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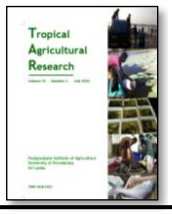

Microcrystalline Cellulose as Reinforcing Agents for Polypropylene Composites

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ABSTRACT

Microcrystalline cellulose (MCC) in its micrometric dimensions has gained a surge of interest in polymer reinforcement. This paper examines empirically the properties of MCC reinforced polypropylene (PP) composites varying the MCC loading from 0-5%. Mechanical, thermal and physical properties were characterized using universal testing machines, shore D hardness tester, TGA, FTIR and XRD machines. The experimental results highlighted that the reinforcement of PP matrix with MCC, improved the properties of the composite gradually as the MCC loading increased. Tensile, hardness and impact properties were improved by 5%, 8% and 51%, respectively for 5% MCC reinforced PP composite. Therefore, these notable results exhibit the potential of reinforcement of PP matrix with MCC.

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INTRODUCTION

There has been extensive work done throughout the industry in reinforcing polymer composites with fibers achieving a unique combination of both phases with a great versatility, tailoring better mechanical properties as well as higher performance. Recently emphasis has shifted from glass or carbon fiber reinforcement to natural fiber reinforced composites to abate the dependence of petroleum based products, focusing on more environmentally friendly low carbon foot print products (Samarasekara *et al.,* 2012a; Samarasekara *et al.,* 2014a; Samarasekara and Jayasuriya, 2014b; Samarasekara *et al*., 2015). Thus, many industries including automotive and aerospace industries have engrossed themselves in using more natural fiber reinforced polymer composites with the advantages of light weight, low density, non-toxicity, non-corrosiveness together with positive impacts on environment (Begum and Islam 2013; Kumar *et al*., 2017).

A variety of natural fibers have been used to supplement synthetic fibers including kenaf, jute, hemp, sisal and cellulose with their abundant availability, low cost, end-of-life degradability, high toughness and low energy usage in production (Joshi *et al*., 2004; Samarasekara *et al*., 2012b; Samarasekara *et al*., 2012c; Samarasekara *et al*., 2014; Liu *et al*., 2017). Being the most available natural polymer on earth cellulose has become one of the major candidates in fabricating polymeric substances. Many research communities have been making efforts on upgrading the properties of cellulose using different isolation methods initiating to diverse dimensions in micrometric and nanometric range and aspect ratios (Lu *et al*., 2014).

Sri Lanka has a high potential for cultivation and processing of a variety of crops leading to a considerable percentage of biodegradable agricultural wastes giving no commercial use but environmental pollution. From these superfluous agricultural wastes, cellulose can be extracted easily as a value-added product with variety of advantages. Microcrystalline cellulose (MCC) which can be isolated from native cellulose has attracted enormous interest in producing micro-composites. The interest arose from the extraordinary capacity of microscale reinforcement with a high degree of ordered crystalline regions, high surface area for interfacial bonding with polymer matrix as well as high mechanical strength and stiffness prior to cellulose (Yue, 2011; Sofla *et al.,* 2016). MCC has lateral dimensions smaller than the wavelength of visible light making MCC free of light scattering. Thus MCC is ideal in fabricating transparent polymeric matrices (Zampaloni *et al*., 2007).

Polypropylene (PP) is one of the most versatile and widely used thermoplastic polyolefin used in many industries due to its low production cost, recyclability, transparency, ability to blend easily and reinforce and low density (Sain *et al*., 2016; Amir *et al*., 2017). However, due to its moderate mechanical properties the durability of the final products has become low. Therefore, to compete with engineering polymers, PP chain reinforcement with natural fibers were introduced, leading to better mechanical, thermal and physical properties (Nanayakkara *et al*., 2017; Rajapaksha *et al*., 2017; Kahawita *et al*., 2018).

This project mainly focused on fabrication of MCC in PP matrix to upgrade the thermal, mechanical and physical properties of the final ecologically innocuous, green MCC-PP composite.

MATERIALS AND METHODS

Materials

Homopolymer Polypropylene (TASNEE PP H4260M) and white crystalline MCC were used as received from The National Petrochemical Industrialization Marketing Cooperation, Saudi Arabia and Sisco Research Lab Pvt. Ltd., India, respectively.

Fabrication of composites

Homopolymer PP was compounded with different loadings (0, 1, 2, 3, 4 and 5%) of MCC to study the property variation with MCC loading. In an internal mixer (MX300-TQ) PP was added and mixed for 5 minutes at 180 ˚C

at 65 rpm with Banbury type rotors. Then appropriate amounts of MCC were added and compounded with PP for another 8 minutes at 180 ˚C. Compounded samples were compression moulded at 180 ˚C for 5 minutes at 10 MPa pressure and demoulded after the cooling process. Five samples were prepared from each composition.

RESULTS AND DISCUSSION

FTIR Analysis

Figure 1 shows the FTIR analysis (Thermo Scientific, USA, Nicolet IS10) of pure MCC, neat PP and PP-MCC 5% composite. Pure MCC contained a broad OH stretching band at 3320 cm^{-1}, H-O-H bending of absorbed water at 1644 cm^{-1} and C-O stretching vibration of cellulose backbone at 1025 cm^{-1}. FTIR spectroscopy of MCC also contained characteristic bands at 1363, 1314 and 1201 cm^{-1} due to OH in plane bending (Akhtar *et al*., 2016; Wei *et al*., 2017). C-O-C asymmetric stretching, C-O stretching at C6, ring stretching in plane, CH_2 symmetric bending, and CH stretching in methyl and methylene group at 897, 1059, 1111, 1430 and 2902 cm^{-1}, respectively suggesting that the MCC comprised of cellulose I structure. Moreover, OH stretching band at 3320 cm^{-1} and OH out of plane bending at 700 cm^{-1} confirmed that MCC sample contained a higher percentage of cellulose I-β (Lu and Hsieh, 2010).

Four characteristic bands of neat PP were also present in PP-MCC composite as well. CH_3 asymmetric and symmetric stretching vibrations, CH_2 asymmetric and symmetric stretching vibrations were obtained at 2950, 2880, 2915 and 2845 cm^{-1} respectively (Morent *et al*., 2008). Bands attributed at 998 and 844 cm^{-1} confirmed the most stable confirmation of 3_1 helix structure in isotactic PP structure (Andreassen, 1999).

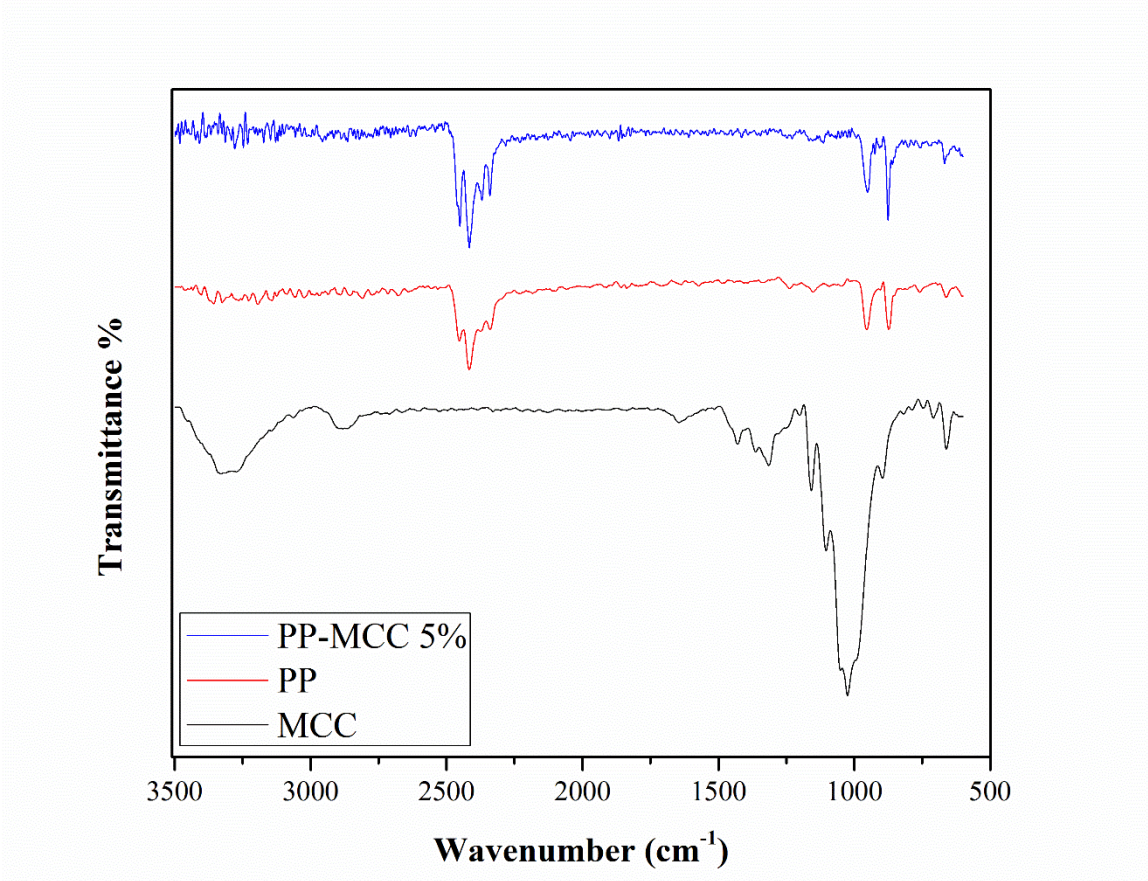

Figure 1. FTIR spectra of MCC, PP and PP-MCC 5% composite

XRD Analysis

XRD (Rigaku Ultima IV, USA) diffractogram of pure MCC showed a characteristic pattern of peaks at 14.9, 16.4, 22.5 and 34.5˚ attributed to (101), (10$\bar{1}$), (002) and (040) diffraction planes of cellulose I respectively, as shown in Figure 2 (Lu and Hsieh, 2010). Occurrence of a more intense sharper peak for (002) suggested the higher degree of crystallinity as well as the higher percentage of cellulose I-β structure (Poletto *et al*., 2013; Lee *et al.*, 2014). Seven characteristic diffraction planes were introduced for both neat and PP composite at 14.3, 16.3, 18.6, 21.5, 22.1, 25.6 and 28.8 ˚ for (110), (300), (130), (131+041), (060+150) and (220), respectively *(*Achaby *et al*., 2012). Absence of (300) plane at 16.3˚ in composite diffractogram suggested that the incorporation of MCC into PP matrix induces changes in crystal structure of PP (Wang and Sheng, 2005).

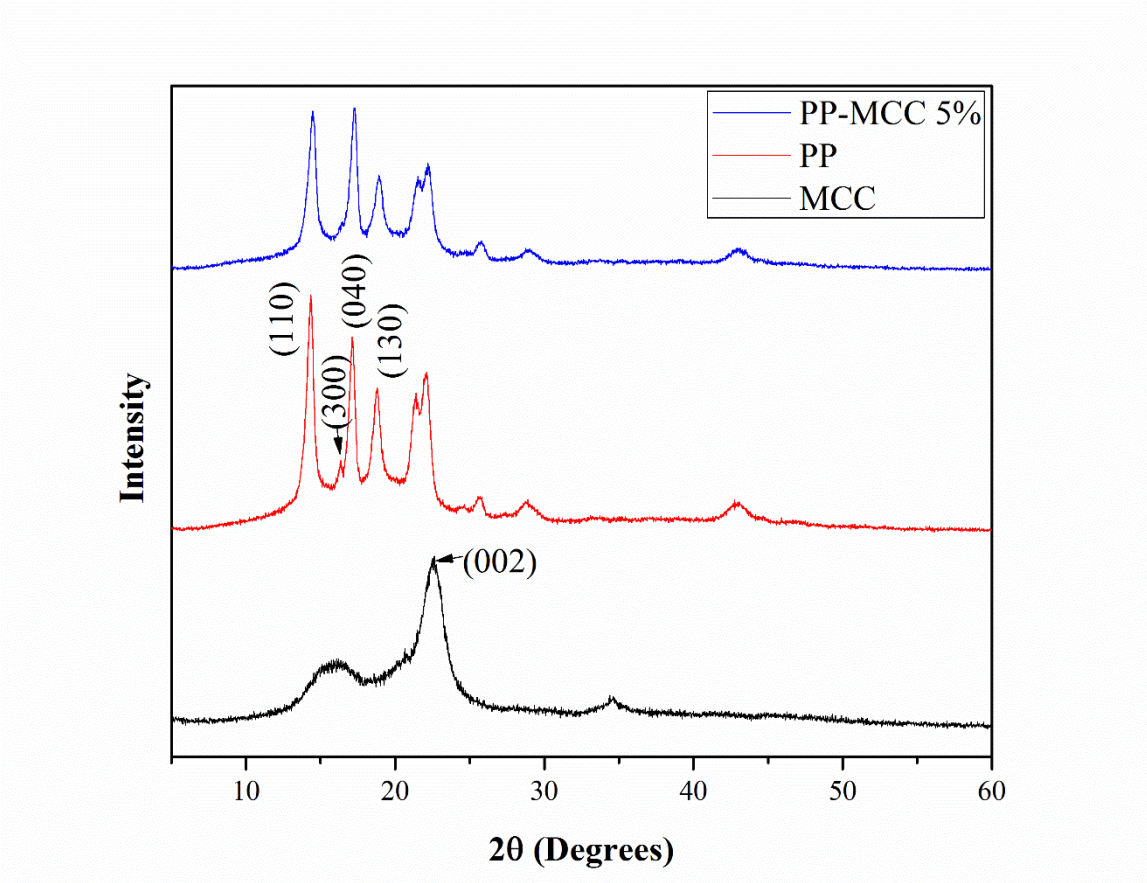

Figure 2. X-ray Diffractogram of MCC, PP and PP-MCC 5% composite

TGA Analysis

Thermal stability variation of neat PP with the incorporation of MCC in composites were analyzed using thermogravimetric graphs using SDT Q600, USA (Quick Start software) (Figure 3). Single step degradation for neat PP showed at 434 ˚C. As PP is reinforced with 5% MCC degradation shifted to a higher a temperature of 455 ˚C implying that MCC reinforcement improved the thermal stability of neat PP matrix (Canetti *et al*., 2006; Bikiaris *et al*., 2008).

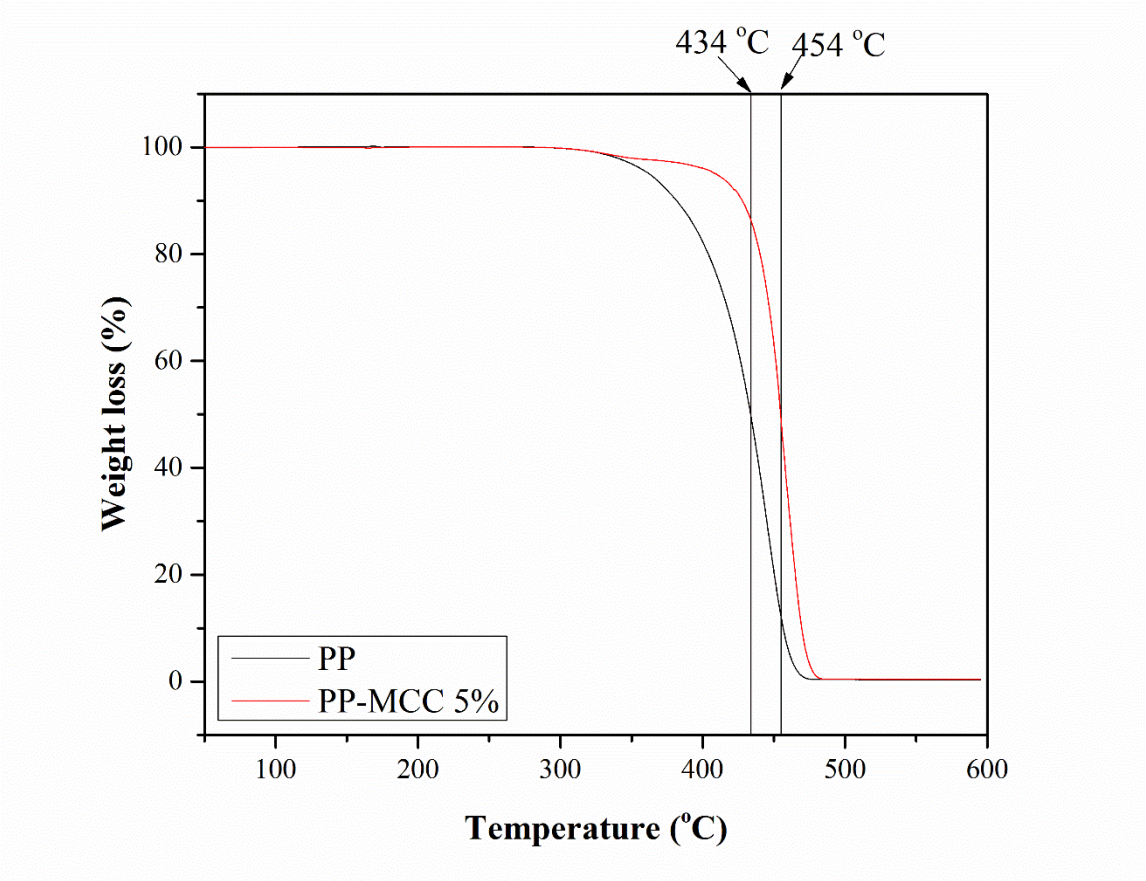

Figure 3. TGA curves of PP and PP-MCC 5% composite

Mechanical Properties

Tensile testing

Universal tensile testing machine (Hounsfield) was used to observe the tensile strength of the composites. Average tensile strength of neat PP was marked at 27.24 MPa which was gradually increased with the addition of MCC (Figure 4). Maximum tensile strength, 28.65 MPa was obtained for 5% MCC incorporation in PP composites. These results implied that as PP matrix reinforced with stiff MCC, the resistance to break at tension increased as well.

Impact testing

Izod Impact tester (HUNG TA Instrument Co. Ltd, Taiwan, HT-8041B) was used to characterize the impact strength. Impact strength variation (Figure 5) in PP composites showed that more energy can be absorbed prior to the fracture by the PP composite as the MCC loading increased. Thus, impact strength of neat PP was improved from 6.85 kJm^{-2} to 10.40 kJm^{-2} with the fabrication of 5% MCC in PP composite.

Hardness testing

Hardness (Shore D durometer, Teclock, GS-702N) of MCC reinforced PP composites (Figure 6) showed higher values than neat PP (61.5). Gradual increment of shore D hardness in MCC-PP composites maximized to 66.5 with the addition of 5% MCC in PP due to its higher reinforcement and decrement in PP chain flexibility.

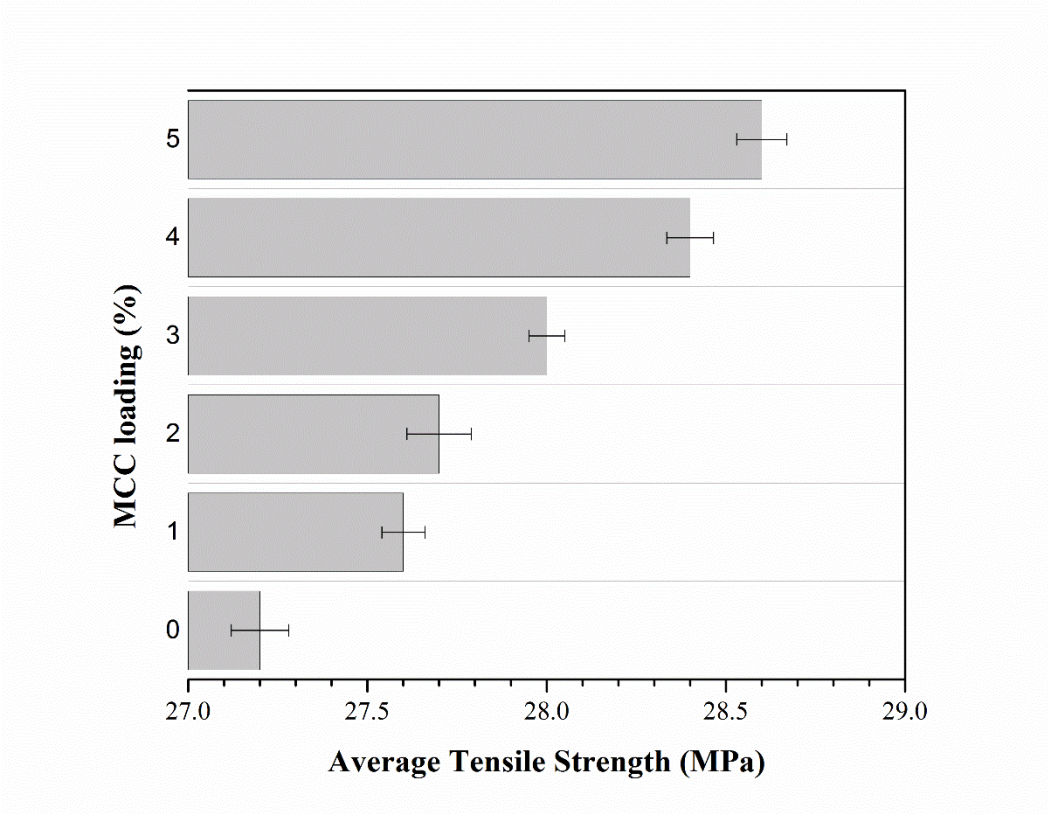

Figure 4. Average Tensile strength variation with respect to MCC loading

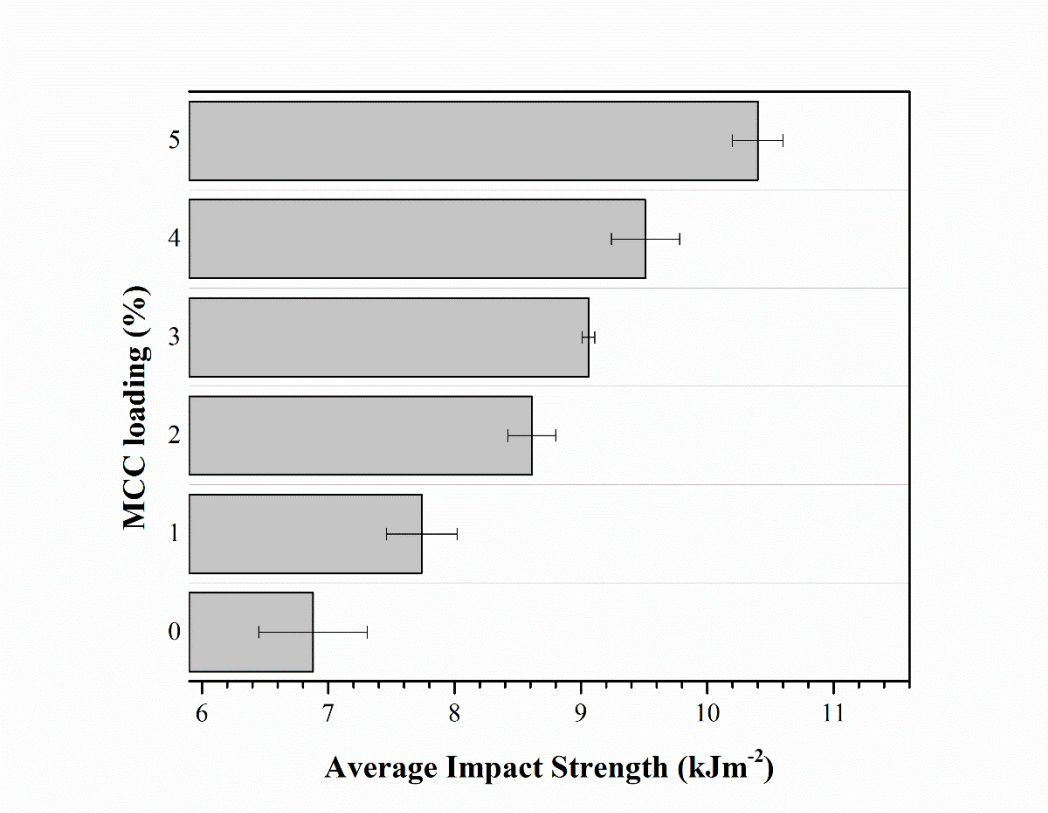

Figure 5. Average Impact strength variation with respect to MCC loading

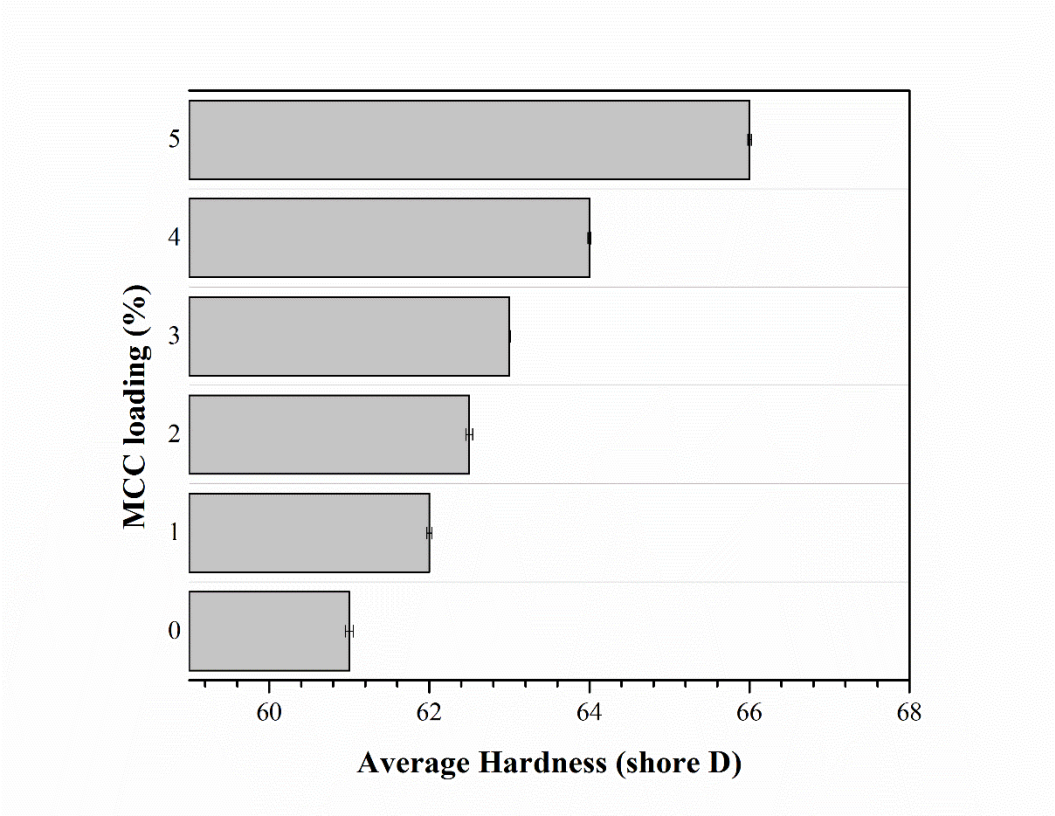

Figure 6. Average Hardness variation with respect to MCC loading

CONCLUSIONS

The experimental results showed a greater improvement in thermal and mechanical properties with the fabrication of MCC in PP matrix. Tensile, hardness and impact properties improved by 5%, 8% and 51%, respectively in 5% MCC reinforced PP composites than non-reinforced PP. The results confirmed MCC comprised of a higher percentage of cellulose I-β structure and isotacticity of PP matrix. This research project prioritized importance of microcrystalline cellulose to reinforce PP matrix to prepare environmentally compatible bio composites.

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