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Mechanically and magneto-dielectric enhanced interpenetrating polymer network of polyurethane/polystyrene for flexible electronic applications

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Abstract

The nonmagnetic/electrical network of two polymers, known as the Polyurethane/Polystyrene Interpenetrating Polymer Network, is utilised in industry, particularly for mechanical applications. As previously stated, polyurethane is most likely utilised in everyday life for things like covering sheets for luggage in the open air and tiny pipes used in the gas business. Although elongation at break and hardness do matter, organic clays and powders have been used to increase mechanical strength. However, these interpenetrating polymer networks are only used for mechanical purposes. Nowadays, ceramic oxide nanoparticles are used to increase mechanical strength in place of organic clays and powders. By being included into a network of polymers, these ceramic oxide nanoparticles not only increase the mechanical strength of the polymer but also develop unique properties such as conductive, dielectric, and magnetic behaviour. By employing in-situ polymerization to incorporate ceramic ferrite nanoparticles, which are employed in the electronic sector, particularly for memory storage, we present the magnetic, dielectric, and conducting responses of a nonmagnetic/electrical network of two polymers.

Keywords: Flexible mats, interpenetrating polymer network (IPN), elongation at break, thermal stability, castor oil, flexible capacitor

Introduction

The integration of ceramics into polymer matrices has emerged as a promising strategy for enhancing the properties of traditional polymer materials. Ceramic-filled polymer nanocomposites combine the advantageous characteristics of both components, leading to materials with significantly improved mechanical and dielectric properties. This innovative class of materials is receiving increasing attention for various industrial applications, ranging from electronics to structural components in aerospace and automotive industries.

Polymers are widely recognized for their lightweight, corrosion resistance, and ease of processing. However, their relatively low mechanical strength and thermal stability limit their application in demanding environments. On the other hand, ceramics are known for their high strength, rigidity, and excellent electrical insulation properties but are often brittle and challenging to process in thin geometries. Ceramic-filled polymer nanocomposites leverage the advantages of both materials, leading to enhanced performance profiles. The addition of ceramic fillers, especially at the nanoscale, can significantly improve the mechanical performance of polymers while retaining or enhancing dielectric properties, making them suitable for a wide range of industrial purposes.

Several ceramic materials can be used as fillers in polymer matrices, including:

- **Silicates:** Such as montmorillonite and kaolin, which improve strength and thermal stability.
- **Oxides:** Like alumina and titania, which enhance mechanical strength and provide electrical insulation.
- **Nitrides and Carbides:** Such as silicon carbide and boron nitride, known for their high thermal conductivity and strength.
- **Epoxy-Silica Composites:** Silica nanoparticles are increasingly used in epoxy resins to refine mechanical properties and add functionality.

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The mechanical properties of ceramic (Magnetic Oxides)-filled polymer nanocomposites are prominently influenced by the type, size, and distribution of the ceramic fillers results in enhanced Strength and Stiffness: The incorporation of ceramic fillers can lead to significant improvements in tensile strength and modulus of elasticity, primarily due to the load-bearing capacity of the ceramic phase as well as impact Resistance contribute to stiffness, careful consideration of filler morphology can enhance energy absorption characteristics, improving fracture toughness. Apart from this, thermal Stability: Ceramic fillers can enhance the thermal stability of polymers, allowing them to maintain performance under elevated temperatures. The dielectric properties of ceramic-filled polymer nanocomposites are crucial for applications in electrical insulation and capacitive devices. Ceramic fillers often exhibit low dielectric loss, making these nanocomposites suitable for high-frequency applications, such as telecommunications and power electronics.

Ceramic-filled polymer nanocomposites find applications in various fields:

- **Electronics:** Used in capacitors, insulators, and as substrates for electronic devices, where both mechanical robustness and electrical properties are essential.
- **Aerospace and Automotive:** Employed in structural components that require lightweight materials with high strength and thermal resistance.
- **Biomedical Devices:** Their biocompatibility and mechanical properties make them suitable for applications in medical devices and implants.

Borda *et al.* ^[1] Utilised poly (MDI) combinations to create biodegradable urethane composites with wheat stalks, to which additional hydroxyl-containing substances like water, flour, etc. were added. The composite produced by the PMDI-water mixture without catalyst has the best mechanical qualities, according to the results. Additionally, the effect of the straw stalk's length on the pressed sheet's physical characteristics was examined, and the results demonstrated that lengthening the straw might result in a composite with good mechanical qualities. The potential chemical reaction's mechanism.

Chen-Yang *et al.* ^[2] made a number of PU/MMT nanocomposites using PU and MMT. Even with up to 7 weight percent of MMT addition, the TEM images showed a highly exfoliated structure, confirming the PU/MMT nanocomposites. This led to significant reinforcements in their mechanical, thermal stability, and anti-corrosion protection. Furthermore, compared to previously published PU/clay nanocomposites with similar clay loadings, the reinforcements were significantly higher. This was attributed to the loaded organoclay's high exfoliation and appropriate dispersion.

Pattanayak and Jana ^[3] discussed how soft-segment affects the exfoliation of clay particles and the resulting mechanical and thermal characteristics of thermoplastic polyurethanes (TPU) and reactive layered silicate clay nanocomposites. The composites were created using a two-step bulk polymerization process using butanediol, diphenyl methane diisocyanate, polyether- and polyester polyol with molecular weight 2000, and up to 5 weight percent reactive layered silicate clay. Although the degree of clay-polymer tethering was the same for both polyester and polyether-polyol, this nanocomposite was only created in the case of polyester polyol because of its significantly higher viscosity during clay-polymer mixing. Additionally, the study demonstrated that clay particles had

no effect on hard segment hydrogen bonding and that hydrogen bonding has no effect on mechanical qualities. The presence of clay particles caused strong improvement in stability against thermal degradation, although clay-polymer tethering showed almost no effect. Clay tethering and resultant clay particles exfoliation, however, caused strong improvement in mechanical properties.

Athawale and Kalekar ^[4] created reinforced elastomeric interpenetrating polymer networks (IPNs) made of polymethylmethacrylate (PMMA) and glycerol modified castor oil polyurethane (CG-PU). The mechanical, chemical, and thermal properties of the IPNs were examined in relation to the impacts of polyol modification on the NCO/OH ratio and the composition of polyurethane/PMMA. The detailed analysis of CG-IPNs showed that as the NCO/OH ratio rose, elongation dropped and tensile strength and hardness increased. The alteration in the NCO/OH ratio and PU/PMMA composition had no effect on the IPNs' chemical or thermal resistance.

Hourston *et al.* ^[5] created and investigated interpenetrating polymer networks of polyurethane (PU) and polystyrene (PS) with ionic groups for mechanical and viscoelastic characteristics. According to Dynamical Mechanical Thermal Analysis (DMTA), the addition of ionic groups significantly alters the viscoelastic characteristics. They demonstrate that while Young's modulus and hardness values improved dramatically with 2% by weight of ionic groups, stress at break values increased significantly.

Vlad *et al.* ^[6] Created a number of interpenetrating polymer networks (IPNs) using polydimethylsiloxane and polyurethane (PU) as the bases. DSC, TEM, TGA, 1H-NMR, and IR spectroscopies, among other methods, have been used to synthesise and characterise these IPNs. Chloroform and n-hexane swelling have been used to describe the homonetworks. A PU based on castor oil and 2, 4-toluene diisocyanate (TDI) combined with varying concentrations of polydimethylsiloxane-a, x-diol (PDMS) yields the IPNs.

Siddaramaiah *et al.* ^[7] reported sequential polymerisation was used to create the two component polymer networks (IPNs) of polyurethane (PU) and polystyrene (PS), which are based on castor oil. Castor oil and MDI-based polyurethane was swelled in polystyrene monomer before being polymerised by a radical polymerisation process that was started with benzoyl peroxide and cross-linked with divinyl benzene (DVB). By casting in glass moulds, a number of PU/PS IPNs were produced as durable films. These films' mechanical qualities, thermal behaviour (TGA), and resistance to chemical reagents were identified as their features. SEM was used to determine the morphology, which shows that adding plastic polystyrene to an elastomeric polyurethane increases its tensile.

Silva R.V. *et al.* ^[8] investigated the fracture toughness of polyurethane composites made of natural fibres and castor oil. The matrix and fibres are both made from sustainable resources. Both with and without sodium hydroxide treatment, sisal and coconut short fibres as well as woven sisal fabric were utilised. The sisal fabric composite showed the best performance in terms of fracture toughness. It turned out that the alkaline treatment was detrimental to fracture toughness.

Rehab and salahuddin ^[9] produced by synthesising using the in situ polymerisation technique to create polyurethane organoclay nanocomposites. Using an ion exchange technique, diethanolamine or triethanolamine is intercalated into montmorillonite clay (MMT) to create the organoclay. The process of creating polyurethane-organoclay hybrids

involved first swelling the organoclay into various diols, then adding diisocyanate. The dispersed structure of MMT was demonstrated by the nanocomposite using X-ray diffraction (XRD) and scanning electron microscopy. The findings indicate that the MMT is uniformly distributed throughout the polymer matrix and that the peak characteristic has broadened with low intensity and shifted to d_{001} spacing to smaller 2θ . When compared to virgin polyurethane, the TGA revealed that the nanocomposites had a greater breakdown temperature.

Vatalis *et al.* [10] reported using a variety of thermal analysis techniques, including differential scanning calorimetry (DSC), thermomechanical analysis (TMA), thermally stimulated depolarisation currents (TSDC), thermally stimulated conductivity (TSC) measurements, and broadband dielectric relaxation spectroscopy (DRS), thermoplastic interpenetrating polymer networks (t-IPNs) were created by melting and pressing crystallisable polyurethane (CPU) and styrene/acrylic acid random copolymer (S/AA) in varying composition.

Experimental Methodology

1. Auto Combustion Method: In this method, a required metal nitrate of above mentioned ferrites has been first weighed according to their stoichiometric proportion. The weighed metal nitrates first dissolved individually in double distilled water. The citric acid has been used as fuel agent also be dissolved in double distilled water according to required stoichiometric proportion. The dissolved individual nitrates now mixed with each other along with continuous stirring and heating. The heating

process continued till liquid converted into powder. The powder is calcined at 1000 °C for phase formation and sintered at 1200 °C.

2. In-situ Polymerization of PU/PS IPNs-Spinel Ferrite
Dehydrated castor oil and MDI were taken in NCO/OH equivalent ratio of 1:1. Reaction was allowed to proceed at the room temperature in the beaker itself with continuous stirring. After almost 60% conversion or 1 hour from the beginning of the reaction, known amount of styrene was added to the mixture along with the ferrite nanoparticles. The reaction mixture was thoroughly stirred so that the particles remain in suspension. Styrene should be added about 50% weight of the total resin. Styrene was added as it forms interpenetrating polymer network with polyurethane which provide improves properties like tensile properties etc. to polyurethane. After all this reagents, 1% Cobalt Octet (of the total weight of the resin) and 1% Methyl Ethyl Ketone Peroxide (MEKP) were added to initiate the reaction and act as a cross linking agent at a difference of 5 mins. 1% Dibutyl Tin Laurate (DBTL) by weight of the mixture was also added which catalyses the reaction. Stirring is continued so that the modified clay was fully dispersed. When the stirring mixture become quite viscous, it should be kept in a mould overnight. Next day the sheet was removed from the mould. For further curing, the sheets were kept in the oven for 2-3 hours at temperature around 80 °C. Absence of smell of styrene and tackiness on the surface means that the sheets have been fully cured. Different sheets with varying amount of modified clay ranging from 5% to 25% by optimization were prepared.



Fig 1: Step by Step Representation of Making of Flexible Mats of PU/PS IPN-CoFe₂O₄

Conclusion

In-situ polymerization of PU/PS with CoFe₂O₄ as a filler, in the presence of a catalyst and chain initiator, has successfully produced flexible sheets of PU/PS IPN-CoFe₂O₄. The presence of CoFe₂O₄ in the IPN matrix is confirmed by the FTIR spectra and X-ray diffraction data. The labyrinth (barrier) effect created by the highly anisotropic oxide, which slows the escape of volatile degradation products, is

demonstrated by the thermal gravimetric data, which also demonstrates improvement in weight loss against temperature inclusion of filler (CoFe₂O₄ integrated) in the matrix of PU/PS IPN. Energy dispersive and elemental mapping spectroscopy have verified the elements' uniform distribution and confirmation. The existence of CoFe₂O₄ as a filler in the PU/PS matrix is further confirmed by the magnetic hysteresis in non-magnetic PU/PS, and magnetization rises as CoFe₂O₄

content rises. The improvement. The increment in elongation at break directly reveals enhancement in mechanical properties with simultaneous magnetic behavior of PU/PS incorporated with magnetic ceramic (CoFe_2O_4) and may be due to interaction of Fe^{3+} ions spinning in both tetrahedral and octahedral sites with Ni^{2+} ions in octahedral sites. The

magnetic nature of polymer used as storage device in computers whereas capacitive behavior of polymers made them useful in electronic industries for charge storage. Apart from magnetic and dielectric applications, flexible nature of ferrite nanoparticles important in modern day flexible electronics devices.

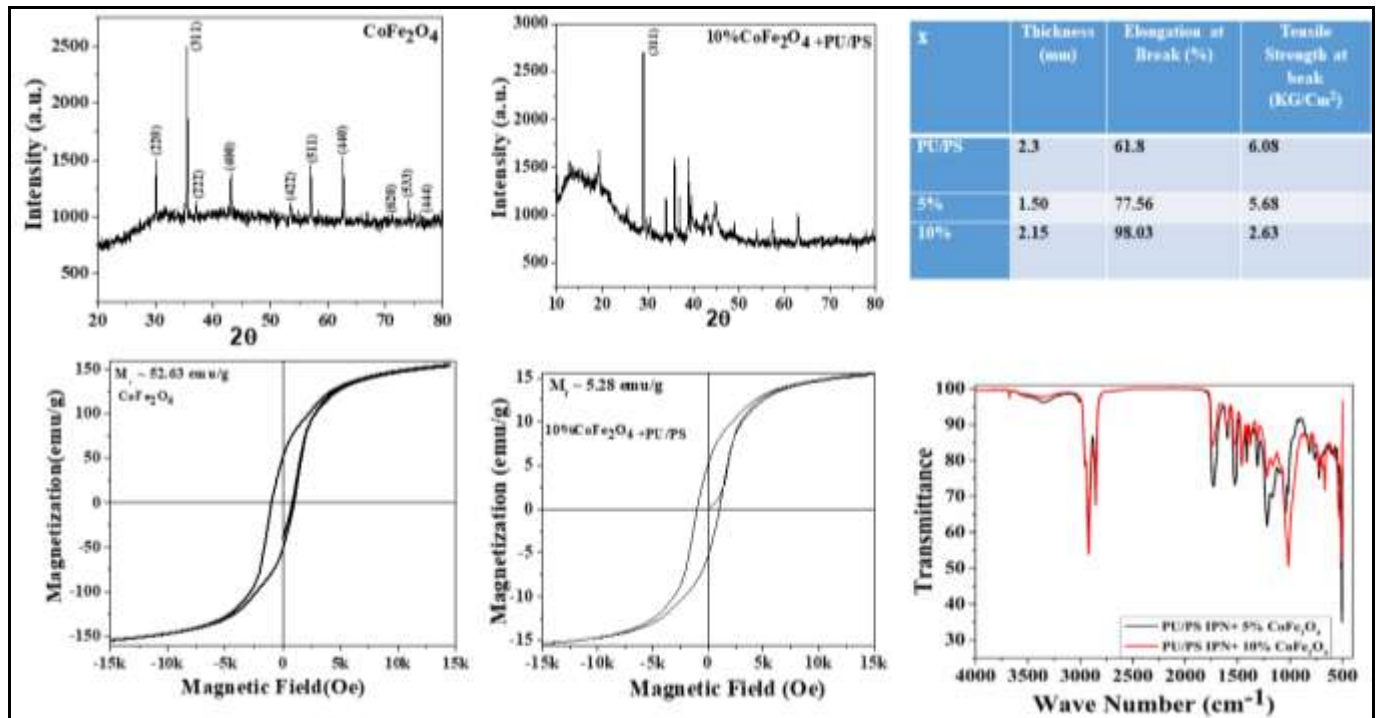


Fig 2: Evidence of Magnetic and Crystalline nature of PU/PS IPN due to incorporation of Ferrite nanoparticles

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