# Abstract

Nowadays, bio-based polymers such as poly (lactic acid) (PLA) and chitosan (CH) have attracted much attention because of their environment friendliness and biodegradability. Poly (lactic acid) has proved to be the most promising biodegradable polymer which has been consumed in wide variety of applications. Due to its desirable properties such as transparency, bio-safety and compostability, it has been used in promising applications such as consumer goods, fibers, biomedicines and packaging. However, PLA has some limitations such as lower barrier properties, glass transition temperature, melting temperature and short degradation rate as compare to conventional polymers such as polyethylene terephthalate and polystyrene. Hence, it cannot be used efficiently in food packaging applications. Such limitations of PLA can be eliminated by mixing it with other biodegradable polymer which should have film forming ability. Literature survey revealed that chitosan, a biodegradable polymer, has good film forming ability with excellent barrier properties. Chitosan has been extracted from the exoskeleton of various natural resources such as crabs, lobsters, shrimps, insects etc. However, as per the literature survey, it has not been extracted from Muga silkworms till date. It is noteworthy to mention that chitosan is not compatible with PLA due to its hydrophilic nature and cannot be utilized in packaging and other applications due to non-melting behaviour. The incompatibility behaviour of PLA with chitosan has been studied by **Suyatma et al., 2004**. They have fabricated PLA/chitosan blend to check the improvement in water vapor transmission rate as well as mechanical property. The results showed an improvement in water vapor transmission rate whereas, elastic modulus and tensile strength were decreased due to agglomeration of filler. Hence, researchers have found many ways such as blending, mixing and grafting to avoid such limitations. As per the available literature, such techniques were conducted by using various initiators, catalysts, additives and solvents in order to improve the functionalization, productivity and transformation of its nature from hydrophilic to hydrophobic. When the functionalized low/high molecular weight copolymer reacts with the matrix polymer, it may result the enhancement in physico-chemical and other properties. Various polymer film/sheet formation techniques such as solution casting, layer by layer approach, melt blending etc. have been used and the resultant polymers have shown improvements upto a good extent in their barrier and other properties. It is noteworthy to mention that the catalyst or initiators used in functionalization are toxic in nature, which cannot be permitted to use in food packaging applications. Further, some techniques such as solution casting and layer by layer approach are not industrially viable. It was also inferred that very less efforts have been made to synthesize a bio-based chitosan grafted copolymer which when blends with PLA, it disperses uniformly in the form of spherical aggregates in hydrophobic PLA matrix. The fabricated bionanocomposite films may sustain better barrier properties as compare to that of PLA, which are also comparable to that of conventional plastics used for food packaging applications. Hence, it was observed that no work has been published on: (a) extraction of chitin and chitosan from Muga silkworms (MS) (*Antheraea assamensis*), (b) synthesis of modified hydrophobic chitosan grafted copolymer using lactic acid, (c) use of chitosan based copolymer in PLA for food packaging applications using solution casting as well as melt extrusion techniques.

As a result, the present doctoral work is mainly focused on food packaging application. In this research work, chitin and chitosan are extracted upto ~8 and ~7 wt% respectively from Muga silkworms and well characterized. Degree of deacetylation is found in the range of 77-97%. Further, chitosan is used as a filler in PLA matrix to fabricate poly (lactic acid)/chitosan based biocomposite films for food packaging application. However, the semicrystalline biocomposite films are not found suitable for same application due to reduction in mechanical strength and increment in water vapour transmission rate due to agglomeration. Moreover, chitosan has been chosen for functionalization with lactic acid due to have reactive amine group. Polycondensation reaction has been used for grafting of lactic acid oligomer on chitosan backbone with the help of conventional heating and microwave heating methods. As a result, it is found that lactic acid oligomer binds selectively at amine group of chitosan, which is also proved with the help of various analyses such as FTIR and NMR. Further, the formulated chitosan copolymer is used as a filler in PLA matrix for the fabrication of PLA/CH-*g*-OLLA bionanocomposite films with the help of various techniques such as solution casting and melt extrusion. The results shows that the glass transition temperature has been reduced more than 18 °C as compare to that of pristine PLA. However, the mechanical properties have been found comparable with PLA except its elongation, which has been improved significantly. The oxygen gas permeability of pristine PLA has been reduced upto ~90.4% with the increment of 5 wt% in filler loading. Further, non-isothermal degradation and isothermal crystallization kinetics of pristine PLA and its bionanocomposite films have been performed in order to check their rate of degradation and crystal growth respectively. Finally, it has observed that the fabricated PLA/CH-*g*-OLLA bionanocomposite films can be one of the best alternatives in food packaging application.