STUDIES ON HDPE / OCTAMETHYL POSS BASED NANOCOMPOSITE FILAMENTS

by

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Dedicated to

My parents, my wife Swapna and my daughter Urvi

CERTIFICATE

This is to certify that the thesis entitled "Studies on HDPE / Octamethyl POSS based Nanocomposite Filaments" being submitted by **Mr. Bhupendra Singh Butola**, to the Indian Institute of Technology, Delhi, for the award of the degree of **Doctor of Philosophy** in the Department of Textile Technology, is a record of bonafide research work carried out by him. Mr. Butola has worked under my guidance and supervision and fulfilled all the requirements for the submission of the thesis.

The results contained in this thesis have not been submitted, in part or full, to any other University or Institute for the award of any degree or diploma.

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ABSTRACT

The area of polymeric nanocomposites has generated a lot of research interest in the last few years, because of the immense potential it offers for the development of materials with unique combinations of properties not achievable with conventional polymer blends and composites.

This thesis is an attempt to explore the development of polymeric nanocomposites based on a novel class of hybrid nanostructured filler, Polyhedral Oligomeric Silsesquioxane (POSS). The system chosen for the study is the simple 'octamethyl POSS', a molecular silica as the nanofiller and HDPE as the polymeric matrix. The HDPE-POSS nanocomposites were prepared by melt compounding on a twin-screw extruder. The nanocomposites were spun into filament form by melt extrusion on a single screw extruder, followed by a two stage drawing process. POSS concentration varied in the range 0.1 to 10 wt %. The morphology of the nanocomposites is investigated using X-ray diffraction, Differential Scanning Calorimetry, AFM, TEM and SEM characterization techniques. The role of POSS in modifying the processing characteristics of HDPE is studied using rheological measurements. The HDPE-POSS nanocomposites have also been evaluated for their mechanical, dynamic mechanical, thermal and UV resistance properties in the filament form.

The incorporation of POSS does not affect the crystallinity or the crystal structure of HDPE upto a loading of 5 wt % POSS, which show a higher crystallinity level of ~75 %. At 10 wt % POSS content however, there is both a drop in the crystallinity level (to 71 %) and change in the crystal structure of HDPE as revealed in DSC and X-ray studies. Presence of POSS changes the morphology of HDPE by changing its crystallization behaviour both in isothermal as well as non-isothermal regimes. While

under isothermal crystallization conditions, the rates of crystallization increase, the crystallization mechanism is also modified in presence of POSS.

The POSS seems to be dispersed in HDPE at molecular/nanolevel with some POSS existing as nanocrystals of size 25 nm upto a POSS concentration of 5 wt %, while at higher concentration of 10 wt % starts agglomerating, as revealed in microscopic studies i.e., AFM, TEM and SEM and also supported by x-ray diffraction studies.

At comparatively very low loadings (0.25-0.5 wt %), POSS actually gives a lubricating effect and facilitates the drawing of filaments, which results in higher tensile strength and modulus. With increase in POSS concentration beyond 1 wt %, POSS existing as nanocrystals/aggregates starts hindering the orientation of HDPE chains leading to a gradual fall in tensile strength and modulus.

The lubricating effect of POSS dispersed at molecular/nanolevel also manifests itself into a drop in complex viscosity for nanocomposites at low POSS contents (0.25-0.5 wt %). Increasing POSS concentration further causes hindrance in chain movement resulting in a steady increase in the viscosity. At higher concentrations (> 5 wt %) POSS nanocrystals form a loose 3-D network, which exhibits a solid like behaviour due to physical gelation.

Due to their highly crystalline and oriented nature, β-transitions are not exhibited by HDPE or HDPE-POSS nanocomposites. However, the presence of POSS shifts the α-transitions of HDPE to a higher temperature. This is attributed to the restrictions on the HDPE chain segments in the intercrystalline regions due to the presence of POSS macromers on crystallite boundaries.

Incorporation of POSS also modifies the thermal degradation behaviour of HDPE and broadens the temperature range of thermal degradation. The thermal behaviour changes from one step thermal degradation for neat HDPE to a two step thermal degradation for HDPE-POSS nanocomposites.

The HDPE-POSS nanocomposites exhibit better UV resistance than neat HDPE filaments, which may be attributed to the scattering/reflective action of POSS. However, the UV stability imparted by this particular variant of POSS is not comparable to the commercially available UV stabilizers like HALS, carbon black etc.

CONTENTS