has been achieved by microwave curing process⁶.

FGS-BMI-BA nanocomposites were fabricated by Muchun Liu et al. (2014). Functionalised grapheme nanosheets (FGS) were prepared by diazonium treatment followed by reduction with hydrazine hydrate. These composites exhibited a notable increase in impact strength, flexural strength and thermal stability by the addition of FGS⁷.

The effect of surface-functionalized reduced graphene oxide sheets, obtained by grafting the hyperbranched polytriazine (HBPT) onto the surface of reduced graphene oxide (GO) sheets, on the mechanical and tribological properties of HBPT-RGO-BMI-BA (diallyl bisphenol A modified bismaleimide resin) were investigated by Chao Liu et al. (2015). At 0.6 weight % loading of HBPT-RGO, the composites displayed better mechanical properties, thermal stability and more stable frictional

coefficient making them wear-resistant composites applicable as high performance composites in many fields⁸.

The effect of distilled water, propylene glycol and a commercially available deicing fluid on dielectric properties were investigated by Luis A. Rodriguez et al. (2015) at X band frequency for three-ply quartz reinforced BMI composites. An increase in 1 weight % of water, deicing fluid and propylene glycol separately in the above fabricated BMI composites results in a 7.85%, 4.5% and 2.5% increase in dielectric permittivity due to water, deicing fluid and propylene glycol. The observation results are described and discussed in detail and they concluded that the use of these common fluids in service aircraft radar systems leads to dielectric property degradation which requires significant concern because the aircraft systems are routinely exposed to these fluids⁹.

BDDP was prepared from 4,4'-bismaleimidodiphenyl methane (BDM), 2, 2'-diallyl bisphenol A (DBA) and [(6-oxido-6H-dibenz [c,e] [1,2]oxaphosphorin-6-yl)-methyl]butanedioic acid (DDP). Glass fiber reinforced (GF)-BMI and GF-BDDP composites were developed by Xiang Xiu Chen et al. (2015). Both the composites exhibit excellent dielectric stability. The lower dielectric loss of GF-BDDP may be due to low dielectric constant and loss of BDDP along with stronger interfacial adhesion¹⁰.

Zhao Yian et al. (2015) proposed a formulation to predict the long term durability of E glass fiber reinforced bismaleimide composites in a sea water environment. Lowering of T_g , accelerated degradation of storage modulus and shortening in service life were exhibited by the E glass fiber reinforced BMI composites with sea water absorption¹¹.

Comparative studies on flexural strength, ILSS and physical properties of microwave cured vacuum bagged carbon fiber-BMI composites with those fabricated by autoclave curing and conventional oven curing. Xuehong Xu et al. (2015) reported that overall cure cycle time was reduced for microwave cured carbon fiber-BMI composites compared with those fabricated using other thermal processing¹².

To produce a high performance polymer, Hongxia Yan et al. (2015) synthesized hyperbranched polysilane grafted reduced graphene oxide (HBPSi-rGO) in bismaleimides. From the systematic study, it is inferred that the addition of HBPSi-rGO into BMI resin not only enhance the mechanical properties like impact and flexural strength but also the thermal stability of the composites. At 0.6 weight % of

loading of HBPSi-rGO, the functional coefficient and the wear rate was decreased significantly¹³.

BMI composites with hybrid nanoparticles consisting of reduced graphene oxide and MoS₂ nanosheets with active amino groups (NH₂-rGO-MoS₂) were developed by Zhengyan Chen et al. (2016). Researchers have reported that the inclusion of 0.6 weight % of the hybrid nanoparticle leads to enhancement in impact and flexural strength. The developed composites exhibited better mechanical, thermal, tribological properties, lower friction coefficient, volume wear rate and enhanced toughness that may be ascribed to the strong interfacial adhesion between NH₂-rGO-MoS₂ and BMI matrix and the synergistic effect of r-GO and MoS₂. These claimed properties make the composite an attractive candidate in the fields where friction reducing and wear resistance composites finds application¹⁴.

Junwei Gu et al. (2016) designed POSS-g-nBN/BMI/DABA nanocomposites for employing it in the integration and miniaturization of microelectronic devices. Surface functionalization of nanosized boron nitride (nBN) was done using γ -glycidoxy propyl trimethoxy silane / polyhedral oligomeric silsesquioxane (KH-560/POSS). The influence of volume fraction of nBN and POSS-g-nBN on dielectric and thermo-mechanical properties of BMI/DABA nanocomposites was also investigated in detail. Upto 428°C, the composites exhibited only 5 % weight loss and gave a detailed account of their outstanding thermal stability. Composites exhibited excellent dielectric properties, high thermal conductivity and outstanding thermal stability at 15.4 volume % of POSS-g-nBN¹⁵.

The effect of BMI content, curing and rheological behaviour of allyl-functionalized phthalonitrile (DABA-Ph) and bismaleimide (BMI) have been investigated in detail by Mingli Jiang et al. (2016). Good processability and thermal stability of DABA-Ph/ BMI copolymer pave the way to fabricate glass fiber reinforced DABA-Ph/BMI composite laminates. DABA-Ph/BMI composites demonstrated remarkable improvement in mechanical properties along with satisfactory thermal stability and stable dielectric constant over a wide range of frequencies so that it could be explored in industrial applications¹⁶.

Co-precipitation and precipitation polymerization process was employed to synthesise a ternary nanoparticle with reduced graphene oxide sheets (GNS), Fe₃O₄ nanoparticles and highly crosslinked polyphosphazene and coded as (GNS- Fe₃O₄@ PZM). Chao Liu et al. (2016) synthesized and characterized three types of nanocomposites such as GNS-Fe₃O₄, GNS-Fe₃O₄@PZM and aligned GNS- Fe₃O₄@PZM/BMI composites. At 0.4 and 0.8 loadings, GNS-Fe₃O₄@PZM exhibited improved mechanical properties such as impact strength and flexural strength, excellent thermal stability along with friction-reducing and anti-wear properties¹⁷.

The effect of loading rate on tensile behaviour of pristine CNT film, random CNT/BMI and oriented CNT/BMI composite films were investigated in detail by Mengsi Zhang et al. (2016). With the increased loading rate, the tensile strength of pristine CNT film decreases while that of random and oriented CNT/BMI composite films are found to be increased. With the increase of loading rate from 0.05mm/min to 40mm/min, there is a significant increase in tensile strength of random and oriented CNT/BMI composite films. The dependence of tensile behaviour of CNT/BMI films at different loading rates may be due to the different induced motions of CNTs in the films¹⁸.

Jing Zhou et al. (2016) prepared carbon fiber/BMI composite for high potential applications in high aerospace structures with improved fracture toughness by employing microwave processing technique. Microwave curing cycle time was cut to 44% compared to thermal curing. The improvement in interlaminar fracture toughness due to enhanced interfacial strength was exhibited by microwave-cured carbon fiber /BMI composites¹⁹.

The effect of temperature on mechanical properties of carbon fiber/BMI composites was studied by Baifeng Yang et al. (2017) by testing the fabricated composites in a wide range of temperature conditions from -120 °C to 200 °C. There is a significant increase in transverse and longitudinal compressive strength while it drops sharply with rise in temperature²⁰.

Phosphorus-containing polyhedral oligomeric silsesquioxane functionalized graphene oxide (P-POSS-GO) / Phosphorus-containing polyhedral oligomeric silsesquioxane (P-POSS) / 4,4'-bismaleimidodiphenyl methane (DBMI) composites were developed and the effect of P-POSS-GO content on mechanical, flame retardant and thermal properties of the composites were investigated in detail by Cheng Tang et al. (2017). Excellent flame retardancy, thermal stability and improved mechanical properties are exhibited by the composites containing P-POSS and modified Graphene oxide (GO). Improvement of flame retardancy may be due to the increase of degradation activation energy or growth of oxygen insulation and heat insulation char layer facilitated by the added P-POSS-GO²¹.

Carbon /BMI composites were fabricated by the out-of-autoclave (OOA) technique and their hygrothermal ageing was investigated in detail by Z. Huo et al. (2017) under seawater conditioning at elevated temperatures of 50 °C and 90 °C for approximately three months. Interfacial crack, identified from SEM analysis of cross section of hygrothermally aged BMI composites resulted in reduction in flexural strength and ILSS and it is more severe at 90 °C than that at 50 °C²².

BMI nanocomposites were prepared with NH₂-rGO / ZnO hybrid nanosheets with loadings ranging from 0.2 to 1.0 weight % by Zhengyan Chen et al. (2018). A simple and green solvothermal method was adopted by the researchers to synthesize r-GO/kapok like ZnO hybrid sheets. BMI nanocomposites with 0.8 weight % of the hybrid nanosheets demonstrate enhanced thermal resistance and highest impact and flexural strength, this makes the composite highly attractive for high temperature performance applications²³.

Graphene oxide synthesized by the Hummers method was reduced separately using three different reductants like hydrazine hydrate (HHA), Chitosan (CS) and KOH. Amino functionalized reduced GO (NH₂IL-rGO) was obtained by modification using ionic liquid (NH₂IL). The MBAE matrix was synthesized from 4,4'-diamino diphenyl methane bismaleimide modified by diallyl bisphenol A and bisphenol A bisallyl ether by Yufel Chen et al. (2019). The effect of incorporation of NH₂IL-rGO on mechanical properties and dielectric constant were studied in detail. Experimentally obtained results indicate that at 2 weight % of the filler, the composite exhibited maximum impact strength and flexural strength, dielectric constant of 84 and thermal decomposition temperature of $435 \, {}^{\circ}C^{24}$.

2.3. Epoxy composites

Epoxy resin has been used in a variety of applications in aerospace industries, adhesives, coatings etc. The brittle nature, poor weathering resistance and impact strength of epoxy resin restricts its high performance applications. Researchers attempted to improve the thermal and electrical properties by modifying epoxy resin with inorganic and organic particles, blending with other resin matrix has been proved to be an efficient way to enhance the performance of epoxy resin. Previous studies and current research literature are reported here.

The effects of various parameters like heat treatment temperature, holding time, grinding effect at different heat treatment temperatures, semiconducting and barrier-layer treatments on the properties of epoxy composite with untreated BT filler and

those with heat treated BT fillers were investigated by Dong–Hau Kuo et al. (2000). An increase in dielectric constant was shown by epoxy composite with semiconducting and barrier layer treated BT filler. A significant increase in dielectric constant was shown by epoxy composites with maximized BT ratio and these multi doped epoxy composites act as promising candidates for application in printed wiring boards²⁵.

Different types of carbon fiber/epoxy composites were prepared separately by C. Nightingale and R. J. Day (2002) using resin–hardner system with higher and lower dielectric loss factors and also employing different processing routes. The flexural strength, interlaminar strength and void content were also measured and a comparison has been made between the two epoxy composites²⁶.

A detailed explanation of the variation of dielectric properties as a function of composition, frequency and temperature obtained by the addition of Al powder in epoxy resin during the fabrication of epoxy composites were given by Vishal Singh, A. R. Kulkarni and T. R. Rama Mohan (2003). An increase in dielectric constant and tan δ of Al–epoxy composites with an increase in volume % of aluminium and temperature may be due to interfacial polarization and orientation of dipoles facilitated by the increased segmental mobility of the polymer respectively²⁷.

Temperature, frequency and volume % dependence of dielectric constant and dielectric loss of the self synthesized BT(SS-BT), commercial BT and lead magnesium niobate (PMN) ceramic filler incorporated epoxy composites was dealt in detail by Dong–Hau Kuo et al. (2004). Among these three ceramic/epoxy composites with perovskite type ceramic filler, the one loaded with SS-BT showed the highest dielectric constant of 44²⁸.

Lai Qui et al. (2005) designed an epoxy composite containing randomly distributed silver nanoparticles for embedded capacitor applications. The high dielectric constant and low dielectric loss of fabricated composites make it a potential candidate to be used as an embedded dielectric²⁹.

Fen Chao et al. (2007) prepared a series of thermosetting polymer/BaTiO₃ (BT) composites using cyanate resin, bismaleimide resin and epoxy resin as thermosetting resin matrices. A detailed explanation of dielectric studies was done and the experimental data were in good agreement with dielectric constants predicted according to the Lichtenecker mixing rule. The experimental results show that the dielectric constants and the dielectric loss of bismaleimide/BT composite and epoxy/

BT composite increase with the increase in BT content. Cyanate/BT composite also exhibited an increase in dielectric constant and decrease in dielectric loss with the increase in BT content. They also concluded that cyanate/BT composites showed more stability and excellent dielectric properties as compared to the other composites³⁰.

Zhi-Min Dang et al. (2007) fabricated epoxy/BT micro composites with differently loaded and microsized BT particles 0.1 μ m and 0.7 μ m coded as BT-01 and BT-02 with cubic and tetragonal crystal phase respectively. Use of an optimum amount of silane coupling agent KH 550 enhances miscibility between the microfiller and the epoxy resin. In this paper, they have reported a detailed investigation on the effect of size of BT and the use of silane coupling agent on the dielectric properties and microstructures of the epoxy/BT composites. Epoxy composites loaded with BT particles with 0.7 μ m size and tetragonal phase showed an increase in dielectric constant of the composites³¹.

Epoxy/BaTiO₃(BT) composites were prepared by dispersing barium titanate, Y5V fillers within DGEBA type epoxy resin, D.E.R. 331, at about 70 °C, followed by the addition of diaminodiphenylmethane (DDM) as the curing agent. The dependence of dielectric constants of the cured composites with various volume % of Y5V fillers and frequency was also analysed by Kuo-Chung Cheng et al. (2007). A model chip antenna was made and packaged with epoxy resin Y5V composite and measured the return loss S11 using a vector network analyzer and based on the results of these observations, it was suggested that this composite could be applied in wireless communications using 204GHz ISM band³².

In order to achieve good dispersion and high compatibility of polyaniline (PANI) with the epoxy resin, the in-situ polymerization method was adopted by Jiongxin Lu et al. (2007) to fabricate PANI/epoxy composite. The dielectric constant of these composites reached a high value close to 3000 and showed a dielectric loss value less than 0.5. Improved dielectric performance of nanocomposites could be tailored by PANI loading levels and appropriate hardner selection³³.

The effect of silane concentration on the dielectric properties and microstructure of epoxy/BaTiO₃ (BT) composites were investigated in detail by L. Ramajo et al. (2007). High silane concentrations resulted in composites with increased porosity and defects with poor dielectric properties. Low silane concentration helps to retain the dielectric properties of the composites³⁴.

Singlewalled carbon nanotube (SWCNT) buckypaper, multiwalled carbon nanotube (MWCNT) buckypaper and carbon nanofiber (CNF) were incorporated onto the surface of carbon fiber reinforced epoxy composites by employing the hand layup method followed by vacuum bagging. In the investigation of flame retardant efficiencies by Qiang Wu et al. (2010). It was found that the epoxy incorporated MWCNT buckypaper showed excellent flame retardancy that may be attributed to the thermo-oxidation stability and low gas permeability³⁵.

The carbon fiber/epoxy composite laminates were prepared by A.Y. Zhang et al. (2011) from tapes made of T 300 carbon fibers pre-impregnated with Hexcel 914 epoxy resin using vacuum bagging and curing method. The effects of parameters like void contents, immersion time on the absorption/desorption behaviour, mechanical properties were also studied. The experimental results show that with the increase of porosity and immersion time, there is an increase in mechanical properties like compressive strength, bending strength and interlaminar strength(ILS) of the aged, unaged and dried samples³⁶.

Surface modification of Al particles was carried out by using four different kinds of coupling agents like silane KH550, Silane KH560, titanate NXT-102 and titanate NXT-201. The effect of these modified Al particles on the microstructure and dielectric properties of the fabricated Al/epoxy composites were analysed in detail by Wenying Zhou and Demei Yu (2011). The epoxy composites incorporated with silane KH560 treated Al particles showed high dielectric constant and low dielectric loss and found application in engineering³⁷.

The effect of loading functionalized and amino functionalized carbon nanotubes (CNT) on the carbon fiber reinforced epoxy composites were investigated in detail by M. Sanchez et al. (2012). The obtained experimental data showed a slight decrease in tensile strength of CNT-NH₂/epoxy composites as compared with CNT/epoxy ones. The presence of agglomerates which act as micro-centres of stress concentration may be the reason for the decrease in tensile strength. The increase in flexural strength and intra and interlaminar strength for the amino functionalised CNT loaded epoxy composites may be attributed to the effective interfacial interaction between the nanofiller and epoxy matrix³⁸.

The influence of high loading lamellar α -Al₂O₃ microparticles on the thermal and dielectric properties of the fabricated epoxy composites was investigated in detail by Xinfeng Wu et al. (2012) and the results obtained are compared with epoxy/Al₂O₃

composites in their previous work. The results show that thermal conductivity and dielectric properties like dielectric constant of the composites were enhanced and there is a reduction in electric resistance and dielectric loss³⁹.

Jin-Ping Peng et al. (2012) fabricated the composites by dispersing the pristine multiwalled carbon nanotubes (p-MWCNTs) and ozone functionalized multiwalled carbon nanotubes (f-MWCNTs) by using methods including three roller mills and high speed stirring in epoxy resin. At the same or fixed filler content, the dielectric constant of MWCNT/epoxy resin composites gradually decreases with an increase in frequency. It was found that the dielectric constant of functionalized MWCNT composites, compared to the pristine ones, was lower at the same content. The composite with high dielectric constant finds use in high energy storage application areas such as mini capacitors⁴⁰.

Surface modification of magnetite (Fe₃O₄) fillers was done using a silane coupling agent and varying treatment time. Fabrication and characterization of magnetite filled epoxy polymer composites have been made and the results were described and discussed in detail in the paper by M. S. Boon and M. Mariatti (2013). This study helps not only to identify the effect of various parameters like silane concentration and treatment time on the magnetic and dielectric properties of magnetite filled epoxy composites but also helps in the optimization of these properties based on the design of the experiment (DOE)⁴¹.

D. Bychanok et al. (2013) fabricated epoxy composites by using three different fillers – carbon black (CB), multiwalled carbon nanotubes (MWCNT) and singlewalled carbon nanotubes (SWCNT) separately. Their detailed analysis helps in identifying the mechanisms that contribute to polarization in these composites which in turn act as a guide for predicting the effective permittivity of the composites with different filler particles⁴².

H. A. Avila et al. (2013) synthesized nanostructured $BaTiO_3$ (BT) fibers by electrospinning method. The composites were prepared by alternating layers of BT nanofibers in an epoxy resin matrix. The influence of the electrode configuration with respect to the BT nanofiber layers on the dielectric properties have been observed and analysed. The obtained experimental data confirms that epoxy/BT composites with perpendicular electrode configuration exhibited higher dielectric permittivity than those obtained for the one with parallel electrode configuration⁴³.

H.Li et al. (2013) fabricated composites with a 1-3 type structure obtained by introducing $Ni_{0.5}Zn_{0.5}Fe_2O_4$ (NZO) ring ceramic rod array in the epoxy matrix. A detailed analysis of frequency dependence of the magnetic and dielectric properties, modelling and simulation analysis as well as comparison with the conventional 0-3 type composites were described and discussed⁴⁴.

Synthesis of a series of titanium dioxide (TiO₂) coated titanium dibromide (TiBr₂) particles with different degrees of coverage were achieved by using the oxidation method and coded as O_n -TiB₂ by Long Cheng Gu et al. (2013). TiBr₂/epoxy and O_n -TiBr₂/epoxy composites were fabricated using both microwave and thermal curing procedures. The influence of various parameters like structure of O_n -TiBr₂, oxidation time and content of O_n -TiBr₂, especially 20 min oxidized O_{20} -TiBr₂ on the dielectric constant value were investigated in detail. The O_{20} -TiBr₂-epoxy composites with 25 volume % O_{20} -TiBr₂ exhibited a dielectric constant value of 407 and low dielectric loss at 1 Hz indicating that this composition is more adaptable in high dielectric applications⁴⁵.

Effect of surface bamboo gradient loadings on epoxy composites with virgin bamboo cellulose fiber (BCF), NaOH treated bamboo cellulose fiber (N-BCF) and silane coupling agent (KH560) treated bamboo cellulose fiber (K-BCF) were fabricated and comparative studies based on the effects of modification of cellulose fibers on the mechanical properties were done by Tingju Lu et al. (2013). The K-BCF/epoxy composites exhibited better mechanical properties than those of BCF and N-BCF incorporated epoxy composites⁴⁶.

The remarkable enhancement of dielectric constant and thermal conductivity at low volume loaded graphite nanoplatelets (GNPs) embedded epoxy composites have been reported by Chao Min et al. (2013). Fabrication of covalent bonding in the interface region by the layers of GNPs and providing conductive channels for thermal support by the inner graphene layers are responsible for the improved dielectric constant and thermal conductivity⁴⁷.

A detailed comparison of dielectric performance has been made between in–situ Cu-epoxy nanocomposites and in-situ–BT/Cu-epoxy nanocomposites by Gang Li et al. (2014). The strong interfacial adhesion between the polymer matrix and nanofiller as well as uniform dispersion of the nanofiller played a vital role in the enhancement of dielectric properties⁴⁸.

The effect of incorporation of polyhedral oligomeric silsesquioxanes (POSS) and cubic boron nitride in epoxy resins was investigated by Thomas Heid, Eric David et al. (2015).Epoxy composites with glycidyl-POSS showed enhanced dielectric properties like breakdown strength and dielectric loss as well as thermal conductivities compared to the neat ones⁴⁹.

Epoxy/graphene composites were fabricated by Qing Hou et al. (2015) by embedding graphene nanoplatelets (GNP) into epoxy resin. The uniform dispersion of GNP in epoxy matrix was enhanced by cryomilling of GNP at liquid nitrogen temperature. Results show that there is an increase in dielectric constant with an increase in weight % of GNP⁵⁰.

A detailed comparison of the physical and mechanical properties has been made between epoxy composites reinforced with kevlar pulp and glass fiber by Simona Matei et al. (2015). It has been inferred that epoxy composites with glass fiber ensure superior mechanical properties like tensile and flexural strength compared with kevlar pulp⁵¹.

The influence of hybridization on mechanical and damping properties exhibited by the kevlar fiber, glass fiber and hybrid Kevlar/glass fiber reinforced epoxy composites has been studied by Mehmet Bulut et al. (2015). The results obtained as numerical and experimental damping vibration responses of samples, tensile strength, damping ratio, storage modulus, loss modulus etc., were described and discussed in this paper. From the results it is concluded that tensile strength damping vibration characteristics of the composites are strongly affected by the hybridization⁵².

Binghao Wang et al. (2015) prepared composites with a series of multilayer composed of multiwalled carbon nanotubes and epoxy resin coded as (MWCNT/EP)_n where n =1-5 by using layer-by-layer casting technique. The dielectric performance and mechanism of these composites were studied in detail. It was found that composites exhibited the highest dielectric constant and low dielectric loss that may be attributed to the space charge polarization (SCP) and blocked transport carriers between layers⁵³.

Carbon fiber/epoxy composites were fabricated by Xuehong Xu et al. (2015) using different processing techniques like vacuum bagging using microwave and thermal curing to improve the compressive strength of the composites and comparison of physical and mechanical properties of the composites were done. Multistep (2 steps or 3 steps) microwave curing process was adopted for further improvement of the

compressive strength of the composites. The obtained experimental data shows that there is an enhancement in specific compressive strength that may be ascribed to strong interfacial bonding between the epoxy matrix and carbon fibers⁵⁴.

Surface modification of Ca and Zr doped $BaTiO_3$ (BCZT) particles were done by Bing cheng Luo et al. (2015) using dopamine and these modified dopamine@BCZT particles were incorporated into the epoxy resin via melt mixing method to get the resultant dopamine@BCZT/EP composites. It was found that the dielectric constant of the fabricated composites increases by eight times when compared with that of neat epoxy resin when the dopamine@BCZT was 25 volume %⁵⁵.

Yong Hu et al. (2016) adopted new processing techniques like gel casting, sintering and vacuum filtration methods to fabricate differently loaded α -Al₂O₃ epoxy composites which find application in electronic packaging. Effects of Al₂O₃ content and the sintering temperature on the thermo-mechanical and dielectric properties were dealt in detail and compared with their previous work⁵⁶.

The effect of donor doped soft lead zirconate titanate (PZ27) and acceptor doped hard (PZ26) filler were incorporated into an epoxy resin and investigations have been made by Jibran Khaliq et al. (2016) to study the effect of piezoelectric performance and ageing in epoxy-PZ27 and epoxy-PZ26 composites. It has been inferred that the dielectric constant of the piezoelectric materials used as filler plays an important role in determining the piezoelectric performance of the composites⁵⁷.

K.Krishnadevi and V.Selvaraj (2016) synthesized functionalized rice husk ash (FRHA) by the treatment of silane coupling agent 3-aminopropyl tri methoxy silane (3-APTMS) on rice husk ash. The synthesized FRHA and amine terminated cyclophosphazene (ATCP) were incorporated into epoxy resin to fabricate the composites. These incorporated materials not only enhance flame retardant properties but also increase the dielectric properties of the developed epoxy composites, which makes the composite find applications in micro electronics⁵⁸.

Effect of different loadings of graphene oxide (GO), reduced graphene oxide (RGO) and diglycidyl ether of bisphenol-A (DGEBA) grafted onto RGO (DGEBA- RGO) in epoxy composites on dielectric properties were investigated in detail by Yan-Jun Wan et al. (2016). From the experimental data, it has been inferred that epoxy with DGEBA-RGO exhibited higher dielectric constant and low dielectric loss that may be ascribed to the strong interaction between the epoxy matrix and the DGEBA-RGO filler as well as the effective dispersion of the filler in the matrix⁵⁹.

Solvent spraying techniques were employed to reduce the agglomeration of CNT and to enhance the uniform dispersion of the incorporated MWCNT during the fabrication of carbon fiber reinforced epoxy composites. The investigation based on the data collected by J. A. Rodriguez Gonzalez et al. (2017) and its further detailed interpretation showed that there is a significant improvement in interlaminar fracture toughness of the epoxy composites modified with sprayed MWCNT⁶⁰.

Epoxy composites incorporated either with exfoliated boron nitride (EBN) nanoplates or nanodiamond attached EBN nanoplates (NDEBN) were prepared separately by Yinhang Zhang et al. (2017) with varying concentrations of EBN and NDEBN and their physical and thermal properties were studied. EP/NDEBN composite exhibited higher thermal conductivity compared to EP/EDN. This may be due to the intrinsic super thermal conductivity of nanodiamond⁶¹.

The influence of incorporation of alumina/polydopamine (AO), core/shell alumina/ polydopamine (AO*) and strawberry-like core/shell structured alumina/polydopamine /silver (AO*@Ag) in an epoxy matrix on the dielectric properties have been studied in detail by Zhengdong Wong et al. (2018). Among the three epoxy composites, the AO*@Ag composites show higher dielectric permittivity and breakdown strength. The incorporation of strawberry-like core/shell structured alumina/polydopamine/ silver (AO*@Ag) in the epoxy matrix enhances the thermo-mechanical properties⁶².

Ling Weng et al. (2018) reported the comparative studies based on thermomechanical and dielectric properties of epoxy composites incorporated either hexagonal boron nitride (h-BN) or with cubic boron nitride (c-BN). Composites with h-BN exhibited higher thermal conductivity whereas c-BN showed slightly higher dielectric constant⁶³.

Based on the variation in dimension of lead zirconate titanate (PZT) ceramic phases, two kinds of 2-2-2 connectivity PZT/epoxy composites were designed for application in the field of ultrasonic transducer by Shanling Yi et al. (2018)⁶⁴.

2.4. Bismaleimide-epoxy composites

Research attempts have been made to improve the toughness, thermo-mechanical and dielectric properties of epoxy resin by adding bismaleimide resins with higher crosslinking ability and thermal stability than epoxy. Researches based on this area are of great interest and a literature review of BMI-epoxy composites is reported below.

The epoxy polymer, tetraglycidyl 4,4'-diamino diphenylmethane (TGDDM) was cured with 4,4'-diamino diphenyl sulphone (DDS) by E. M. Woo et al. (1987). An intercrosslinked network was formed during curing with no phase separation confirmed by DMA and SEM analysis. This blended matrix can be used for fabricating high performance composites with an intermediate service temperature⁶⁵. Allyl modified epoxies (AE) were synthesized by the reaction of epoxy with 2-allyl-4-methyl phenol by Aijuan Gu and Guozheng Liang (1997). BMI resins modified with AE (BMI/AE systems) exhibit high thermal stability, improved toughness and

A typical tetraglycidyl 4,4'-diamino diphenylmethane (TGDDM) and diamino diphenyl sulfone (DDS) blended with BMI was prepared and investigated by P. Musto et al. (1998). The addition of BMI to the TGDDM–DDS blend enhances the flexural elastic modulus and compressive yield stress while reducing the equilibrium water uptake⁶⁷.

outstanding tack and drape properties⁶⁶.

A. Ashok Kumar et al. (2000) prepared siliconized epoxy resin by treating it with hydroxyl-terminated polydimethylsiloxane (HTPDMS) with γ -aminopropyltriethoxy silane (γ -APS) as a compatibilizer and dibutylindilaurate as a catalyst. The systems were further modified by BMI and cured by diaminodiphenylmethane (DDM). Comparative studies based on thermal properties were done. HTPDMS modified BMI-epoxy systems exhibited higher thermal stability as well as heat distortion temperatures⁶⁸.

The influence of void content percentage, size and shape of the voids on the interlaminar shear strength (ILSS) of carbon fabric reinforced epoxy and carbon fabric reinforced BMI composites were analyzed in detail by Michelle Leali Costa et al. (2001). From the observations, it is clear that triangular shaped voids are created at the crossing of the woven fabric in the case of carbon fabric/epoxy laminates whereas in carbon fabric/BMI laminates, voids are present at the interface of woven fabric. Both composites with void content above 0.9 % showed a decrease in interlaminar shear strength⁶⁹.

Effect of unsaturated polyester content as well as BMI content on tensile modulus, flexural modulus, fracture toughness and Tg were studied in detail by K. Dinakaran and M. Alagar (2001) for which they developed unsaturated polyester (UP)–bismaleimide (BMI) modified epoxy matrix systems. Epoxy/UP/BMI system

with compositions in the ratio 100:30:15 exhibited higher mechanical and thermal properties⁷⁰.

A. Vanaja, R. M. V. G. K. Rao (2002) prepared and characterized epoxy-novolac resin (EPN)/BMI blends. From the DSC scans, it is clear that with the increase in BMI content, there is an enhancement in Tg due to higher crosslinking density attributed to the homopolymerization of BMI. With the addition of BMI to the EPN system, there is an improvement in thermal stability⁷¹.

The presence of BMI in TGDDM/DDS/BMI system decreased the equilibrium water uptake, enhanced the diffusivity and reduced the activation energy for diffusion were investigated by Pellegrino Musto et al. (2002) from gravimetric sorption measurements and near infrared (NIR)-FTIR techniques. Their assignments were also supported by a dual-mode model for diffusion and the estimated ratio for free and bound water was in good agreement with that obtained from the spectra⁷².

Epoxy-cyanate ester (CE), epoxy-BMI and BMI-CE-epoxy matrices were fabricated and their mechanical and thermal properties were investigated by K. Dinakaran et al. (2003). An increase in both BMI and CE content resulted in epoxy composites with improved thermal stability, enhanced degradation temperature and reduced moisture absorption. Epoxy/CE system showed increased flexural and impact strength with a marginal reduction in tensile strength whereas epoxy/BMI system shows enhanced tensile and flexural strength with a reduction in impact strength⁷³.

Ru-Jong Jeng et al. (2003) designed 3,3'-bis(maleimidophenyl)phenyl phosphine oxide (BMPPPO) embedded epoxy resin for application in microelectronics. The introduction of phosphorus-imido containing BMI (BMPPPO) into the epoxy resin exhibits higher thermal properties and excellent flame retardancy⁷⁴.

A detailed comparison has been made between polyurethane modified epoxy, BMImodified polyurethane epoxy, BMI-modified epoxy and unmodified epoxy on the thermal and electrical properties by K. P. Ochathevar Mahesh et al. (2003). BMImodified polyurethane epoxy systems exhibited increased electrical properties and thermal properties⁷⁵.

K. P. O. Mahesh et al. (2003) prepared different kinds of epoxy systems which include polyurethane-epoxy matrices, BMI-epoxy matrices, BMI modified polyurethane -epoxy matrices and a detailed comparison has been made based on their mechanical, thermal and morphological behaviors. Bismaleimide modified polyurethane-epoxy matrices show an increase in thermal stability, tensile and

flexural strength, decrease in impact strength, T_g and heat distortion temperature (HDT) whereas polyurethane incorporated epoxy matrices exhibited increased mechanical properties and decrease in T_g , thermal stability and HDT. SEM micrographs revealed heterogeneous microstructure of polyurethane modified epoxy and bismaleimide modified polyurethane epoxy system and homogeneous microstructure of bismaleimide –epoxy systems⁷⁶.

The influence of introduction of diglycidylether terminated polydimethylsiloxane (DGETPDMS) to bismaleimide (BMI) as well as both DGETPDMS and BMI into the epoxy resin matrix on the thermo-mechanical properties and the morphology of the developed matrices were investigated in detail by R. Suresh Kumar and M. Alagar (2005). Introduction of BMI into epoxy resin enhances both mechanical and thermal properties with BMI content whereas incorporation of DGETPDMS improves impact strength thereby reducing tensile strength, flexural strength and T_g. DGETPDMS-BMI-epoxy matrix exhibited enhanced thermo-mechanical properties according to their percentage content of the incorporated DGETPDMS and BMI⁷⁷.

Aromatic and aliphatic bismaleimides were separately incorporated into the polyurethane-epoxy intercrosslinked system by K. P. O. Mahesh et al. (2006). The thermal stability, T_g and electrical properties of aromatic bismaleimide embedded polyurethane –epoxy matrices increased with increasing the BMI content. This system is more adaptable for fabricating composites for industrial and engineering applications⁷⁸.

Laleh Rajabi and Ghodratollah Malekzadeh (2006) investigated the effect of BMI resin on the dielectric and mechanical properties of the prepared glass fabric DGEBPA-MDA (Methylene dianiline) and DGEBPA-DDS (Diaminodiphenyl sulfone) laminates and the results showed that there is a considerable increase in T_g , flexural strength and shear storage modulus⁷⁹.

Siliconized epoxy aliphatic bismaleimide and siliconized epoxy aromatic bismaleimide intercrossed network matrices were developed by M. Alagar et al. (2007). Thermal and mechanical properties of the fabricated matrices were compared with unmodified epoxy matrices and siliconized epoxy systems. The fracture toughness increases with an increase in siloxane content and decreases with an increase in BMI content. The matrix with aromatic bismaleimide resin exhibited higher mechanical properties and this system can be used to fabricate advanced composite for aerospace and engineering applications⁸⁰.

Effects of incorporation of polysulfone and various types of bismaleimides such as N, N'-bismaleimido-4,4'-diphenylmethane (BMI-1), 1,3-bis(maleimido)benzene (BMI-2) and 1,1'-bis(4-maleimidodiphenyl) cyclohexane (BMI-3) into epoxy matrix has been investigated in detail by R. Rajsekaran and M. Alagar (2007). The introduction of polysulfone into epoxy resin enhanced impact strength whereas the introduction of BMI improved both tensile and flexural properties. The epoxy matrix with 8% polysulfone and 8% BMI-2 exhibited excellent mechanical properties than other modified epoxy systems indicating that this composition can be used for the fabrication of advanced composites in industrial and engineering applications and the matrix system as such can also be used in the form of adhesives and coatings⁸¹.

Rajangam Rajasekaran (2008) prepared Polyethersulfone (PES)-epoxy matrices, BMI-modified epoxy matrices and PES-epoxy-BMI matrices. The thermal properties of the developed matrices have been compared with those of unmodified epoxy systems. PES-epoxy-BMI matrices exhibited enhanced thermal stability, mechanical properties and degradation temperature. With the increase in PES and BMI content, these matrices show an increase in water absorption resistance. The results were described and discussed in detail and concluded that the PES-epoxy-BMI matrix could be used for the fabrication of high performance composites⁸².

In order to fabricate advanced composites with improved toughness and excellent thermo-mechanical properties for engineering applications S. Premkumar et al. (2008) made attempts to develop caprolactum blocked methylene diphenyl diisocyanate (CMDI) and BMI modified epoxy resin systems. It was observed that the incorporation of BMI has increased thermal properties like heat distortion temperature (HDT), T_g and thermal stability along with a significant increase in tensile and flexural strength and decrease in water absorption⁸³.

Li Yuan et al. (2008) reported that the inclusion of microcapsules filled with epoxy resins (MCE) to BMI-diallyl bisphenol A (BA) system could lead to enhancement in flexural strength, impact and water resistance with a slight decrease in initial thermal decomposition temperature (T_d). The mechanical properties of BMI-BA systems were influenced by the size and the content of MCE⁸⁴.

Three kinds of PUF microcapsules filled with bisphenol-A epoxy resins (MCE) were prepared with mean diameters of 200, 140 and 86 μ m respectively and investigated the effect of inclusion of these into fiber reinforced BMI-BA composites by Li Yuan et al. (2010). From the experimental data, it is inferred that with the same MCE

content, the composite with a smaller MCE diameter exhibited better mechanical properties compared to others that may be attributed to the large surface area provided by MCE with a small diameter that may increase the interfacial interaction between MCE and the matrix⁸⁵.

Carbon fiber fabric (IM7), epoxy-IM7, epoxy-IM7-SWCNT-buckypaper (BP), epoxy-IM7-MWCNT-BP, BMI-IM7-SWCNT-BP, BMI-IM7-MWCNT-BP composites were fabricated and comparative studies based on flame retardancy has been made by Qiang Wu et al. (2010). After combustion of epoxy-IM7-SWCNT-BP, SWCNT-buckypaper was burnt out completely without affecting the composite, in the case of BMI-IM7-SWCNT-BP, due to the synergic effect of BMI and SWCNT-bucky paper (BP) partially survived indicating that SWCNT-BP can act as an excellent fire retarding shield⁸⁶.

The influence of incorporation of bismaleimide containing phthalide cardo structure (BMIPP) in diglycidyl ether of bisphenol A epoxy resin (DGEBA) in the presence of 4,4'-diamino diphenyl sulfone (DDS) as hardner have been investigated in detail by Xuhai Xiong et al. (2010) and a detailed comparison has been made on the thermal, mechanical, physical properties and also on the curing behavior. The blend showed increased fracture toughness and maximum impact strength at 5 weight % loading of BMIPP. The blend also exhibits improved thermal stability, reduced equilibrium water uptake as well as storage modulus with decreased T_g^{87} .

A new bismaleimide monomer,2-((4-maleimido phenoxy) methyl)-5-(4-maleimido phenyl)-1,3,4-oxadiazole (Mioxd)–modified epoxy resins with various concentrations of mioxd were developed and their curing behavior as well as thermal properties were studied by Xuhai Xiong et al. (2011). The results showed that there is an increase in both T_g and storage modulus⁸⁸. Siliconized epoxy-clay nanocomposites and BMI modified siliconized epoxy-clay nanocomposites were synthesized and characterized by Selvaganapathi Anbazhagan et al. (2011)⁸⁹.

The effect of loading 4,5-epoxy cyclohexane-1,2-dicarboxylic acid diglycidyl epoxy resin (TDE 85) in BMI-bisphenol A–dicyanate (BADCy) were investigated in detail by Guanglei Wu et al. (2012). At 20 weight % of TDE 85-epoxy resin, the polymer network exhibited excellent mechanical properties, dielectric constant as well as thermal stability⁹⁰.

The influence of sizing agents in the interfacial properties of carbon fiber-BMI and carbon fiber (CF)-epoxy (EP) were investigated by Lirui Yao et al. (2012). From

FTIR spectra, it is revealed that these sizing agents (T700 grade high strength carbon fibers) are chemically reactive at 200°C and they can react with the polymer matrices. This type of reaction is observed for the CF-EP matrix while only partial reactions are observed in the CF-BMI matrix. Surface roughness, chemical properties and wetting properties of CF with BMI and EP matrix are modified by the use of a sizing agent that is confirmed from AFM and XPS studies. This study also proved that the sizing agent could significantly enhance the interfacial adhesion property between the CF and epoxy resin while no significant effects are observed in the CF-BMI matrix⁹¹.

BMI and epoxy modified novolac phenolic resin systems were prepared and their thermal stability and thermal degradation kinetics were investigated by Xiachuan Ye et al. (2012). The TGA and DSC results indicate that the fabricated matrix exhibits better thermal stability than the pure novolac resin⁹².

Tzu Hsuan Chiang et al. (2013) made attempts to chemically modify epoxy resin with melamine phenol formaldehyde novolac (MPN) and bismaleimide and investigated the effects of the BMI content on the reaction kinetics and processing conditions and also on thermal, flame retarding and dielectric properties of the epoxy-MPN system. From the results, it is concluded that the epoxy-MPN system had a lower dielectric constant compared to the epoxy-MPN-BMI system⁹³.

C₄-ether linked bismaleimide (C₄ e-BMI) epoxy composite consisting of nano-Al₂O₃ and silane treated Al₂O₃ were developed by M. Mandhakini et al. (2014). From the experimental data, it is inferred that the addition of 3-glycidoxypropyl methoxy silane (GPS) treated Al₂O₃ into the epoxy matrix enhances the thermo-mechanical properties of the nanocomposites significantly such that it can be used for structural applications⁹⁴.

The effect of carbon black (CB) on impact strength, toughness and electrical conductivity of epoxy matrix toughened with C8 ether linked (C8e-BMI) were investigated by M. Mandhakini et al. (2014). From the experimental data, it has been inferred that at 5 weight % of CB loading, there is a significant enhancement in impact strength, toughness and electrical conductivity of the nanocomposites⁹⁵.

Functionalization of recycling waste material like rice husk ash (RHA) by treating it with 3-glycidoxypropyl trimethoxysilane was done by K. Kanimozhi et al. (2014) followed by developing RHA-DGEBA-BMI composites using epoxy (DGEBA) as the matrix and BMI as the toughening agent. Experimentally obtained results indicate that different loadings of functionalized RHA have brought about significant changes

in thermo-mechanical, dielectric properties of the developed nanocomposites. These composites exhibit improved thermo-mechanical properties, excellent flame retardancy, and the ability to withstand higher temperatures, making them to use in the form of adhesives, coatings, and matrices to fabricate advanced composites for high performance applications⁹⁶.

Mandhakini Mohandas et al. (2014) employed nanoindentation tests to evaluate the mechanical properties of the epoxy-C4 e-BMI-GPS functionalized nano Al₂O₃ nanocomposites. The addition of 5 weight % GPS functionalized Al₂O₃ nanoparticles to 15 weight % C4e-BMI-epoxy matrix increases elastic modulus and nanohardness⁹⁷. The effect of incorporation of unfunctionalized and amino functionalized multiwalled carbon nanotubes (MWCNT) into BMI-diallyl bisphenol-A (DBA) epoxy resin and BMI-DBA-DGEBA (diglycidylether of bisphenol A) epoxy matrix on mechanical and thermal properties were investigated by R. K. Jena and C. Y. Yue (2015). The improvement in the thermomechanical properties demonstrates the significant benefits of incorporating NH₂-MWCNT into BMI-DBA-DGEBA epoxy systems⁹⁸.

A series of silicon containing hyperbranched epoxy (SHBEp)-BMI thermosets were prepared and their mechanical properties like flexural strength, impact strength and thermal stability were investigated by Song Niu et al. (2016). The obtained experimental data indicate that at 8 weight % of SHBEp, the polymer exhibits improved toughness and thermal property⁹⁹.

TaoWang et al. (2016) proposed the synergistic effect of phosphorus and maleimide groups on flame retardancy and thermal properties of epoxy resin by modifying epoxy resin with 10-(2,5-dihydroxylphenyl)-9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide (DOPO) and 1,1'-(methyldi-4,1-phenylene) bismaleimide (BDM). The experimentally obtained data indicated that the cured blends showed high T_g as well as remarkable flame retardant properties¹⁰⁰.

Epoxy (EP)-chain extended BMI (CBMI) polymers with high cycle-life shape memory property, thermal stability, mechanical stability, reconfiguration and selfhealing abilities were fabricated by Zhenjie Ding et al. (2018). These polymers have the ability to recover their original shapes and fracture toughness and also reconfigure their shapes upon thermal activation. As a result of these excellent integrated properties, they find application in smart structure materials¹⁰¹.

Nazrul Islam Khan et al. (2018) made attempts to design self healing polymers for use in sustainable engineering applications employing the matrix hybridization of thermoirreversible DGEBA epoxy resin with thermo-reversible Diels Alder (DA) based resin system (HPM) reinforced with bismaleimide grafted graphite nanoplatelets (B-GNP). The incorporation of B-GNP in HPM enhances the tensile strength and modulus along with successful interfacial healing¹⁰².

PES-EP-BMI composites were prepared by Yufei Chen et al. (2018) by uniformly dispersing polyethersulfone (PES) in the epoxy-bismaleimide matrix. Bending and impact strength were enhanced but there was only a slight increase in dielectric constant and dielectric loss observed by the addition of PES to the EP-BMI matrix compared with those of EP-BMI matrix¹⁰³.

Shintaro Kamiyama et al. (2018) studied the effect of matrix resin properties on the lightning strike damage behavior in carbon fiber reinforced (CFRP) laminates with three different kinds of resin matrices such as epoxy, BMI and polyether ether ketone (PEEK). Carbon fiber breakage, changes in color and delamination on the front surface was shown by CF-BMI and CF-PEEK composites. Their investigations revealed that electrical conductivity, char yield, the onset temperature of thermal decomposition and interlaminar fracture toughness affect the lightning strike damage¹⁰⁴.

EP- 2MI (2-methylimidazole), EP- 2MI -N, N'- bismaleimide- 4,4'-diphenylmethane (BDM), 9,10-dihydro-9-oxa-10- phosphaphenanthrene -10- oxide (DOPO) -EP-2MI, DOPO-EP-2MI-BDM systems were fabricated and their curing behavior flame retarding properties were studied by Qianqian Zhan et al. (2019). DOPO-EP-2MI-BDM thermoset exhibited better flame retardant properties with a high loss on ignition(LOI) value that may be ascribed to the synergistic flame retardant effect of phosphaphenanthrene in DOPO and BMI¹⁰⁵.

2.5. Polymer matrix composites with barium titanate (BaTiO₃) fillers

Due to high permittivity, BaTiO₃ (BT) nanoparticles are considered as the promising ceramic filler to disperse into the polymer matrix to form a composite with enhanced dielectric properties. Many attempts by researchers have been made to prepare polymer-BT composites and their dielectric studies for a variety of applications such as dielectric layers in flexible organic electronics, high performance nanogenerators, embedded capacitors, printing circuit boards (PCB), charge storage devices etc. A brief description of the research efforts is given below.

The influence of temperature treatment on dielectric permittivity of the polyesterstyrene-BT nanocomposites for optimized high k polymer composites was investigated by Benedikt Schumacher et al. (2009). The composites were exposed to a wide range of temperatures between 500°C to 1200 °C. As a result significant enhancement in dielectric permittivity and decrease in dielectric loss was obtained¹⁰⁶. Polyetherimide (PEI)-BT nanocomposite films were developed by Arup Choudhury (2010) employing solution casting followed by thermal imidization. The piezoelectric coefficients of PEI-BT nanocomposites increased upto 40 volume % BT loading thereafter it decreases that may be due to the formation of voids or pores. At 50 volume % of BT, the nanocomposites exhibited improved dielectric constant and dielectric loss along with good dielectric stability¹⁰⁷.

Yong Li et al. (2011) prepared two phase composites like PVDF-BT composites and PVDF- β SiC composites along with three phase composites PVDF-BT- β SiC and comparison has been made on their dielectric as well as thermal properties. Among these developed composites, PVDF-BT- β SiC exhibited higher dielectric constants and thermal conductivity compared to others¹⁰⁸.

M. E. Hossain et al. (2013) adopted different models such as Lichtnecker model, Maxwell Wagner model, Yamada and modified Kemer model to evaluate the frequency dependent dielectric properties of the fabricated BT-Parylene(polymer of para-xylylene) and BT-PVDF-HEP (Poly(vinylidenefluoride-co-hexafluoropropylene) composites. Comparison between the experimental data and theoretical calculations have been made. For dielectric constant, the first three models show reasonable agreement with the experimental data, while the dielectric loss simple rule of mixture with good predictions showed good agreement with experimental results. Based on the above studies on dielectric properties, the developed BT-PVDF-HEP composites can be used for designing high dielectric materials in future¹⁰⁹.

BT nanoparticles embedded polyester nanocomposites were developed by I. A. Asimakopoulos et al. (2014) by a compression moulding technique. With the increase in BT content, T_g of the composites decreases whereas the mechanical properties, dielectric properties and energy storage capability increases¹¹⁰.

Polyimide-BT nanocomposites were fabricated by electrospinning method by Wenhui Xu et al. (2014). The developed composites exhibited remarkable enhancement in dielectric permittivity, dielectric breakdown, mechanical properties with low dielectric loss and excellent thermal stability. The claimed properties make the composites attractive for use in the embedded capacitors on flexible printing circuit boards¹¹¹.

The effect of BT particle size on the dielectric permittivity of polystyrene (PS)-BT composites were investigated by O. Boorman et al. (2014). The particle dimension of BT filler was varied by ball milling at different time intervals. Ball milling of BT particles for 30 minutes was found to be effective for dispersion of BT particles in PS. An increase in real permittivity with BT content was observed for both non-milled and ball-milled samples¹¹².

Yifei Wang et al. (2015) paved the way to fabricate a sandwich structured BT-PVDF nanocomposites by solution casting layer by layer method for energy storage applications. With the increase in BT content, the dielectric constant increases gradually beyond 30 volume %, a leaping rise appears which may be due to the formation of a conductive path. At 20 volume % loading of BT, the composite showed maximum breakdown strength¹¹³.

Yujuan Niu et al. (2015) prepared BT-PVDF composite films with enhanced dielectric properties by adopting the ball-milled technique and compared with those prepared by the stirring method. The experimental results indicate that due to the well dispersion of BT particles in PVDF matrix without agglomeration, the interfacial interactions are strengthened significantly, thereby leading to the formation of polymer composites with high energy density, high permittivity and enhanced breakdown strength¹¹⁴.

In order to explore the potential applications of polycaprolactone (PCL) materials in tissue engineering, Jinyan Liu et al. (2018) prepared and characterized PCL-BT composites. Enhancement in piezoelectric effect was observed with an increase in volume % of BT. Cytomorphology observations obtained from SEM analysis and MTT test results indicate that the cytoactivity is not damaged by PCL-BT composites¹¹⁵.

Kunming Sing et al. (2018) constructed high performance piezoelectric nanogenerators (PENG) by using cellulose-BaTiO₃ (C-BT) aerogel papers based polydimethylsiloxane (PDMS) nanocomposites.

In order to enhance the performance of PENG, it is coupled with single electrode triboelectric nanogenerators (TENG). This coupling effect significantly enhanced the output voltage and power of the nanogenerator¹¹⁶.

In an effort to produce efficient electromagnetic interference shielding effectiveness (EMI SE) material Aqib Muzaffar et al. (2019) synthesized polyvinyl chloride (PVC)-BT-nickel oxide (NiO) nanocomposite films by solvent casting method. Homogeneous nanofiller dispersion within the PVC matrix obtained from SEM analysis and significant interaction between PVC, BT and NiO obtained from FTIR analysis confirms the successful formation of PVC-BT-NiO nanocomposites. The variation of BT and NiO nanofiller concentration in the PVC matrix has influenced the dielectric constant as well as EMI SE of the composite¹¹⁷.

In order to design an eco-friendly high performance piezoelectric composite, Hyeong Yeol Choi and Young Gyu Jeong (2019) prepared BT-cellulose composite films by aqueous solution casting technique. The effect of inclusion of BT nanoparticles on dielectric, piezoelectric, thermal, mechanical properties and microstructures of the fabricated composites were investigated in detail. The improvements are not only in dielectric properties like dielectric constant and dielectric loss but also in terms of piezoelectric outputs¹¹⁸.

The stored and harvested energy of the epoxy-BT nanocomposites under DC conditions and at various temperatures were investigated by G. C. Manika and G. C. Psarras (2019). With the increase in DC voltage, the stored energy and harvestability of the composites also increase. The coefficient of energy efficiency increases with BT filler content whereas it decreases with increases in temperature¹¹⁹.

Youngho Jin et al. fabricated BT-MWCNT-PVDF composites by effectively dispersing BT-MWCNT hybrid fillers using miscible–immiscible coagulation in PVDF matrix followed by hot pressing. At 37 volume % of BT and 3 volume % of MWCNT, the composites exhibited high permittivity with low dielectric loss. The high flexibility and excellent dielectric properties make the composite find applications in embedded and charge storage devices¹²⁰.

2.6. Polymer matrix composites with Rochelle Salt (RS) as filler

RS is well known for its piezoelectric effect and its anomalous dielectric behavior. In earlier research, parameters like temperature, humidity on dielectric behavior are investigated in detail. Current research efforts to develop RS filled composites, their synthesis and applications are also included in this literature review.

The effect of variation in frequency, electrostatic field and temperature on reversible inductivity of RS crystals was studied by John. C. Frayne (1908)¹²¹. The relation of relaxation time in RS crystals with temperature, field and thickness of the crystal are studied in detail by R. D. Schulwas et al. (1934)¹²². The influence of frequency and applied field strength on the variation of dielectric constant of RS crystals was investigated in detail by Anthony Zeleny and Joseph Valasek (1934)¹²³. The effect of

temperature and hydrostatic pressure on the electric susceptibility of RS crystals was studied by Dennison Bancroft (1938)¹²⁴.

Hans Mueller (1940) explained piezoelectric effect and anomalous thermal expansion of RS and phase transitions based on strains produced by the polarization or cooling of the RS crystals¹²⁵. The effect of temperature and humidity on the piezoelectric behavior and dielectric constant of RS was studied by Heiji Kawal (1948)¹²⁶. Humio Akao and Taizo Sasaki (1955) measured the dielectric dispersion of RS at 3000 - 20,000Mc/s and found that the relaxation frequency of RS exists in the decimeter wave region¹²⁷.

The effect of varying particle size and temperature on the dielectric properties of the compressed RS powders were measured under different humidity conditions by A. Mansingh and S. S. Bawa (1974)¹²⁸. Investigative studies based on the phase transitions of RS and deuterated RS crystals have been made by Bijay Krishna Chaudhari (1980) with two sublattice pseudo spin model¹²⁹.

I. H. Ismailzade et al. (1986) studied the temperature dependence of dielectric constant of RS crystals at E=0 and E=7 kV/cm and the Curie temperatures-low temperature T_{C1} and high temperature T_{C2} displacements of RS crystals upon the electric field strength¹³⁰. Critical dielectric parameters depend on the applied electric field in a parallel direction. L. Kalisz et al. (1993) made attempts to measure the relaxation effects and dielectric measurements in non-parallel electric fields¹³¹.

Comparative studies on the dielectric properties of pure RS nanocrystals grown in porous glass have been made by E. K. Jang et al. (1995)¹³². U. Schneider et al. (1999) studied ammonium doped RS crystals using linear and non-linear frequency dependent dielectric spectroscopy¹³³. Investigations based on different dielectric responses shown by the RS and deuterated RS crystals in high temperature ferroelectric phase transition are carried out by Inna A. Malyshkina and Nadezhda D. Gavrilova (2002)¹³⁴.

D. Yadlovker and S. Berger (2005) have grown RS crystals inside an array of alumina pores with different diameters and investigated the density of the array, polarization orientation and size for future application in ferroelectric–based devices¹³⁵.

Cheng Tien et al. (2008) carried out NMR studies to investigate the crystalline structure and ferroelectric effect exhibited by the RS embedded in molecular sieves which act as nanoporous matrices¹³⁶. C. H. Gorbitz et al. (2008) investigated the

structural features of RS crystals at 105K and provided the existence of low temperature paraelectric phase of RS crystals¹³⁷.

In this study, Balkrishna Kandpal and Trilok Chandra Upadhyay (2009)¹³⁸ calculated the dielectric constant and loss tangent of RS crystals under microwave absorption by green function method with the experimental results of Sandy and Jones¹³⁹. D. V. Rogazinskaya et al. (2009) prepared triglycine sulphate (TGS) and RS embedded nanoporous aluminum oxide and their dielectric studies are also investigated in detail.¹⁴⁰

S. V. Baryshnikov et al. (2010) prepared porous alumina films in which Rochelle salt (RS) was embedded in the pores of alumina and the dielectric properties of the films were investigated in detail in the temperature range from 80K to 325K. Sharp anomalies of permittivity will be observed at the upper and lower ferroelectric phases of RS at two Curie points such as 255K and 297K. Maximum permittivity of the developed alumina films was observed at 245K at a lower phase transition temperature of RS. There is no anomaly observed in the permittivity of the alumina films at the upper phase transition temperature¹⁴¹.

O. M. Golitsyna et al. (2010) prepared triglycine sulphate (TGS) and RS incorporated porous Al₂O₃ samples and the effect of absorbed water and temperature on the dielectric properties of these composite structures were investigated in detail¹⁴². The temperature dependence of dielectric susceptibilities of RS crystals was investigated by S. Miga et al. (2010) and confirmed the validity of Mitsui ferroelectric theory of RS with the results obtained¹⁴³. RS-PVA nanocomposites were prepared by Md Jamal Uddin et al. (2012) with RS loadings ranging from 0-16 weight % using the solvent cast method and investigated the dielectric, ferroelectric properties of the developed composites. With the increase in frequency, dielectric permittivity decreases whereas it increases with increase in temperature and RS content. Effective permittivity as well as effective conductivity increases with increase in RS content with very sharp rise in permittivity near the percolation threshold (f_c)¹⁴⁴.

The conventional solution method and Sankaranarayanan-Ramasamy (SR) method have been introduced for the growth of sodium potassium tartarate tetrahydrate (PST) by T. S. Shyju et al. (2012) and comparative studies based on the growth aspects and variation in properties has been made. The experimental data revealed that compared to conventional solution grown PST, SR method grown PST crystal exhibits excellent mechanical and dielectric properties¹⁴⁵. Toshio Kikuta et al. (2012) investigated the

influence of non-parallel DC electric field (transverse electric field) on remanent polarization in RS. It is observed that there is a decrease in remanent polarization with an increase in strength and time of the applied transverse electric field¹⁴⁶.

In order to study the effect of polymer matrix on the dielectric properties, A. B. Plaksitsky and A. G. Gorshkov (2014) prepared three different composites with different ferroelectric materials such as triglycine sulphate (TGS), Rochelle salt and sodium nitrite (NaNO₂) with polyvinyl alcohol (PVA) as the polymer matrix because of its high dipole moment which plays an important role in the dielectric measurements and ease of manufacture. Comparative studies of PVA-TGS, PVA-NaNO₂ and PVA-RS composites, anomalies in the capacitance were observed in the same temperature region for both PVA-RS composites as well as the bulk RS crystals. There is a reduction in the capacitance of the composite with an increase or decrease in RS content that may be due to the weak coupling of RS with PVA matrix¹⁴⁷.

Hard and stiff piezosalt textile was prepared by E. Lemaire et al. (2015) loading RS filler into polypropylene based absorbent textile. Piezotextile with actuated microcantilever makes the RS stay piezoelectrically active¹⁴⁸.

N. G. Popravko et al. (2015) prepared the nanocomposites by introducing the fillers like triglycine sulphate (TGS) and RS separately into the porous alumina films and compared the IR spectra of both composites to identify the existence of strong interactions like hydrogen bonds¹⁴⁹. Frode Mo et al. (2015) investigated the high temperature paraelectric phase of RS crystals at 308K by using synchrotron X-ray diffraction¹⁵⁰. Comparative studies based on the photoluminescence, photoconductive nature and piezoelectric behavior of pure triglycine sulphate (TGS) and RS doped TGS have been studied by P. Rajesh et al. (2016)¹⁵¹. The dielectric constant and dielectric loss of RS crystals was calculated by using the green function technique¹⁵² and compared with experimental values obtained from other works by Balkrishna Kandpal and Trilok Chandra Upadhyay (2016)¹⁵³. In the paper, V. Mathivanan et al. (2015) reported a detailed comparative study on thermal, magnetic, dielectric behavior and antimicrobial activity of pure RS and copper doped RS crystals¹⁵⁴.

Aanchal Rawat and Trilok Chandra Upadhyay (2017) investigated the temperature dependence of dielectric constant and loss tangent with the help of modified soft mode frequency and this mode clearly explains the ferroelectric transition in RS crystals¹⁵⁵. Hoai Thuong Nguyen and Dung Mai (2019) prepared ferroelectric composite from varying the concentration of the cellulose nanoparticles (CNP) with a

fixed amount of RS. With the increase in cellulose content, expanded region 3000-3500 cm⁻¹ was obtained in FTIR spectra that may be due to the number and strength of hydrogen bonds between CNP and RS. The ferroelectric phase of RS is maintained upto its decomposition temperature, even at high CNP content¹⁵⁶.

Aachal Rawat and Trilok Chandra Upadhyay (2019) explained the temperature dependence of soft mode frequency, dielectric constant and loss tangent of deuterated RS crystals (DRS) by calculating the values and comparisons are made with experimental results of other works¹⁵⁷.

2.7. Polymer matrix composites with surface modified barium titanate as fillers

In order to enhance the dielectric properties and compatibility of the interface, surface modification of nanofillers serve as the most effective technique. Research based on the composites embedded with surface modified fillers revealed remarkable enhancement in the thermo-mechanical as well as dielectric properties of composites which may be attributed to the uniform dispersion of filler in the matrix and strong interfacial interaction between the filler and the matrix. Different surface modification of the filler and their composites are investigated by the researchers and are discussed here as the literature review.

Commercial raw materials for multilayer ceramic capacitors (MLCC) are BT and ammonium polyacrylate dispersant (APA). BT being reactive in an aqueous environment as well as moisture and CO_2 in the air will affect the performance and reproducibility of MLCCs. It was found that there was more chemically adsorbed APA dispersant on BT surface when the surface of the latter was more hydroxylated and less carbonated. Burtrand I. Lee (1998) examined lot to lot variations in pH and ammonium content for the APA dispersant¹⁵⁸.

Akio Takahashi et al. (2005) reported the fabrication of polyamide (PA)-BMI-BT composites for embedded capacitors. In order to increase the dielectric constant of polymer ceramic composites, surface modification of BT particles was done by methallophthalocyanine oligomers. PA-BMI composite film with the above surface modified BT exhibited a dielectric constant of 80 and a dielectric loss of 0.02 at 1 MHz¹⁵⁹. Philseok Kim et al. (2009) investigated the role of volume fraction of fluorinated phosphonic acid modified BT on the dielectric properties such as dielectric permittivity, dielectric breakdown strength and dielectric loss on the polyvinylidene fluoride-co-hexafluoropropylene (PVDF-HFP) nanocomposites¹⁶⁰.

2,3,4,5-tetrafluorobenzoic acid modified BT nanoparticles were synthesized by Yujuan Niu et al. These fluorocarboxylic acid modified BT (F4CBT) nanoparticles were incorporated into PVDF polymer matrix. The above fabricated nanocomposites showed high breakdown strength, low dielectric loss and good compatibility. F4CBT modified BT nanoparticles act as an excellent modifier for the nanocomposites which find application in the field of pulsed power capacitors¹⁶¹.

Chang et al. (2009), in their paper, proposed a novel approach to hydroxylate BT nanoparticles and further chemical modifications of surface hydroxylated BT nanoparticles were done using sodium oleate (SOA). Samples of BT nanoparticles were treated separately with H_2O_2 and with HCl solutions in order to compare the influence of the acidic properties on the surface treatment of BT. From the FTIR spectra, they concluded that a much weaker peak of O-H vibration was obtained for the HCl treated BT, indicating that highly surface hydroxylated BT nanoparticles were produced by the oxidation of BT nanoparticles using an aqueous of $H_2O_2^{162}$.

Ohia Chen Li et al. (2010) investigated the chemical behaviour of the –OH groups produced on the surface of BT nanoparticles as a result of H_2O_2 treatment and how these –OH groups affect the adsorption selectivity of BT nanoparticles as surface modification agents, i.e., the surface reactivity of BT nanoparticles in addition to the quantitative analysis of the hydroxylation efficiency including acid base properties and chemical reactivity. According to XPS data, there is a significant increase in the concentration of –OH groups on the surface of BT nanoparticles as a result of H_2O_2 treatment. They have reached a conclusion based on the XPS data and zeta potential values that –OH is attached to Ba instead of Ti making the –OH Bronsted-basic and this increases the adsorption selectivity of BT nanoparticles for further surface modification¹⁶³.

Tao Zhou et al. (2011) prepared crude BT-PVDF and hydroxylated BT-PVDF nanocomposites and comparison studies were conducted. The hydrogen bond between hydroxylated BT and PVDF matrix leads to stronger interaction between the nanofiller and the polymer matrix, which might be the reason for the higher breakdown strength of the fabricated nanocomposites¹⁶⁴. Yu Song et al. (2012) reported (polyvinylidene fluoride trifluoroethylene) PVDF-TrFE-dopamine modified BT nanocomposites with enhanced dielectric permittivity and breakdown strength. The enhancement in dielectric properties is attributed to polydopamine layers on BT nanofibers and the improved crystallinity induced by these BT nanoparticles¹⁶⁵.

Mohmoud .N. Almadhoun et al. (2012) reported the influence of surface hydroxylated BT nanoparticles on the performance of polyvinylidene fluoride trifluoroethylene (PVDF-TrFE) nanocomposites. Loading of hydroxylated BT in this inorganic-organic hybrid nanocomposites shows improved breakdown strength and dielectric permittivity. The surface modification of BT nanoparticles results in a higher degree of polymer matrix interaction¹⁶⁶.

Several modifications are done on BT by Jelena Culic Viskota et al. (2012) in order to introduce it in Second harmonic generating (SHG) nanoprobes for invivo imaging in zebra fish. At first surface hydroxylation of BT was done by using H₂O₂ treatment, then the resulting –OH groups are allowed to react with N-aminoethyl-2,2,4-trimethyl-1-aze-2-silacyclopentane exposing terminal NH₂ domain which acts as a platform for further modification and NH₂ surface reacted with 6-azido-hexanoic acid which further exposes azide moiety and further modification by biotin-PEO-cyclooctyne resulting with biotin BT terminal end that can be reacted with Streptavidin, further reaction with Methoxy poly(ethylene glycol)succinimidyl valerate (mPEG-SVA) to cover the surface of BT with PEG (Poly(ethylene glycol)). This coating assists in keeping the particles well dispersed for the effective introduction of BT into biological samples¹⁶⁷.

Yu Song et al. (2012) points out the effect of filler shape and surface modification on the dielectric properties of PVDF and epoxy based composites for which they have synthesized dopamine modified nanofibers and dopamine modified nanoparticles. Their results suggest that dopamine modified nanofibers could be used to improve dielectric properties and electric energy density in polymer nanocomposites¹⁶⁸.

Arup Choudhury (2012) prepared Polyetherimide (PEI) surface hydroxylated BT nanoparticles and these nanocomposites showed enhanced dielectric permittivity compared with PEI-BT nanocomposite. These nanocomposites showed good dielectric stability over a wide range of frequencies and temperatures and these results suggest that PEI-surface functionalized BT nanocomposites are promising candidates for the development of embedded capacitor devices¹⁶⁹.

Penghao Hu et al. (2012) made attempts to enhance the energy density in BSBT-PVDF-TrFE composites by incorporating dopamine modified Bi₂O₃-doped strontium barium titanate (BSBT). The enhancement in the dielectric constant, breakdown strength and energy density may be attributed to the combined effect of the surface modification, large aspect ratio and paraelectric polarization behaviour of BSBT nanofibers¹⁷⁰.

Xianhong Zhang et al. (2014) presented a novel strategy to fabricate PVDF-BT nanocomposites in which BT nanofibers are surface modified with 1H, 1H, 2H, 2H perfluoro octyltrimethoxy silane. Nanocomposites of PVDF with fluoro silane modified BT nanofiber exhibited high dielectric constant and reduced tan δ values¹⁷¹. PVDF-BST nanocomposites with barium strontium titanate nanofibers (BST60NF) and BST60NF-OH nanofibers were fabricated separately and comparative dielectric studies were carried out by Liu Shaohui et al. (2014). Their results suggest that the nanocomposite loaded with BST60NF-OH nanofibers exhibited enhanced relative permittivity, low dielectric loss and improved breakdown strength that find applications in high energy storage devices¹⁷².

Rongyan Zhao et al. (2015) focussed on solving the sedimentation problem associated with solvent casting processing during the preparation of polymer nanocomposite films. Most of the nanofillers have a strong tendency to agglomerate due to the small size and high surface to volume ratio. Surface hydroxylation by H_2O_2 or surface amination by γ -aminopropyl triethoxy silane (γ -APS) was carried out to modify the surface of BT fillers. Then agglomeration and sedimentation of these surface modified fillers during solution casting processing with N, N-dimethyl formamide (DMF) as solvent was checked and compared with the raw BT filler. They also made comparative studies on the dielectric properties such as dielectric constant, dielectric loss and break down strength for the fabricated nanocomposite films of PVDF-surface hydroxylated BT (BT-OH) films and PVDF-surface aminated BT(BT-NH₂). It was found that both nanocomposite films exhibited better dielectric properties than the one with raw BT filler. Among the PVDF-BT-OH and PVDF-BT-NH₂ nanocomposite films, the one with hydroxylated BT filler (BT-OH) shows a better effect than aminated BT-NH₂ ones¹⁷³.

Nao Kamezawa et al. (2015) prepared PVDF nanocomposite film by the homogeneous dispersion of BT nanoparticles with 3,3,3-tri fluoro propyl methoxy silane (FPS) modifier. At first, they prepared a double alkoxide of barium and titanium as the precursor for the BT nanoparticles by using metallic barium and tetraethyl orthotitanate (TEOT) and 2-methoxy ethanol as solvent. FPS was added to the solution containing freshly synthesized BT nanoparticles after ultrasonication. The

fabricated BT-FPS-PVDF transparent nanocomposite films showed a high dielectric constant of 44 and a relatively low dissipation factor¹⁷⁴.

Nikita Emelianov's (2015) research work includes a detailed study of the dielectric properties of PS-BT nanocomposites in which BT nanoparticles were the first surface hydroxylated and then chemically modified by Sodium oleate (SOA). The chemical modification of BT nanoparticles by SOA reduces their agglomeration in the polymer matrix and the nanoparticles are more uniformly dispersed in PS matrix¹⁷⁵.

Shaohui Liu et al. (2016) paid attention to develop ceramic polymer composites with enhanced dielectric properties and high energy storage density due to their potential application in energy storage and power capacitors. They synthesized cube-shaped barium strontium titanate (BST) nanoparticles using a molten salt method and adopted a hydroxylation strategy to obtain surface hydroxylated BST nanoparticles (BST-NP-OH)¹⁷⁶.

Yanyan Fan et al. (2016) reported the silanization on the hydroxylated BT nanoparticles and characterized these alkoxy silane modified BT nanoparticles. The introduction of alkoxysilanes on the surface of BT nanoparticles significantly enhances the compatibility between the alkoxy silane modified BT nanofiller and PVDF matrix. These fabricated PVDF-BT nanocomposites are more adaptable for high dielectric applications¹⁷⁷.

Mei Du et al. (2016) reported the enhancement in dielectric properties of PVDF composites reinforced by blended fillers of surface hydroxylated BT and exfoliated silanized graphene. The functionality of both BT and EG improves the interfacial bonding between fillers and the matrix, thereby increasing the dispersion of nanoparticles in polyvinylidene fluoride (PVDF) which is responsible for high dielectric constant values of the composites¹⁷⁸.

The electrospinning process was employed by Shaohui Liu et al. (2016) to synthesize barium zirconate titanate nanofibers (BZTNF). Surface modification of BZTNF was done by polyvinyl pyrrolidone (PVP) due to its strong interfacial adhesion strength to the surfaces including polymers and metal oxides. Fabrication of nanocomposite films of BZTNF-PVDF and surface modified BZTNF by PVP-PVDF were done by solution casting method and comparative dielectric and energy density studies were carried out. The results indicate that even a small loading of surface modified BZT by PVP in PVDF results in nanocomposites with enhanced dielectric constant, reduced loss

tangent and high energy storage density made to use these fabricated nanocomposites in energy storage applications¹⁷⁹.

Srikanta Moharana et al. (2017) prepared PVDF-BT composites consisting of BT particles modified with a non-ionic and hydrophilic surface agent like Polyethylene glycol (PEG) which further modify the interfaces between PVDF and BT. These composites exhibit higher dielectric constant \approx 192 and lower dielectric loss. The enhancement of dielectric constant attributed to the mobility of PEG and increase of interfacial polarization effects. PEG-BT-PVDF composites with enhanced dielectric and ferroelectric properties find application in the field of high energy storage capacitors¹⁸⁰.

Y. N. Hao et al. (2017) synthesized hydroxylated colloidal BT nanoparticles with perovskite structures via gel collection method and used these particles as fillers directly to fabricate polyvinylidene fluoride–co-hexafluoro propylene (PVDF-hfp)-BT nanocomposite films. Their investigation reveals the influence of colloidal perovskite nanoparticles on the fabrication and dielectric properties of the nanocomposite films¹⁸¹. Neydys Gonzalez et al. (2017) carried out an extensive study on the thermal, mechanical and dielectric properties of natural rubber (NR) films based on the effect of the nanofiller content–both functionalised and non-functionalised BT nanoparticles. The addition of different weight percentages of surface hydroxylated BT nanoparticles enhanced the dielectric properties such as breakdown strength¹⁸².

Youngtae Kim et al. (2017) reported the design of highly fluorinated–ligand modified and ligand exchanged BT nanoparticle composites with high dielectric constant. Here hydroxylated BT surface was further successfully modified by ligand substitution reactions. Their findings suggest that these types of polymeric fluorous ligand modified BT nanoparticles can be adopted as useful components in printed electronic devices¹⁸³.

Francesco Piana et al. (2018) reported the preparation of high dielectric constant polystyrene (PS) composite films with surface functionalized barium titanate (BT) nanoparticles. Surface modification of BT nanoparticles was carried out by hydroxylation with H_2O_2 and further functionalization by treatment with palmitic acid. PS-BT(PAc) films were prepared by the typical floating method. These composites are suitable for applications in flexible organic electronics as dielectric layers¹⁸⁴.

Barium titanate foam (BTF) with three dimensions (3D) open foam structure was prepared from BT ceramic particles by adjusting the number of coatings (n) for a fixed sintering temperature of 1200 ⁰C. The resultant BTF was coded as BTFn and the surface hydroxylation of BTFn was carried out using H₂O₂ and the resultant product was coded as BTFn-OH. A similar procedure was adopted for obtaining hyperbranched polysiloxane modified BTF denoted as BTF@HSi from BTFn-OH. Longhui Zheng et al. in 2018 investigated the dielectric properties of the fabricated BTFn-CE (Cyanate ester), BTF@HSi-CE, BTFn-cPES [poly (ether sulfone)]-CE and BTF@HSi-cPES-CE composites. The introduction of cPES into CE matrix effectively blocks the cracks caused by the stress concentration of the composites. BTF@HSi-cPES-CE composites showed a high dielectric constant, low dielectric loss, and better frequency stability, making them interesting candidates for fabricating integral capacitors in the system on package substrate¹⁸⁵.

Penghao Hu et al. (2018) prepared surface hydroxylated BT nanoparticles by H_2O_2 treatment and further carried out the modification with titanate coupling agent (TC-2) in the form of surface coating via solution method. The fabricated PVDF-modified BT nanocomposite films exhibited enhanced dielectric breakdown strength and improvements in the energy storage performance and these high energy density nanocomposites act as a potential candidate for advanced dielectric capacitors¹⁸⁶.

2.8 Polymer nanocomposites with MWCNT fillers

Lathapriya Vellingiri et al. (2018) synthesized multiwalled carbon nanotube (MWCNT)-SnO₂ nanocomposites with different weight % of SnO₂ (3, 5, 7 and 9) by impregnation method and investigated the hydrogen storage capacity of these composites. MWCNT-SnO₂ nanocomposites with 3 weight % of SnO₂ exhibited the highest hydrogen intake capacity¹⁸⁷.

Bismaleimide (BMI) modified with diallyl bisphenol A (BA)-Carbon nanotube (CNT) hybrid and BMI-BA-aminated CNT (A-CNT) hybrids were prepared by Aijuan Gu et al. (2007) and the synthesized hybrids exhibited higher impact strength, low glass transition temperature (Tg) as well as low storage modulus¹⁸⁸.

High performance bismaleimide polymer nanocomposite conductor reinforced with high concentrations of MWCNT was fabricated and investigated their high mechanical and electrical properties by Qunfeng Cheng et al. (2009)¹⁸⁹.

T. L. A. Montanheiro et al. (2015) adopted a solution casting method to synthesize (poly(hydroxybutyrate-co-valerate)PHBV-MWCNT nanocomposites loaded with 0.5 weight % of pristine MWCNT, carboxylated MWCNT (MWCNT-COOH), hydroxylated MWCNT (MWCNT-OH) and studied the thermal and electrical properties of these samples in detail¹⁹⁰.

The effect of graphene nanoplatelets (GNP) and MWCNT on dielectric and optical properties as well as electrical conductivity were investigated by Omer Bahadir Mergen et al. (2020) in polystyrene-graphene nanoplatelet (PS-GNP) and PS-MWCNT nanocomposites, the latter was developed by solution mixing technique. These composites with enhanced dielectric permittivity and ac conductivity find applications in EMI shielding and enhanced energy storage devices¹⁹¹.

Nuzul Fatihin Izatil Azman et al. (2021) carried out an experimental study to investigate the morphology and thermal properties of PET (polyethylene terephthalate) nanocomposites loaded with MWCNT and GNP at different concentrations (0.5 weight % and 0.1 weight %). The incorporation of a small amount of the fillers-MWCNT and GNP improved the crystallization of PET nanocomposites along with a slight increase in thermal properties of the fabricated PET nanocomposites as compared to pure PET¹⁹².

The effect of addition of MWCNT filler on electrical properties of MWCNT-epoxy nanocomposites was investigated by Manindra Trihotri et al. (2016). Dielectric properties like dielectric permittivity, dielectric loss and ac conductivity of the fabricated MWCNT-epoxy nanocomposites exhibited maximum enhancement at 0.5 weight % loading of MWCNT filler¹⁹³.

Vinylester resin (VER)-epoxy (EP) nanocomposites were prepared by Ankita Pritam Praharaj et al. (2015) with loadings in the range from 0, 1, 3, 5 and 7 weight % of amine functionalized MWCNT (f-MWCNT). Interestingly it has been observed that the nanocomposite with 5 weight % of MWCNT filler exhibited remarkable enhancement in thermo-mechanical as well as electrical properties¹⁹⁴.

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