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# Thermal conductivity and mechanical properties of high density polyethylene composites filled with silicon carbide whiskers modified by cross-linked poly (vinyl alcohol)

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ABSTRACT

A thin layer of poly (vinyl alcohol) (PVA) was coated on the surface of silicon carbide whiskers (SCWs) and crosslinked by glutaraldehyde, and then these modified whiskers (mSCWs) were incorporated into high density polyethylene (HDPE) to prepare HDPE/mSCW composites with a high thermal conductivity. The thermal conductivity, mechanical properties, heat resistance, thermal stability and morphology of HDPE/mSCW and HDPE/SCW composites were characterized and compared. The results reveal that the thermal conductivity of both HDPE/SCW and HDPE/mSCW composites increases with the increase of filler loading, and reaches a maximum of 1.48 and 1.69 W/(m K) at 40 wt% filler loading, which is 251.20% and 300.75% higher than that of HDPE, respectively. Significantly, HDPE/mSCW composites have a higher thermal conductivity than their HDPE/SCW counterparts with the same filler loading. In addition, the heat resistance, Young's modulus and yield strength of both HDPE/SCW and HDPE/mSCW composites are also improved compared with that of HDPE. mSCW can be homogenously dispersed in the HDPE matrix, which contributes to the formation of thermally conductive networks by the inter-connection of mSCWs.



## 1. Introduction

In recent years, there is an increasing demand for thermally conductive materials in heat exchange and transfer, electrical and electronic fields [1,2]. However, the application of traditional thermally conductive materials such as metals are seriously limited due to their undesirable properties, such as heavy weight, susceptibility to corrosion, difficulty in processing, and high manufacturing cost [3]. Most polymers have the advantages of light weight, high corrosion resistance and ease of processing, thus making them promising candidates for thermal conductivity. A potential problem that can seriously compromise the application of polymers is their low thermal conductivity, and thus there is a practical need for the preparation of polymers with a high thermal conductivity [4,5]. This can be achieved by two approaches. The first one is to synthesize polymers with a π-π conjugated structure or extremely high crystallinity, as the former allows for the motion of free electrons and the latter allows for the high speed of phonon with less defects and a better intrinsic order [2]. A second approach is to incorporate fillers with high thermal conductivity into the polymer matrix. A wide variety of fillers have been used for this purpose, such as metals [6,7], carbon materials [5,8], ceramic materials [4,9], silicon carbide (SiC) [10], silicon nitride [11–13] and boron nitride [14]. This is perhaps the most convenient and effective way to obtain desired thermally conductive materials.

It is suggested that increasing the filler content contributes to the formation of thermally conductive pathway or network and thus the improvement of the thermal conductivity [2]. It is also noted that such a network can be more easily constructed with the use of one-dimensional or two-dimensional fillers. One dimensional fillers such as nanowires, fibers, rods, whiskers and nanotubes have a high aspect ratio and the heat energy can be transferred in the longitudinal direction. Two dimensional fillers such as BN platelet, graphene and graphite tend to align parallel to each other, and thus they can provide a high degree of contact in the parallel direction. Xiao et. al. [15] introduced carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) into the poly (vinylidene fluoride) (PVDF) matrix simultaneously to prepare ternary composites, and the rheometer analysis showed that the CNTs and GNPs formed a three-dimensional hybrid network structure.

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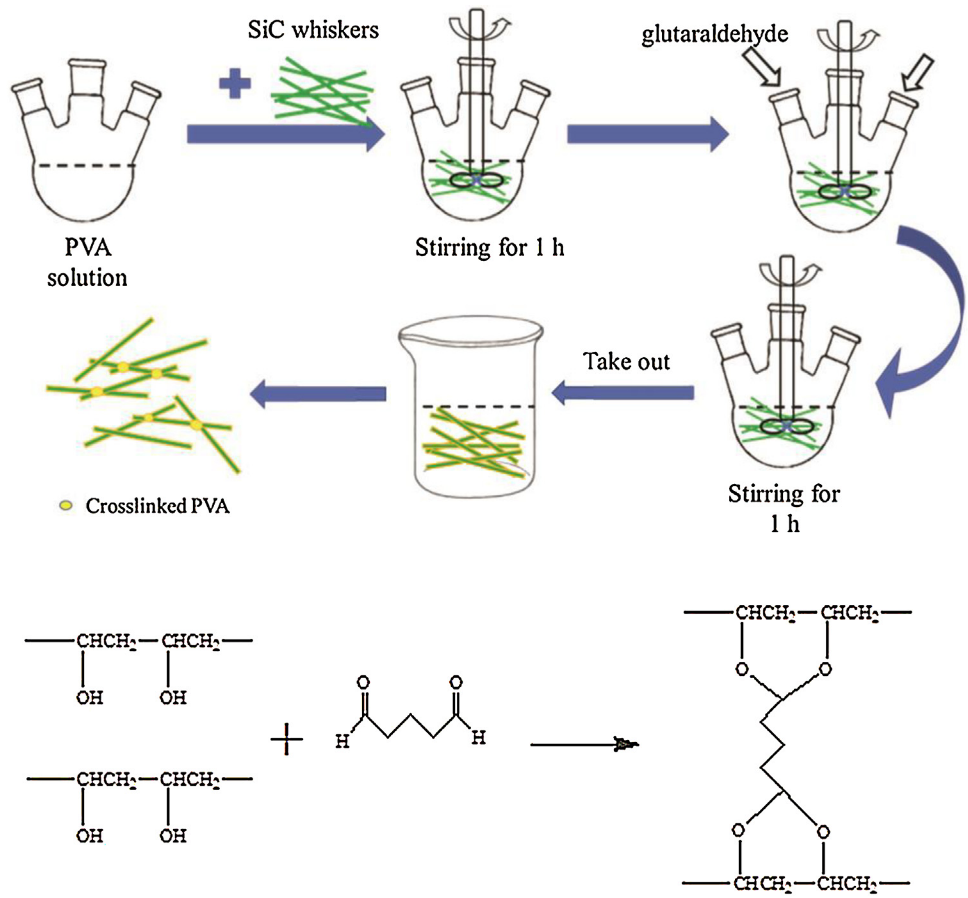


**Fig 1.** Preparation process of mSCWs and the cross-linking reaction between glutaraldehyde and PVA.

Chemical connection is a possible way to form the thermally conductive network. Gu et. al. [16] added thermally conductive SiC particles grafted with polyhedral oligomeric silsesquioxane into the ultrahigh molecular weight polyethylene to fabricate composites. The $-NH_2$- groups on the polyhedral oligomeric silsesquioxane could connect several SiC particles, resulting in an increase in the homogeneous dispersion and interfacial compatibility. The reaction between polyhedral oligomeric silsesquioxane and particles contributes to the formation of thermally conductive networks. Physical combination is also helpful for the formation of thermally conductive networks. Different polymers in the matrix can have different affinities for the fillers, and those with high affinities can act as adhesives and centralizes towards the fillers. Li et. al. [17] investigated the effect of relative interfacial affinity on the formation of glass fiber-polyamide 6 (GF-PA6) networks in different polymer matrices. Double percolation was often used to form thermally conductive pathway in heterogeneous system. Two immiscible polymers constitute the matrix, at least one of which is continuous and has better compatibility with the fillers. These fillers would be selectively and preferentially localized in the continuous polymer phase, and thus the network would be formed in the continuous phase. This method makes it possible use less fillers to achieve a high thermal conductivity [18–22]. Cao et. al. [22] investigated the effect of selective localization of SiC and polystyrene-coated SiC (p-SiC) on the thermal conductivity of PS/PVDF, and their results showed that SiC enabled the PS/PVDF (70/30) composites to have a higher thermal conductivity than p-SiC.

SiC whisker has a high thermal conductivity of about 80 W/(m K), and its one-dimensional shape makes it more convenient to contact with each other. The addition of SiC whiskers would not compromise the tensile properties of the polymer. However, the effect of SiC whiskers on the thermal conductivity of high density polyethylene (HDPE) composites remains poorly understood [10]. Previous studies have focused on the surface modification of SiC to decrease the interfacial thermal resistance and improve the thermal conductivity of the composites. However, there is a lack of effective methods to construct a thermally conductive network or pathway with SiC whiskers. Kim et. al [23] reported that more units linked together were easier to contact and helped to improve the thermal conductivity of the composites. Inspired by Yuan's method [24], SiC whiskers are treated by cross-linked PVA to improve their dispersion in the HDPE matrix in this study, and then these modified SCWs (mSCWs) are added into HDPE to prepare the composites with high thermal conductivity. The thermal conductivity, heat resistance, tensile properties and thermal stability of HDPE/mSCW and HDPE/SCW composites are characterized and compared.

## 2. Experimental

### 2.1. Materials

HDPE (DMDA-8920, $\rho = 0.948\ g/cm^3$) was purchased from Sinopec Dushanzi Co., Ltd. (Xinjiang, China); SiC whisker ($D \leq 2.5\ \mu m$, $L/D \geq 20$) was purchased from Hongwu Nano Technology Co., Ltd. (Jiangsu, China); polyvinyl alcohol (PVA; 1750) was purchased from Shanghai Lingfeng Chemical Regents Co., Ltd. (Shanghai, China); and 25 wt% glutaraldehyde solution was purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China).