



Thermal conduction behaviors of chemically cross-linked high-density polyethylenes[☆]



Seunggun Yu^{a,b}, Cheolmin Park^b, Soon Man Hong^a, Chong Min Koo^{a,c,*}

^a Center for Materials Architecturing, Korea Institute of Science and Technology, Hwarangno 14-gil 5, Seongbuk-gu, Seoul 136-791, Republic of Korea

^b Department of Materials Science and Engineering, Yonsei University, 50 Yonsei-ro, Seodaemun-gu, Seoul 120-749, Republic of Korea

^c Nanomaterials Science and Engineering, University of Science and Technology, 176 Gajung-dong, 217 Gajungro, Yuseong-gu, Daejeon 305-350, Republic of Korea

ARTICLE INFO

Article history:

Received 28 October 2013

Received in revised form 7 March 2014

Accepted 19 March 2014

Available online 28 March 2014

Keywords:

High-density polyethylene

Crystallinity

Thermal conductivity

Crosslinking

ABSTRACT

The effect of chemical crosslinking on the thermal conductivity of high-density polyethylene (HDPE) was investigated. The crosslinking density of HDPE was controlled by the content of the crosslinking agent of dicumyl peroxide (DCP) in molten state. As the content of DCP increased, the gel content of HDPE increased, and crystal structure such as spherulite size and crystallite size, and crystallinity decreased, as network formation suppressed the crystallization of polymer molecules. The thermal conductivity of crosslinked HDPE was proportional to the crystallinity and reciprocal to the gel content.

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1. Introduction

Thermal management of materials has received a considerable amount of attention in various industrial fields of electronics, lighting, automobiles, aircraft and solar cells, as the failure of heat dissipation increases the temperature of devices and deteriorates their lifetime, reliability, and performance [1–5]. Among the materials used for thermal management, polymer and polymer composite materials have been considered as next-generation thermal conduction materials due to their light weight, easy processability and low cost [6,7].

However, in general, polymers have very low thermal conductivity (on the order of $0.1 \text{ W m}^{-1} \text{ K}^{-1}$) because they have numerous structural defects, such as those at the boundary between crystal domains, and an amorphous structure in a disorder state [8–10]. The thermal conduction of insulating polymer materials is carried out by phononic heat conduction through acoustic phonons, unlike that of metals, which is governed by electronic heat conduction through mobile electrons [11,12]. The acoustic phonon energy is

likely to propagate through the harmonic oscillation of the crystal solid. The structural defects of polymers give rise to phonon scattering, reducing the thermal conductivity [13,14].

High-density polyethylene (HDPE), one of the most popular general polymers, has thermal conductivity of $0.6 \text{ W m}^{-1} \text{ K}^{-1}$, which is the top-level value among polymer materials. The high thermal conductivity of HDPE basically stems from the high crystallinity, which can reach a maximum of 90% [15–19]. The high crystallinity also accompanies a high yield point, as well as high internal tearing stress of polyethylene. However, the use of HDPE as a thermal conduction material is limited due to its low melting temperature, below 140°C , and its poor dimension stability at high temperatures. Crosslinking reactions compensate for the disadvantages of HDPE. The three-dimensional network structure formed in HDPE via the crosslinking process is responsible for improved mechanical properties, such as the tensile strength and hardness and the dimension stability [20–22]. A crosslinking reaction of HDPE can be carried out in three ways: high-energy radiation crosslinking [23,24], a chemical crosslinking reaction by organic peroxide [25,26], and silane–water crosslinking [27,28]. Among them, a chemical crosslinking reaction using organic peroxide is widely used owing to its many advantages, such as an economic process, easy control of the peroxide decomposition rate and fewer by-products.

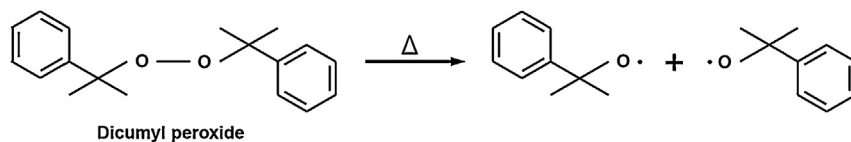
In this study, we demonstrate that the chemical crosslinking of HDPE using a peroxide crosslinking agent controls gel content,

[☆] Presented at The 10th Asia Thermophysical Properties Conference, Jeju, Korea, September 29–October 3, 2013.

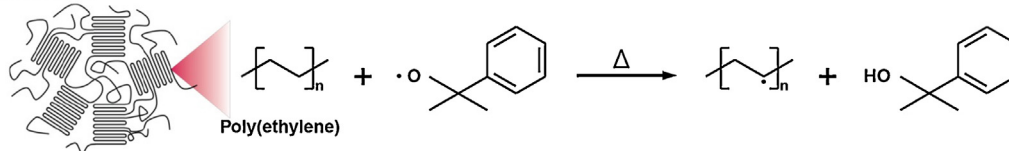
* Corresponding author at: Center for Materials Architecturing, Korea Institute of Science and Technology, Hwarangno 14-gil 5, Seongbuk-gu, Seoul 136-791, Republic of Korea. Tel.: +82 2958 6872; fax: +82 2958 5309.

E-mail addresses: koo@kist.re.kr, chongminkoo@gmail.com (C.M. Koo).

Thermal decomposition



Initiation



Crosslinking reaction

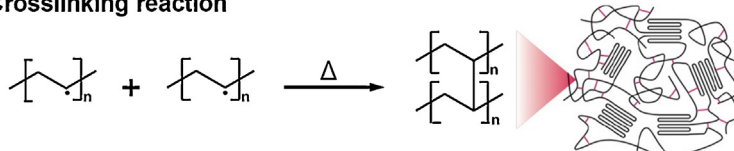


Fig. 1. Scheme of the crosslinking reaction of polyethylene molecules using a peroxide crosslinker.

crystal structure and crystallinity and thermal conduction behavior. The thermal conductivity was determined by crystal structure and crystallinity of the HDPE.

2. Experimental

2.1. Materials

HDPE (2600J) with a melt index of $20 \text{ g } 10^{-1} \text{ min}^{-1}$ at 190°C and density of 0.959 g cm^{-3} was supplied from Honam Petrochem in South Korea. Dicumyl peroxide (DCP, 98.0%), xylene (99.9%) was purchased from Sigma–Aldrich in the USA.

2.2. Preparation of the crosslinked HDPE

The HDPE was melted in an internal mixer (Haake polydrive mixer, Thermo Fisher Scientific Inc., Germany) at a speed of 100 rpm at 160°C for 5 min, after which DCP was added and mixed for 2 min at the same temperature. The DCP content varied from 0.5 to 3.0 wt.%. High-pressure molding was performed in a hot press (Auto series, Carver Inc., USA) at 180°C and 12 MPa for 5 min. The sheet samples were then cooled to room temperature at a cooling rate of $10^\circ\text{C min}^{-1}$.

2.3. Characterizations

The gel content of the crosslinked HDPEs was evaluated by Eq. (1).

$$F_{\text{gel}} = \frac{w}{w_0} \times 100 \quad (1)$$

where w and w_0 are the weight of the samples before and after solvent extraction, respectively. The extraction process was performed using a xylene medium at 110°C for 12 h with the reflux condition according to the ASTM D2765-11 standard. The thermal melting behaviors of the HDPEs were investigated by means of DSC (Differential scanning calorimetry, Q20, TA Instruments) under nitrogen atmosphere at a heating rate of $10^\circ\text{C min}^{-1}$. Synchrotron wide-angle X-ray scattering (WAXS) measurements at the beam line 4 C of the Pohang Light Source, South Korea, using an X-ray beam were performed at room temperature. The wavelength of the electron beam light source was $\lambda = 1.54 \text{ \AA}$. Crystal

morphology of the HDPE was investigated by polarized optical microscopy (DM2500P, Leica, Germany) under crossed polarizers, using thin HDPE films crystallized at 117°C on a glass slide. The thermal diffusivity of the HDPEs was measured using the laser flash method (LFA-447, Netzsch, Germany) based on an xenon flash lamp source at room temperature. Both sides of the semi-transparent HDPE samples were coated with Au and graphite to prevent light energy from passing through and to absorb reflected light energy without loss, respectively. The Au was coated using a sputtering machine (KIC-1A, COXEM, Korea) and the graphite was coated using a graphite spray (Graphite 33, CRC Industries, USA). The thermal diffusivity and specific heat capacity were determined using an LFA-447 device in comparison with pyroceramic references. The thermal conductivity was calculated from the equation of $k = T_d \times d \times C_p$, where T_d , d and C_p are the thermal diffusivity ($\text{mm}^2 \text{ s}^{-1}$), density (g cm^{-3}) and specific heat capacity ($\text{J kg}^{-1} \text{ K}^{-1}$), respectively. The bulk density d was calculated using the weight and volume of a specimen with dimensions of $10 \text{ mm} \times 10 \text{ mm} \times 2 \text{ mm}$ (Width \times Length \times Height). The average bulk density of five specimens was used to minimize errors.

3. Results and discussion

3.1. Crosslinking reaction

Fig. 1 illustrates the crosslinking process of polyethylene molecules using a DCP crosslinking agent in a melted state. Generally, the peroxide bond between oxygen atoms in a DCP molecule is decomposed and produces radical pairs at a high temperature. The cumyl peroxide radical faces the polyethylene molecule, and the radical is transferred to the hydrogen atom site in the polyethylene molecule according to the electronegativity difference. Finally, the polyethylene molecules with the radical are reacted with each other and form a crosslinking bond. The DCP used here had an activation energy of $34.3 \text{ kcal mol}^{-1}$ for radical decomposition and a 1 min half-life at 170°C [29,30]. HDPE and DCP were pre-mixed at 160°C for 2 min using an internal mixer and the mixture was then hot-press molded using a hot-press to complete the crosslinking reaction at 180°C and 12 MPa for 5 min.

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