Form 2

**Dissertation Abstract**

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| Dissertation title | Fabrication of Flexible Transparent Fluorinated Nanohybrids Including Organo-modified Nanoparticles and Its Spherulitic Observation (有機修飾ナノ粒子を含んだフレキシブル透明フッ素系ナノ複合体の創製とその球晶観察) | | |


Abstract

Partially fluorinated crystalline polymer nanohybrids were prepared by incorporation of different types of organo-modified nanoparticles into the polymer matrix via melt-compounding process. Poly[ethylene-*co*-(tetrafluoroethylene)] (ETFE) nanohybrid was fabricated by incorporation of organo-modified montmorillonite (organo-MMT) in the polymer matrix. A new technology for the production of transparent material, using a “crystalline” polymer, is proposed in this work. In addition, a heat-resistant transparent flexible plastic film with a high hydrophobic surface and a thermal decomposition temperature near 400 ℃ was created. Partially fluorinated crystalline polymer with switchboard-type lamellae results high transparency as a consequence of the formation of a high-density amorphous structure based on high-temperature drawing just below the melting point at 250 ℃. Melt-compounding with montmorillonite modified by the long-chain quaternary phosphonium with high coverage induces formation of a nanohybrid that retains transparency and also results in an increase in the thermal degradation temperature by over 50 ℃. Through this technology, which results in heat-resistance, transparency, and flexibility, the nano-micro-millimeter structures of solid-state polymers are hierarchically controlled, which enables the creation of new materials.

Poly[vinylidenefluoride-*co*-(tetrafluoroethylene)] (P(VDF-TeFE)) nanohybrid was fabricated by incorporation of organo-modified nanodiamond (organo-ND) in the polymer matrix. Here, also a new technology for the production of transparent material using a “crystalline” polymer, is proposed in this study. Further, transparent and flexible crystalline polymer nanohybrid film containing well-dispersed nanodiamond filler was fabricated. In this case, crystalline partially-fluorinated copolymer having switchboard-type lamellae indicates excellent

transparency based on the formation of “high-density amorphous region” by drawing at high-temperature just below the melting point at 110 ℃. Although the formation of nanohybrid materials composed of fluorinated-polymer/organo-modified nanocarbon is generally difficult, it confirmed the formation, via melt-compounding, using atomic force microscopy (AFM) and wide-angle X-ray diffraction (XRD). Even though the polymer matrix/nanodiamond hybrid has remarkable aggregation properties, a well-dispersed state was achieved because of improvement in wettability obtained through surface modification of filler. The resulting nanohybrid demonstrates transparency, increased thermal degradation temperature, and enhanced mechanical properties, which seem to be derived from the nucleation effect caused by the adsorption of the terminal polymer chain onto the organic modifier.

P(VDF-TeFE) exhibited clear spherulitic texture with negative birefringence. The number and growth rates of the spherulites decreased at high crystallization temperature than at low crystallization temperature. Nonetheless, overall larger spherulites were found at high crystallization temperature and the brightness of the spherulites increased very fast at low crystallization temperature, thereafter it seemed as diminution of birefringence. AFM was used to investigate the impact of organo modified nanodiamond(ND) on spherulitic textures, lamellar thickness, and thickness distribution of P(VDF-TeFE) copolymer. ETFE also confirmed spherulites structure and the boundaries could be clearly observed. By incorporation of the organo modified nanodiamond (ND) and organo-modified montmorillonite (MMT) in fluropolymer matrix, it was found that spherulitic texture was seriously disordered and their nanohybrids was found only to have poorly developed spherulite structure. Both of the nanohybrids samples show better crystallization temperature as compared to their neat copolymer samples. Furthermore, the incorporation of nanoparticles decreased the size of the spherulites. Since nanoparticles act as the nucleating agents, a large number of nucleus generated and spherulitic texture was seriously changed in the nanohybrids spherulitic texture.