# Chapter 2

# Literature Survey

This chapter presents a synopsis of the research works reviewed pertaining to this present investigation that has been carried out. This review is intended to offer a history of the information available beforehand which would assist in the selection of objectives and methodology for the present research. This dissertation encompasses the various aspects of composite materials required to develop industrial lime sludge waste filled polymeric composites with a special focus on the properties of the composites. The literature survey includes reviews of following topics:

- Composites classification into particulate and fibre reinforced composites with a special focus on CaCO<sub>3</sub> filled polymeric composites and coir fibre reinforced hybrid composites respectively.
- Role of coupling agents on enhancing properties of polymeric composites.
- Role of industrial wastes as fillers in polymeric composites.
- Industrial lime sludge waste and its alternative applications.

A review on the numerous research works conducted on various types of composites (particulate and fibre reinforced) and their properties have been discussed in the first section. Since, lime sludge is a  $CaCO_3$  based waste, hence special focus has been given on the research works conducted in developing  $CaCO_3$  filled polymeric composites. It is a known fact that the interfacial adhesion at the filler-matrix boundary is highly instrumental in enhancing the properties of reinforced composites. Hence, a survey on the various chemical and physical modifications techniques proposed through various research works, and the efficiency of these techniques has been discussed in the second section. Moreover, since the major theme of the intended research is reuse of an industrial waste in polymeric composites, hence the feasibility and viability of different types of industrial wastes being used as filler in polymeric matrix and their effects on the composite properties has been surveyed in the third section. The fourth section comprises of a survey of lime sludge waste in general, and the various alternative applications of reusing industrial lime sludge waste that have been suggested through numerous research works. In the concluding section, a summary of the literature survey has been presented with a view to determine the knowledge gap present in previous investigations.

### 2.1 Composites

The most widely accepted definition of composite has been given by Jartiz [107] who defined composites as multi-functional material systems which show properties that are not exhibited by any distinct material. Composites are a unified configuration made by blending two or more materials, dissimilar in constitution and characteristics but retaining their identities in the combination. Kelly [123] stressed that the composites are not just combination of two materials but it has its own distinctive properties. In terms of inherent properties, a composite exhibits better properties than either of its components and is radically different from its constituents. Beghezan [37] defined composites as - compound materials which differ from alloys by the fact that the individual components retain their characteristics but are so incorporated into the composite that would take advantage only of their attributes and not of their short comings - resulting in materials with improved properties. Additionally, Van Suchetclan [257] defines a composite as a heterogeneous material constituting two or more phases, which are in intimate contact with each other on a microscopic scale. They can be also considered as homogeneous materials on a microscopic scale in the sense that any portion of it will have the same physical property. Although composite materials are broadly categorised into three different types based on their matrices viz. polymer matrix composites (PMC), metal matrix composites (MMC) and ceramic matrix composites (CMC); PMCs find the maximum use in our lives due to their low cost, ease of manufacturing, better strength to weight ratios and ability to be used in diverse applications.

Thus, a polymeric composite is a functionally graded material consisting of two phases - reinforcing phase and polymeric matrix phase; both of them mostly maintaining their individuality in the mixture but still exhibit superior characteristics which usually are not attained with any of the components acting alone. As mentioned earlier, a composite differs from an alloy in the sense that although the reinforcing phase is bonded to the matrix, yet they retain their own identities unlike alloys [259]. Usually, the matrix acts as the binder and provides the required toughness to the composites (transfers stress to the reinforcing phase upon loading) while the reinforcing phase provides the required strength and stiffness to the composites upon loading; thus, incorporating the advantageous properties of both the constituents [156]. The application of composite materials in various walks of life has been so diverse that they play a pivotal role in manufacturing of highly sophisticated machines and equipment. They exhibit many advantages over conventional materials such as light weight, high specific strength and stiffness, high fatigue strength, cheap and easier mode of fabrication and flexibility in tailoring the properties of the composites as per the customer's needs [120]. Composites are used to prepare many products with low cost, low density and better overall properties which find diverse applications in various sectors such as aviation and aerospace (plane components and parts), automobile (brakes, drive shafts, flywheels), manufacturing (pressure vessels), defence (bulletproof jackets, helmets, armoured plating), sports items (golf clubs, tennis and badminton racquets etc) and so on [189]. Thus, researchers over the years have been incessantly toiling to develop composite materials with improved properties for diverse applications and also develop proper combinations of reinforcing and matrix materials.

## 2.2 Classification of polymeric composites

Polymeric composites consist of two constituents - reinforcing agents and matrix phase. The matrix phase usually consists of polymers which can be classified broadly into two categories: (1) thermoplastics *e.g.* high density polyethylene (HDPE), low density polyethylene (LDPE), polypropylene (PP), poly vinyl chloride (PVC) etc; and (2) thermosetting plastics *e.g.* phenols, epoxy and polyester resins etc. The matrix is usually less rigid when compared to the reinforcing phase and therefore results in the higher ductility shown by composites. Additionally, the matrix also acts as the protection for the reinforcing agents from external environmental damage during manufacturing of the composite and while in-service. The reinforcing phase may be broadly categorized into (1) particulate (particles of any size or shape or configuration suspended in a continuous matrix), and (2) fibrous components. Fibre type reinforcing agents are further categorized into (a) short/whiskers type (length to diameter ratio is small) and (b) continuous fibre type (length to diameter ratio is high). Furthermore, the long fibre type reinforcing agents can be arranged unidirectionally or in different orientation (such as  $0^{\circ}$ ,  $45^{\circ}$ ,  $90^{\circ}$  and so on) which would yield different composite properties; while the short fibres can be randomly dispersed or unidirectionally arranged in the matrix.

#### 2.2.1 Particulate composites

Due to their low manufacturing costs and ease of production into near net complex shapes, particulate filled polymeric composites are widely used for various applications. Additionally, particulate composites are isotropic in nature such that their properties are not directionally oriented unlike that directionally aligned fibre composites which are affected by the direction of fibre orientation and also, dissimilarity in the coefficients of thermal expansion of matrix and fibres [206, 252]. Traditionally, particulates in composites are added as filler in order to reduce costs. However, over the years, these fillers have been modified or replaced by other particulates which would add value to the composites in terms of enhancing their properties *viz*. improved processing, mechanical properties, hardness and wear resistance, flame retardancy, thermal conductivity, better electrical, magnetic and dielectric properties.

Numerous polymeric composites are being reinforced with various types of particulates (viz. inorganic, wastes, metallic, ceramics etc) to improve their properties [236], so that they can extend their applications towards electrodes [104] and products with high thermal stability [174]. Bishay et al. [44] investigated the mechanical, thermal and electrical properties of aluminium powder filled polyvinyl chloride composites and found that certain properties of the composites were enhanced. Similarly, research was also conducted on silica particles as fillers in polymeric matrix, whereby they observed enhancement in thermal, electrical and mechanical properties [137, 189]. The characteristics of particulate reinforced polymeric matrix depend on adhesion at the particle-matrix interface, particle size and filler loading. Earlier reports show that the fracture toughness improved when of polymeric composites were filled with nano calcium carbonate [80], layered silicate [81], nano-scale silica [250], and smaller alumina trihydrate particles [198]. It was observed by Rong et al. [210] that nano particle agglomerates filled in polymeric composites led to reinforcing and toughening effects in the composite. Additionally, the size and shape of filler particles also affected the properties of the polymeric composites. This was observed by Yamamoto et al. [267] where they showed that the fracture and tensile properties of polymeric composites were considerably affected by the size as well as the shapes of silica particles. Nakamura et al. [168, 169] also observed that the shape and size of silica particles affected the strength and fracture toughness of composites. It is usually also observed that the strength of a composite is also affected by the interfacial adhesion at the filler matrix boundary. An effective stress transfer between the filler and the matrix upon loading leads to better strength of the composites [195]. Additionally, the type and content of filler particles also significantly affect the mechanical characteristics of composites [187]. Tagliavia et al. [251] investigated the bending characteristics of vinyl ester composites filled with hollow glass particles with an intention of using them for the marine industry. Hassan et al. [100] examined the characteristics of bio-composites reinforced with carbonized maize stalk waste. Dependence of particle size on the static and dynamic compression properties of polypropylene/silica composites was reported by Omar et al. [177]. Gupta and Satapathy characterized the properties of borosilicate glass microspheres filled in epoxy composites and studied their wear characteristics [91]. In an another study, Yadav et al. investigated the impact and flexural properties of saline water treated wheat flour particles reinforced epoxy composites [266].

Various researchers also proposed many models regarding the prediction of composite properties through theoretical modelling. Fracture toughness of particulatepolymer composites was theoretically modelled by Lauke and Fu [138] where they proposed a geometrical model of particle-particle interaction in a regular particle arrangement and studied the influence of particulate filler loading on the mechanical characteristics of the matrix. Jerabek et al. [113] used volume strain measurements for the detection of debonding in particulate filled polypropylene composites under tension. Agrawal and Satapathy [8] investigated the thermal and dielectric properties of AlN reinforced in epoxy and polypropylene and also developed a heat conduction model depicting the thermal conductivity.

#### 2.2.1.1 Calcium carbonate filled polymeric composites

Thermoplastic polymers are often supplemented with inorganic/mineral fillers. Initially, the main objective of adding mineral fillers to polymers was cost reduction. However nowadays, the fillers not only reduce the product cost but also improve their mechanical, thermal and electrical properties, thereby increasing their commercial viability of the composites. Filler materials are used increasingly in order to accomplish added functionality such as enhancing stiffness, decreasing dielectric loss or increasing absorption of infrared radiation [131]. Several studies have been conducted which have demonstrated an increase in toughness with rigid particulate fillers in various systems, *e.g.*, filled polypropylene [27, 194] and filled polyethylene [24, 78–80]. Among the fillers, CaCO<sub>3</sub> is the most widely used filler in polymeric composites not only due to its abundant availability and cheapness, but also due to its ability to improve certain properties in the the composites. CaCO<sub>3</sub> finds its significance in the composites industry during the production of polymeric composites: (i) as reinforcing fillers in thermoplastic industry [136], (ii) for increasing the viscosity of thermoplastics [129], (iii) for toughening (Izod impact toughness) HDPE and PP [32, 286], (iv) to improve Young's modulus of polymers and increasing mechanical properties such as impact strength and hardness [80, 286]; and (v) most importantly, to reduce cost and increase economic viability of the composites [32, 80, 136, 286]. Additionally, several studies have been conducted which have demonstrated an increase in the impact toughness upon addition of rigid particle fillers of very small particle size in polymeric systems, e.g., filled polyethylene [80] and filled polypropylene [194].

Bartczak et al. [32] developed HDPE composites modified by rigid particulate  $CaCO_3$  fillers whereby mechanical properties including notched Izod impact energy of the polymeric composite samples were examined as a function of filler particle size and filler volume fraction. The results showed that not only the toughness of the  $CaCO_3$  filled HDPE composites increased dramatically but the Youngs modulus and impact energy also considerably improved . An impressive increase of impact energy by a factor of nearly 4 was also reported by Wang et al. [80] for polyethylene filled with calcium carbonate particles.

Zuiderduin et al. [286] studied the influence of CaCO<sub>3</sub> particle size (0.07 - 1.9 mm) and surface treatment of the particles (with and without stearic acid) on the toughening properties of CaCO<sub>3</sub> (0 - 32 % volume) filled polypropylene composites. They found that the modulus of the composites increased but the yield stress was lowered with filler content (due to debonding of filler particles from the polypropylene matrix). The impact resistance showed large improvement with particle content - the brittle to ductile transition was lowered from 90 to 40 °C with the addition of CaCO<sub>3</sub> particles and notched Izod fracture energy was increased from 2 to 40 - 50 kJ/m<sup>2</sup>. The stearic acid coating on the particle surface showed better impact strength due to improved dispersion of the CaCO<sub>3</sub> particles. Of all particle types used in the study the stearic acid treated particles of size less than this, tend to agglomerate providing poor dispersion and thus decreasing the impact strength.

Deshmukh et al. [68] also studied the influence of surface treatment of filler particles (with and without stearic acid) on mechanical, thermal and structural properties of poly-butylene terephthalate (PBT)/CaCO<sub>3</sub> composites (0 - 30 % by filler weight).

The composites containing coated CaCO<sub>3</sub> filler particles were found to exhibit better mechanical properties than compared to the composites containing uncoated CaCO<sub>3</sub> particles, with the composite *i.e.* 20 wt % of stearic acid coated CaCO<sub>3</sub>, exhibiting best combination of mechanical properties. Thermal analysis showed that particle type and content had no influence on the melting temperature but the crystallization temperature, % crystallinity and thermal stability increased on increasing the CaCO<sub>3</sub> content in PBT matrix. Morphological observation indicated stearic acid is a favorable coupling agent rendering the filler polymer interfaces compatible thus paving the road towards better mechanical and thermal properties. However in an another research, Osman et al. [179] showed that excess coating of filler particles (calcium carbonate) with stearic acid or calcium stearate proved to be detrimental to the mechanical properties of low density poly ethylene (LDPE)/CaCO<sub>3</sub> composites and suggested only optimal coating of the filler particles for improved mechanical properties.

Zhou et al. [283] studied the properties of calcium carbonate-poly ether ether ketone (PEEK) composites with filler particles surface treated with sulfonated PEEK (SPEEK) and found that both filler particle size and surface treatment of filler particle affected the mechanical properties of the composite. Kamble et al. [116] blended thermoplastic polyurethane (TPU) and poly olefins (LD, HD and PP) while using polypropylene copolymer (PPCP) as compatibilizer and CaCO<sub>3</sub> as filler. They showed that PPCP was a good compatibilizer for TPU/LD, TPU/HD and TPU/PP blends and addition of calcium carbonate as filler increased the mechanical properties such as tensile strength, impact strength and hardness. Kwon et al. [136] studied the mechanical properties and melt viscosity of unfilled and calcite (calcium carbonate) filled polyethylene using HDPE, LDPE, and LLDPE composites. In addition, they also proposed a mechanism of formation of air holes between the calcite and the resin matrix upon stretching. The tensile stress and the complex melt viscosity of the calcite filled polyethylene composites were higher than that of unfilled ones, thus proving the reinforcing effect of the calcium carbonate.

More recently, Kato [119] developed a new hybrid material - polymer/calcium carbonate layered thin film composites by combining insoluble chitin or chitosan solid matrices with soluble acidic macromolecules such as poly (acrylic acid) to induce thin-film crystallization of CaCO<sub>3</sub> on the solid matrices. Hybrid composite material formed that way has the potential of showing high functionality as well as environmental benignity. Mohesenzadeh et al. [164] studied the deformation techniques of medium density polyethylene/calcium carbonate nano-composites using advanced techniques such as essential work of fracture (EWOF) and transmission optical microscopy (TOM). EWOF analysis proved that nano-particles of calcium carbonate added to polymer matrix decreased the essential work of fracture causing a detrimental effect due to agglomeration of nano-particles in the matrix.

#### 2.2.2 Fibre reinforced composites

An important category of structural materials which are in vogue nowadays are fibre reinforced polymeric composites due to their ease of manufacturing, flexibility in design and ability to exhibit properties tailor made for specific applications. Fibre reinforced composites consists of fibres implanted in a matrix with discrete interfaces between the two constituent phases. The fibres exhibit properties such as superior rigidity and strength, acting as the main load bearing component while matrix offers the toughness in order to sustain the stress applied. Additionally, the matrix also acts as a shield, thereby protecting the reinforcing fibres from weathering or atmospherical degradation all throughout fabrication and subsequently, while in-service. A variety of fibres are available for use *viz*. synthetic fibres such as glass, aramid and carbon; and natural fibres viz. coir, jute, hemp, sisal etc. At present, fibre reinforced composites are regularly employed in varied applications viz. household goods, aeroplanes, offshore structures, automobiles, piping, sporting goods, space vehicles, electronics and appliances. The various advantageous properties of fibre reinforced composites are light weight, high specific strength, high corrosion and impact resistance and excellent fatigue resistance. The fibres can be aligned and layered as per tailor made combinations which provide excellent structural and functional properties viz. high specific stiffness and strength, to these composites.

Ramesh et al. [203] studied the properties of sisal-jute-glass fibre reinforced polyester hybrid composites with respect to the fibre content (ratios 40:0:60, 0:40:60, and 20:20:60) and fibre orientation (0° and 90°). They found that these parameters affected the strength and water absorption properties of the composites. Cho et al. [58] investigated the properties of graphite nanoparticle filled carbon fibre-epoxy composites and found that a 5 wt % graphite nanoparticles and 55 % fibre volume fraction, provided 16 % improvement in longitudinal compressive strength and inplane shear properties. Effects of coupling agents on the enhancement of properties of sisal fibre reinforced polymeric composites were also studied earlier by Haque et al. [97]. Ota et al. [180] found that the melt flow index, tensile strength and elastic modulus of glass fibre reinforced polypropylene composites depended on the fibre length and fibre content. In another research, micro-fibres were used to improve

the tensile strength of cement based composite materials by Betterman et al [39]. Oliveira et al. [64] introduced 0, 0.5, 1.0 and 1.5 vol % of fibres to improve the properties of a typical mortar mix of cement, hydrated lime and natural sand (ratio of 1:1:6) and found no fibre balling/bundling or other abnormalities. Berthelot and Sefrani [38] calculated the damping of glass and kevlar fibre reinforced composites using the impulse hammer technique and compared the results with existing models. Velmurugan and Manikandan [258] found that the mechanical properties such as tensile, impact, shear and bending properties of rooflite composites improved by the addition of randomly oriented short fibres of palmyra and glass. The optimum properties of the composites were observed at fibre content of 50 wt % and and fibre length of 50 mm. Kaichang et al [143] evaluated the feasibility of improving the interfacial adhesion of natural fibre based polymeric composites by the using various chemical methods. Khalil et al. [126] reviewed the reinforcing and plasticizing effects of natural fibre reinforced poly vinyl chloride composites with a focus on the effects of coupling agents on composites. Abdulmajeed et al. [5] and Garoushi et al. [83] discussed about the role of unidirectional long and short glass fibres on improving the mechanical characteristics of polymer composites. Karsli and Aytac [118] investigated the thermo-mechanical characteristics of polyamide-6 (PA6) composites filled with short carbon fibres, while Bhattacharjee and Bajwa studied the degradation in the properties of natural fiber reinforced polymeric composites due to recycling [42]. Sridhar et al. [248] focussed his research towards fabricating customized hip prosthesis made out of poly-ether-ether-ketone composites filled with carbon fibres. Also, continuous quartz fibre reinforced functionally graded composites (FGCs) were developed for aerospace applications by Kumar et al. [135]. Pandey et al. reused fibres from distiller's dried grains with solubles (DDGS) and corn grains as fillers in HDPE obtained from bio-based and petroleum sources [183]. Safwan et al. investigated the tensile and impact characteristics of bamboo-kenaf fibre reinforced epoxy composites [217], while Saba et al. improved the flame retarding capacity of polymeric composites reinforced with kenaf fibres [216]. Zadeh et al. enhanced certain properties of polymeric composites reinforced with date palm leaf fibres in case of recycled ternary polyolefins composites [278]. Earlier, Singh and Palsule self compatibilized jute fibre-polypropylene composites and coconut fibre-HDPE composites by Palsule process and characterized their properties [238, 239].

#### 2.2.2.1 Coir fibre based composites

Rising environmental awareness has brought over a paradigm shift towards designing eco-friendly materials and reusing/recycling hazardous wastes. Hence, over the past few years, natural fibres have drawn substantial interest from researchers as a potential replacement for synthetic fibres such as glass, aramid etc in polymeric composites [115]. In addition to mitigating environmental issues of using hazardous and nonbiodegradable synthetic materials, natural fibres also offer advantages such as low cost, versatility, lightweight, biodegradability, renewability, high specific strength and modulus, lower equipment abrasion, and energy consumption during processing [235].

Among all natural fibres, coir is an important lignocellulosic fibre obtained from coconut trees grown extensively in tropical countries like India [34]. Because of its hard-wearing quality and durability, it is used for making a wide variety of floor furnishing materials, yarn, rope etc. [212]. Traditional coir products consist of a small fraction of the total production of coir world over. Hence, efforts for alternative uses of coir have been underway, including utilization of coir as reinforcement in polymer composites [212]. Coir fibre has low cellulose content, high microfibrillar angle, is not as brittle as glass fibre, can be chemically surface modified, is non-toxic (no waste disposal issues), water proof, resistant to salt water, and has lower tensile strength and high elongation compared to other lignocellulosic fibres [111, 165]. Hence, numerous researches on coir fibre based polymeric composites have been conducted till now. Earlier, coir fibre-polyester composites were tested as helmets, roofing, and post-boxes [234]. Moreover, mechanical properties of randomly oriented coir-epoxy composites exhibited tensile, flexural, and impact strengths of 17.86 MPa, 31.08 MPa, and 11.49  $kJ/m^2$  respectively [98]. Similar research on the mechanical properties of coir fibre polyester composites were evaluated [213]. Effect of length and weight fractions of coir fibre on mechanical properties was also studied [110].

However, the main hindrances in using natural fibres in polymeric matrices are incompatibility at the interface of hydrophilic fibres and hydrophobic matrix, and also high moisture absorption of fibres causing dimensional changes in them [212]. This is an important fact as the efficiency (mechanical properties) of a composite depends on the fibre/matrix interface and the ability to transfer stress from the matrix to the fibre. Moreover, moisture content in hydrophilic natural fibres can vary from 3 - 13 %, resulting in a weak interface [209]. Additionally, the performance of coir as a reinforcement in polymer composites is unsatisfactory due to its cellulose (36 - 43 %) and hemicelluloses content, high lignin content (41 - 45 %), and high microfibrillar angle. It has been reported that removal of the outer sheath of lignin usually results in a better bonding at the interface. Hence, coir fibres are surface treated with an alkali (such as NaOH) in order remove the lignin on the surface, before using it in composites, so as to improve the bonding at the interface between the matrix and fibres [84, 212]. Rout et al. [212] studied the mechanical properties of chemically treated coir fibre reinforced polyester composites whereby coir surface was modified by alkali, bleaching and vinyl grafting. They found that 5 % alkali (NaOH) treated fibres showed improvement in the properties of coir composites. Li et al. [145] reviewed the alkaline, acetylation, silane, benzoylation, isocyanates, permanganate and other types of treatments on natural fibre composites and reported that modification of the fibre surface could enhance the compatibility of hydrophobic thermoset matrix and hydrophilic cellulose fibres. Orue et al. [178] investigated

(NaOH) treated fibres showed improvement in the properties of coir composites. Li et al. [145] reviewed the alkaline, acetylation, silane, benzovlation, isocyanates, permanganate and other types of treatments on natural fibre composites and reported that modification of the fibre surface could enhance the compatibility of hydrophobic thermoset matrix and hydrophilic cellulose fibres. Orue et al. [178] investigated the effect of alkaline and silane treatments on mechanical properties and breakage of sisal fibre composites. They found that alkali and alkali+silane treated composites showed better tensile strength values. Mulinari et al. [166] reported that the fatigue life of alkaline (sodium hydroxide solution 1 % w/v) modified coconut fibre reinforced polyester increased upon surface modification. Nam et al. [170] studied the mechanical properties of the alkali treatment (5 wt % NaOH) coir fibre filled poly (butylene succinate) biodegradable composites which revealed that a remarkable improvement was achieved in the interfacial shear strength (IFSS) and mechanical properties of treated coir fibre/polybutylene succinate (PBS) composites (25 wt %fibre content exhibit higher mechanical properties). Wong et al. [264] studied the interfacial adhesion between the fibre and matrix of alkali treated fibre reinforced composites. In this study, two types of composites were made by using untreated bamboo fibres and NaOH treated bamboo fibres under the concentrations of 1, 3 and 5 wt %. The study found that due to the chemical treatment, the strength and stiffness of composites were improved than the untreated fibre composites specifically for 5 wt % NaOH treated fibres. Prasad et al. [190] alkali treated coir fibres with 5 % NaOH solution and used them in polyester composites. They found out that debonding stress of alkali-treated fibres was 90 % higher than that of untreated fibre reinforced composites. Haque et al. [96] treated raw palm and coir fibres with benzene diazonium salt and 5 wt % NaOH in order to improve their compatibility with a polypropylene matrix. It was observed that the treated fibre reinforced composites exhibited better mechanical properties than untreated ones. This shows that chemical modification of natural fibres including coir fibres enhances the compatibility of fibres with the polymeric matrix, which consequently improves the overall

characteristics of the composites.

#### 2.2.3 Multi component hybrid composites

A sensible mixture of filler or reinforcing agent and a matrix can develop a composite whose properties such as strength and modulus can outmatch those of conventional structural metallic materials [106]. It has been observed though various researches that the physico-mechanical features of a composite can be further modified by the addition of a particulate filler and a fibre to the matrix during fabrication. This is done in order to reap the benefits of both reinforcing agents. Hence, studies on a variety of hybrid composites with fibre-fibre, fibre-filler and filler-filler combinations are in vogue now, as they tend to further improve the properties of composites; thereby increasing their commercial viability.

Of late, it has been proved that solid particulate fillers incorporated in a fibre reinforced composites yielded advantageous results in the form of increased rigidity (modulus), reduced product costs but with certain shortcomings such as lower strength and impact toughness [74, 130]. Gracia at al. [86] were among the first researchers to propose the concept of hybrid composites for improved characteristics of fibre reinforced composites. Subsequently, Liau et al. [147] observed that the impact toughness significantly improved upon the addition of ceramic whiskers or particulates in the hybrid composites. Similar researches have been attempted in the recent past in order to study the physico-mechanical and tribological behaviour of hybrid composites [43, 260]. In recent times, a substantial amount of research on hybrid composites with a variety of filler-fibre combinations have been conducted viz. bioparticles-coir [204], crab carapace-coir [201], carbon black-glass [89], soysugarcane bagasse [50], powdered hazelnut shells-jute [160] etc. in various polymer matrices. Hartikainen et al. [99] observed that the fibre orientation, length, distribution and mechanical properties of long glass fibre reinforced polypropylene filled with  $CaCO_3$  are all affected upon filler addition. Herrera et al. [101] fabricated a hybrid composite material of HDPE, sand and short henequen fibres. They found that the tensile modulus and flexural properties improved upon sand filler addition.

Additionally, tailor made properties of hybrid composites are also obtained through the reinforcement of two or more types of fibres in a polymeric matrix. It was observed earlier that the ductility of hybrid composites filled with carbon and glass fibres increased upon the addition of glass fibre. It was evident that the higher strength carbon fibres imparted low ductility to the composites. This was compensated by the addition of ductile glass fibres which increased the overall elongation of the composites [62, 63, 157]. Dong and Davies [70] studied the flexural

properties of S-2 glass and T700S carbon fibre reinforced epoxy composites. Raja et al. [200] demonstrated the mechanical characteristics of E-glass fibre filled polymeric composites impregnated with fly ash. Patil et al. also studied the effects of adding nanostructured fly ash as filler on the properties of chitosan composites [186]. Mahapatra [152] fabricated a multi-component hybrid polyester based composite filled industrial waste solids and reinforced with E-glass. Gangil et al. [82] fabricated vinyl ester composites filled with cement by-pass dust (CBPD) particles and also reinforced with short kevlar fibres. Saradava et al. [223] investigated the mechanical properties of red mud filled coir fiber reinforced polymer composites. Moreover, the composites can be also tailor made in different arrangements (fibres may be properly mixed in the matrix and arranged in specific manner in terms of interlaminates and orientation). This would provide specific properties based on the design of the composites.

### 2.3 Role of compatibilizers in composites

The efficiency with which a matrix transfer stress upon loading to the reinforcing agents in a composite is reduced to a great extent due to weak adhesion at the interfacial boundary. Thus, a huge amount of research has been conducted till date, focussing on the improvement of interfacial strength of a composite through chemical treatments and surface modification of either the matrix or the reinforcing phase. The observed trend suggests that the chemical and physical modification of either the reinforcing phase or the matrix phase is significant in increasing the interfacial adhesion and thereby, is instrumental in enhancing the properties of composites. A variety of chemical modification techniques have been proposed over the years *viz.* alkalis, silanes, fatty acid (stearic acid), acetylation, benzoylation, acrylation and acrylonitrile grafting, maleic anhydride coupling, permanganate, peroxide, and isocyanate treatment [75]; along with a few physical modification techniques *viz.* corona and plasma treatment [35, 75, 241]. However chemical modification techniques outperform its physical counterparts in providing the better results, and hence are more popular [75].

Among the chemical modification techniques, stearic acid treatment, alkaline treatment (with NaOH), acetylation, maleic anhydride grafting on polymers and silane treatment are the most popular due to their cheapness and ease of treatment.

Homogenous dispersion of inorganic particles in an organic polymer matrix is

difficult due to the strong tendency of the particles to agglomerate and generate high viscosity during composite processing. Moreover, inorganic mineral fillers are hydrophilic while organic polymers are hydrophobic; this causes the problem of low interfacial adhesion [105]. Hence, inorganic fillers are often surface modified by fatty acids, silanes, titanates, phosphates, etc. in order to improve their compatibility and dispersability [3]. In case of calcium carbonate based fillers (such as lime sludge) the most widely used surface coatings are fatty acids, usually stearic acid where a layer of hydrophobic organic molecules is attached to the filler surface [133]. When fatty acids are adsorbed onto carbonate surface, a monolayer of hydrophobic organic molecules is attached to the mineral surface. This ultrathin organic film strongly influences the final properties of the composites as it is the interface, which determines the particle-particle as well as the particle-matrix interactions. Fatty acids such as stearic acid help in uniform dispersion of particles in the polymeric matrix as the tendency of the coated filler particles to agglomerate reduces substantially. Additionally, stearic acid is cheaper and more widely available than its contemporary surface modifiers such as silanes, maleic anhydride, titanates, etc. Premphet et al. [192] fabricated stearic acid coated CaCO<sub>3</sub> filler in polypropylene and found that the stearic acid coating promoted  $\beta$ -hexagonal crystallization of PP which resulted in an increase in the impact strength of the composites. Zuiderduin et al. [286] found that stearic acid coating on  $CaCO_3$  particles exhibited a large increment in impact strength and led to the toughening of the composites due to the improved dispersion of the  $CaCO_3$  particles. Deshmukh et al. [192] studied the properties of stearic acid coated  $CaCO_3$  filled poly-butylene terephthalate (PBT) composites and found that the systems containing stearic acid coated  $CaCO_3$  exhibited better mechanical properties when compared to the uncoated ones, with 20 wt % of steraic coated  $CaCO_3$  filled PBT composites exhibiting best mechanical properties. Maged et al. [179] found that coating the filler with an optimal amount of coupling agent so that it covers the filler surface with an organic monolayer is most advantageous for the composite properties. Kim et al. [127] realised that stearic acid on zeolite surface enhanced flexibility of polymer matrices; while in case of LLDPE and HDPE composites, the modulus, elongation and impact properties improved upon higher filler loading.

Coupling agents were also used in case of fibre reinforced composites for effective interfacial adhesion. Earlier, Rout et al. [212] studied the mechanical properties of chemically treated coir fibre reinforced polyester composites whereby coir surface was modified by alkali, bleaching and vinyl grafting. They found that 5 % alkali (NaOH) treated fibres showed improvement in the properties of coir composites. Li et al. [145] reviewed the alkaline, acetylation, silane, benzoylation, isocyanates, permanganate and other types of treatments on natural fibre composites and reported that modification of the fibre surface could enhance the compatibility of hydrophobic matrix and hydrophilic cellulose fibres. Orue et al. [178] investigated the effect of alkaline and silane treatments on mechanical and breakage of sisal fibre composites. They found that alkali and alkali-silane treated composite showed better tensile strengths. Mulinari et al. [166] reported that the fatigue life of alkaline (sodium hydroxide solution 1 % w/v) modified coconut fibre reinforced polyester increased upon surface modification. Saidi et al. found that the various properties of rice husk reinforced polymeric composites were improved when titanate was used as the coupling agent [219].

Acetylation of reinforcing fibres for increasing the interfacial adhesion results in esterification whereby acetyl groups (CH<sub>3</sub>CO) and hydroxyl groups (OH) of natural fibres react with each other leading to increased hydrophobocity [102]. This results in enhanced interfacial adhesion, tensile and flexural strength, stiffness, thermal stability and resistance to fungal attack in case of natural fibre reinforced composites [48, 125, 255]. However, it is important to note that excess acetylation treatment results in degradation of impact properties of the composites.

Silanes are also used for chemical modification at the interface as they form a bridge between hydrophilic groups of the fibre and hydrophobic groups in the matrix. The most commonly reported silanes used are amino, methacryl, glycidoxy and alkyl silanes. It has been observed that silanes increase the hydrophobicity of natural fibres; thereby improving the mechanical properties of natural fibre based composites. This is due to the improvement in the interfacial bonding which increases on account of the covalent bonds linking silanes and the matrix [1, 197].

Over the years, maleic anhydride (MA) grafted polymers are commercially the most widely used coupling agents used to improve the interfacial bonding in composites. MA is grafted on to the same polymer which is intended to be used as the matrix. This is done in order to warrant compatibility between the matrix and the coupling agent. One of the most common coupling agent of this class is maleic anhydride grafted polypropylene (MAPP) which reacts with the hydroxyl groups on the reinforcing fibres or fillers, leading to covalent or hydrogen bonding at the interface. This increases the interfacial adhesion leading to the enhancement of mechanical properties of the composites. Maleic anhydride grafted polymers can be used as an additive along with the matrix and reinforcing phase during fabrication or grafted to the reinforcing phase prior to fabrication. It was observed that the tensile and flexural properties have improved in addition to increased stiffness in polypropylene composites when MAPP is used as a coupling agent. Although the impact strength was observed to decrease with addition of natural fibres to tough matrices such as PP, the reduction was less significant upon the addition of MAPP. In addition to improved interfacial adhesion, the maleic anhydride grafted polymers were instrumental in increasing the wettability of the reinforcing phase and enhanced dispersion which resulted in increasing the mechanical properties [122]. In similar lines of work, use of MA grafted poly lactic acid (PLA) as the compatibilizer enhanced the mechanical properties of PLA composites as well as their thermal stability [21, 117, 277]. Similar enhancement in the mechanical and thermal properties were obtained when maleic anhydride grafted polyethylene (MAPE) was used [163, 176, 282]. Earlier, curaua fibre reinforced HDPE composites were treated with MAPE coupling agent which yielded better thermal stability and higher fibre-matrix interaction at the interface [16]. Mohanty et al. [16] found that the properties of jute fibre reinforced HDPE composites increased significantly upon the addition of MAPE as compatibilizer when compared with untreated ones. It was observed that the strength of the composites increased by almost two folds upon using MAPP as a compatibilizer when compared with those obtained by using silanes [36]. Maleic anhydride coupling agent was also used as the coupling agent to enhance the physical and mechanical properties of wood filled biodegradable polymer composites [274]. Thus, from literature it is observed that maleic anhydide grafting on polymers can be regarded as one of the most efficient method of improving the interfacial adhesion among all the chemical modification processes.

#### 2.4 Industrial waste based composites

Recycling and reuse of wastes has become a mandatory requirement today due to environmental concerns raised on account of unscientific landfilling, incineration and lack of biodegradation. Reuse of industrial wastes in polymeric composites as particulate or fibrous fillers has generated immense interest from researchers as well as industrialists due to their ability of enhancing the properties of the composites, extending the ranges of their applications in addition to mitigating the disadvantages of environmental pollution. Thus, numerous researches have been conducted till date to reuse various industrial wastes as fillers/reinforcing agents in polymeric composites, as a process of sustainable waste management. Industrial wastes *viz.*  biochar [61], post consumer recycled wood [242], electronic wastes [243], non-metals recycled from waste printed circuit boards [270], thal silica [9], mechano-chemically and mechanically activated waste fly ash [31], industrial fly ash cenospheres [66], red mud wastes [196], discarded nylon carpets [182] etc., are being used as fillers or reinforcing agents in a polymeric matrix in order to increase their functionality and decrease pollution.

Das et al. [61] reported that highly carbonised fine biochar particles with high surface area acted as a reinforcing agent in the wood/polypropylene biocomposites. A biochar content of 24 wt % exhibited the best mechanical properties with improved tensile and flexural properties while a biochar content of 12 wt % and 18 wt% showed the maximum ductility and thermally stability respectively. Sommerhuber et al. [242] revealed that composites made out of recycled waste HDPE and post consumer recycled wood exhibited comparative properties with those composites made from virgin resources. Sommerhuber et al. [243] also used recycled acrylonitrilebutadienestyrene and polystyrene obtained from post-consumer electronics in wood-polymer composites and showed that properties of the composites such as stiffness and strength (both tensile and flexural) improved as a result of waste filler addition. Yang et al. [270] used non-metals derived from recycled waste printed circuit boards as reinforcing fillers in HDPE composites. They revealed that the high aspect ratios of non-metals and better interfacial adhesion resulted in higher stiffness of the composites. Ahmad et al. [9] observed that the tensile, hardness and impact properties of epoxy based hybrid composites increased with the addition increasing thal silica sand nano-particles derived from waste desert sand. Deepthi et al. [66] used fly-ash cenospheres derived from the coal fired power plants as fillers in HDPE composites after surface modifying them with silane coupling agents. They found that the composites were comparatively light in weight due to the addition of hollow fillers and also exhibited better tensile properties and thermal stability. Pan et al. [182] prepared composites using nylon 6/nylon 6,6 waste carpets and found that these composites have much higher sound absorption, water stability and mechanical properties than its jute-polypropylene counterparts. Additionally, they observed that the styrene-butadiene-rubber/calcium-carbonate binders and unshredded nylon 6,6 carpet yarns used in the composites made them strong. Ahmetli et al. [11] studied the properties of epoxy-epoxy toluene oligomer composites filled with marble processing waste. They also found that the composites exhibited higher thermal stability in addition to higher tensile strength and hardness. Zheng et al. [99] used the non metallic materials obtained from waste printed circuit boards and used them

as fillers in polypropylene composites to find out that the mechanical properties such as tensile strength, tensile modulus, flexural strength and modulus are improved upon filler addition. Chaowasakoo and Sombatsompop [56] investigated the properties of fly ash/epoxy composites using conventional thermal and microwave curing methods. Various other researches were also conducted using fly ash waste as filler in epoxy matrix and features such as effect of porosity and damping properties were studied [87, 249]. Patnaik et al. [187] investigated the wear characteristics of polyester composites reinforced with glass fibres and filled with waste fly ash, using Taguchis design of experiments. Patnaik et al. [188] also studied the effects of adding kiln-dust of a cement plant and flakes of pine-bark as the filler in polyester. In an another research, Asaad and Tawfik [17] used cement dust waste as filler in recycled polystyrene. Siddhartha et al. [237] investigated the wear properties of CBPD filled epoxy based functionally graded composites. In similar lines of work, an industrial waste obtained as a by-product of alumina production was used as a filler in coirfibre-reinforced polymer composites [196]. Results showed the compressive strength, density and hardness of the composites improved with the addition of waste filler due to the presence of metallic oxides in red mud. Recently, Mahata et al. [153] used titanium enriched red mud in order to develop aluminium titanate-mullite composite. One other researches include studies of thermal and mechanical characteristics of polypropylene composites filled with red mud [12, 281]. However, most of the studies with red mud as a filler in polymeric composites are targeted with the prospect of making low cost composites in addition to reusing an industrial waste, thereby avoiding pollution [41]. Additionally, another industrial waste, copper slag was used by Biswas and Satapathy [46] as filler in glass-epoxy composites and they found that the tensile rigidity, flexural strength, hardness and impact strength increased with increasing filler loading. Subsequently, Biswas et al. [47] investigated the combined outcomes of using copper slag and red mud on the physico-mechanical properties on bamboo reinforced epoxy hybrid composites. Moreover, of late, blast furnace slag derived from iron making has also been used in polymeric composites to improve certain properties [181]; while property study of original and modified steel slag reinforced epoxy composites was also conducted [94, 95]. Saidi et al. studied the effect of rice husk filler on the biodegradability and mechanical properties of montmorillonite reinforced low density polyethylene nanocomposite films [154].

## 2.5 Industrial lime sludge waste

Lime sludge mainly consists of lime as calcium carbonate (CaCO<sub>3</sub>) which on heating provided a chemical composition of 40-50 % CaO, along with small amounts of Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, Fe<sub>2</sub>O<sub>3</sub> and traces of alkali oxides after a loss on ignition (LOI) of 35-40 %. This implies that almost all of lime sludge consists of calcium carbonate and some other ceramic oxides (small amounts Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub>) [2]. Lime sludge is primarily produced from the paper, carbide, fertilizer and soda ash industries and consists of lime in the form of calcium carbonate (CaCO<sub>3</sub>) as the major constituent. Since, lime sludge waste from the paper industry is used in this research, hence the lime sludge production process is described below.

The principal process for producing chemical pulps in paper industries is the Kraft process, wherein the mixtures of sodium sulphide and sodium hydroxide are used as pulping chemicals. In this process, calcined lime is used for regeneration of caustic soda by conversion of soda ash (green liquor) leaving behind lime (calcium carbonate) sludge as a waste [2]. The reaction depicting Kraft's process is shown in Eq. (2.1).

$$Na_2CO_3 + CaO + H_2O \rightarrow NaOH + CaCO_3$$
 (2.1)

The calcium carbonate generated as a result of Kraft's process is washed with water and filtered to recover the alkalis. The calcium carbonate thus produced can be again converted to CaO through recycling process. But, the process is uneconomical and expensive which results in lime sludge being disposed off as wastes. As per the report of the Central Pollution Control Board (Govt. of India), lime sludge produced from the various industries in India, is usually disposed off in open or closed dumps or used in unscientific land filling causing environmental pollution [2].

Until a few decades ago, utilization of paper and pulp industry solid wastes entirely involved the conventional methods of incineration, land filing and land application; which are actually potential pollution hazards and threat to the surrounding ecological systems. However, triggered by the urgent need to develop environmental friendly waste utilization methods, Wiegand et al. [262] suggested some unconventional methods of pulp and paper industry lime sludge waste utilization - such as recovery of raw materials from sludge, production of bricks, ceramics and building materials from sludge, sludge pelletization for alternative fuel, feedstock for ethanol production and even as potential composts. Recently, Torkashvand et al. [254] studied the effect of paper mill lime sludge as a soil adjustment in acid soil and found that a 2 % addition of lime sludge remarkably increased shoot dry matter and micronutrient uptake in *sorghum vulgaris* plant. Evalyno et al. [73] conducted feasibility studies of using dewatered paper sludge from Virginia Paper Corporations as a potential potting soil replacement. They found that paper sludge acted as stable compost which may be used as a fertilizer, soil amendment material or nutrient supplement source along with a potting medium rather than as a potting medium alone. Moreover, Ahmadi et al. [10] utilized paper sludge as a potential building block by adding it to concrete. They found that although addition of sludge decreased concrete density but a compressive strength of 8 MPa was achieved by using 5 % of paper sludge waste, and it is acceptable for use in masonry construction.

In India, Singh et al. [240] had deduced the application of waste lime from fertilizer industry when they showed that vateritic nature of lime sludge does not pose problems for production of E class lime (used in masonry mortars). Additionally, the hydrated lime is suitable for making composite mortars of improved strength by addition of calcined clay or flyash or Portland cement and sand. A report by the National Council for Cement and Building Materials (India) [4], showed that conversion of lime sludge from paper mills into quick lime in vertical shaft kilns is an attractive and economically feasible proposition which will enhance productivity of lime kilns. Deka et al. [67] showed that lime sludge waste mud obtained from the paper mill at Jagiroad, Assam (Hindustan Paper Corporation Limited) can be successfully used in management of pH in acidic waters of fish ponds as it showed some beneficial effects on the production of fish. Although 1.5 times the amount of lime sludge was required to be added in comparison to pure lime, the parameters such as pH, conductivity, free carbon dioxide, total hardness and dissolved oxygen increased with the addition of lime sludge which was beneficial for fish production.

In the last few decades, paper sludge waste was also used as filler in polymeric and rubber composites [103, 244, 245]. However, paper sludge consists of only 70 % inorganic matter (mainly (CaCO<sub>3</sub>)) along with 30 % cellulosic fibres in it which has its own disadvantages [85, 245]. It is observed that cellulose in natural fibres hinder bonding between the filler and matrix due to hydrophilic fibre and hydrophobic polypropylene. Hence, most natural fibres are pre-treated with a coupling agent before their use as reinforcing agent in polymeric composites [85, 245]. Since the main objective behind using industrial wastes as filler is cost reduction, the pretreatment of paper sludge adds to additional expenditures when done on industrial scale. Moreover, particle size of paper sludge is usually much higher (up to 5 mm) due to the presence of cellulosic fibres [85]. This increases stress concentration sites in composites leading to decreased mechanical properties. Higher particle size also leads to high water absorption rate of the composites which is detrimental for their properties. Additionally, low temperature resistance of organic fibres pose another problem of degradation/decomposition at extrusion/molding temperatures around 200 °C during processing of thermoplastic composites [16]. As per report of the Central Pollution Control Board (Govt. of India), lime sludge waste from various paper industry mainly consists of lime as  $CaCO_3$  along withe some other minerals and traces of alkali oxides. However, no traces of cellulosic fibres are reported to be present in lime sludge waste from the paper industry. Additionally, particle size of raw lime sludge varies from 30-45 m and is stable at higher processing temperatures due to the absence of any organic matter [2]. Feasibility studies of using paper industry lime sludge as potential filler in the manufacturing of polyethylene composites have not been conducted till now. Hence, in this research, lime sludge obtained from the paper industry is used as a filler in polymeric composites.

## 2.6 Prediction of composite properties

An effective way of reducing complications and rigors of experimental tests is to develop robust mathematical models for predicting parameters related to composites viz. process parameters, mechanical properties etc. Numerous attempts have been made over the years to develop and optimize mathematical models for accurate prediction of composite properties. Earlier, regression based models were developed for glass fibre reinforced composites and compared with fuzzy models in order to develop predictive models for thrust force and drilling during drilling of these composites [108]. Regression based models coupled with evolutionary algorithms were also proposed earlier for optimization of machining characteristics of glasscoirpolyester hybrid composite [109]. Yildiz et al. [276] used regression analysis to correlate the variation in the mechanical properties of wood-polymer composites with respect to polymer-filler loading. Yang et al. [268] used linear regression analysis in order to determine the relation between the composite parameters viz. thickness swelling and water absorption rate, as a function of filler loading in case of lignocellulosic filler-polyolefin bio-composites. Athijayamani et al. [19] used nonlinear regression analysis in order to establish quantitative relationships between the tensile and flexure properties of randomly oriented natural fibres/polyester hybrid composite with respect to fibre content and length. Braem et al. [51] used an an exponential regression analysis in case of dental composites, to associate each composite property as a function of volumetric filler content and found an excellent

correlation between the parameters and the filler content. Balaji and Javabal [29] developed an artificial neural network algorithm to predict the mechanical behaviors of zea fibrepolyester composites and found an average absolute error percentage of less than 4 % in the models developed for tensile, flexural and impact properties. The requirement of regression and ANN models in calcium carbonate impregnated coir-polyester composites paved the way towards development of regression models for the multi objective problems [112]. Earlier, optimization of mechanical behaviour of alumina impregnated coir-polyester composites were conducted using response surface methodology [231]. In another study, the mechanical behaviour of hybrid bioparticles (alumina and calcium carbonate) impregnated coir-polyester composites were predicted using soft computing. Additionally, soft computing techniques such as artificial neural network and response surface methodology were used for prediction and optimization of mechanical behaviors of particulate coirpolymer composites [29, 232]. Li et al. [146] modelled the compressive and flexural properties of hemp fibre reinforced concrete composites using a simple empirical linear mathematical expression based on statistical analysis and regression. Leprince et al. [142] performed linear regression analysis in order to correlate the dynamic and static elastic moduli with the filler mass fraction in case of inorganic filler added low shrinkage resin composites. Rajmohan et al. [202] developed a linear regression model in order to predict the mechanical properties as a function of nano copper oxide (CuO) filler loading in case of glass fibre reinforced - nano CuO dispersed in polystyrene composite. Pujari et al. [193] modelled jute and banana fibre reinforced polymeric composites as a function of fibre loading using regression analysis and artificial neural networks and found satisfactory results. Thus, prediction models based on computing techniques proved effective in modelling properties of composite materials.

## 2.7 Opportunities for research and development

Today polymers and polymeric composites constitute the building blocks of almost every commercial item surrounding us due to their various inherent features such as light weight, functional and structural flexibility, low cost and ease of manufacturing - to name a few. With the ever increasing demand for structurally and functionally graded materials, it is imperative to find avenues for creating polymeric composites with better properties than the virgin polymer. On the other hand, industrial lime sludge is a hazardous waste causing pollution to the environment and there is an urgent need to find an alternative solution for reusing this waste which is commercially and structurally viable. This means that the alternative process needs to be cheaper so that it would be profitable to employ this process industrially. Additionally, wherever this process is applied, the end product would have to be better than its existing counterpart in terms of product properties.

Based on the literature survey conducted above, the following gaps in knowledge is observed:

- Although researchers have proposed alternative utilization of industrial lime sludge waste in various areas, any research regarding re-utilization of industrial lime sludge waste as a filler in polymeric composites have not been conducted yet. Needless to say, the feasibility and commercial viability of lime sludge filled polymeric composites in terms of mechanical and thermal properties have not been discussed yet.
- Numerous investigations on the use of coupling agents or compatibilizers in enhancing the interfacial adhesion at filler-matrix boundary in polymeric composites have been conducted over the years. However, the effect of stearic acid or maleic anhydride grafted polymers as compatibilizers in lime sludge filled composites have not been examined yet.
- It is also not known whether and how lime sludge would effect as a filler in hybrid composites. It has been observed through literature that particulate fillers add a variety of positive dimensions to the properties of a hybrid composites.
- Although numerous investigations have been conducted in order to model composite properties as a function of filler loading, no such models are presented for lime sludge waste filled polymeric composites.

Most importantly, successful development of industrial lime sludge waste filled polyethylene composite will not only open another door towards utilization of these potentially hazardous industrial wastes, but also result in production of a commercially viable low cost polyethylene composite. Lime sludge based polymeric composite is a novel idea which would focus on integrating two advantages - (i) the scientific value - utilization of industrial lime sludge waste thereby decreasing pollution, and (ii) commercial value - reducing the manufacturing cost of polymeric composites and increasing the viability of the industrial waste.

## 2.8 Aim of the thesis

Based on the knowledge gaps, the main aim of the thesis is to study the feasibility of using industrial lime sludge waste in polymeric composites which would provide low cost structural materials with superior properties in addition to mitigating the problem of environmental pollution.

#### 2.8.1 Hypothesis

The hypothesis of this research is:

"Industrial lime sludge waste used as filler in polymeric composites would produce composites with comparable properties with that of pure polymers; thus, proving it to be an effective reinforcing filler."

#### 2.8.2 Objectives

Based on the hypothesis, the main objectives of this thesis are:

- To characterize polymeric composites filled with industrial lime sludge waste having comparable mechanical, thermal and morphological properties to its existing counterparts.
- To evaluate the effect of using stearic acid as a surface modifier on lime sludge filled polymeric composites and compare their mechanical, thermal and morphological properties with those of raw lime sludge filled composites.
- To develop various lime sludge filled hybrid composites (with different matrices and along with other reinforcing agents such as coir fibres) and evaluate their mechanical properties in order to assess the versatility of lime sludge as a reinforcing filler in different types of composites.
- To develop a regression based consolidated model which would provide predictive properties of composites as a function of lime sludge weight fraction in them.