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Non-isothermal crystallization kinetics and characterization of biodegradable poly(butylene succinate-co-neopentyl glycol succinate) copolyesters

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ABSTRACT

Both biodegradable aliphatic neat poly(butylene succinate) (PBS) and poly(butylene succinate-co-neopentyl glycol succinate) (P(BS-co-NPGS)) copolyesters with different 1,4-butanediol/neopentyl glycol ratios were synthesized through a two-step process of transesterification and polycondensation using stannous chloride and 4-Methylbenzenesulfonic acid as the co-catalysts. The structure, non-isothermal crystallization behavior, crystalline morphology and crystal structure of neat PBS and P(BS-co-NPGS) copolyesters were characterized by ^{1}H NMR, differential scanning calorimetry (DSC), polarized optical microscope (POM) and wide angle X-ray diffraction (WAXD), respectively. The Avrami equation modified by Jeziorny and Mo's method was employed to describe the non-isothermal crystallization kinetics of the neat PBS and its copolyesters. The modified Avrami equation could adequately describe the primary stage of non-isothermal crystallization kinetics of the neat PBS and its copolyesters. Mo's method provided a fairly satisfactory description of the non-isothermal crystallization of neat PBS and its copolyesters. Interestingly, the values of $1/t_{1/2}$, Z_c and F(T) obtained by the modified Avrami equation and Mo's method analysis indicated that the crystallization rate increased first and then decreased with an increase of NPGS content compared that of neat PBS, whereas the crystallization mechanism almost kept unchanged. The results of tensile testing showed that the ductility of PBS was largely improved by incorporating NPGS units. The elongation at break increased remarkably with increasing NPGS content. In particular, the sample with 20% NPGS content showed around 548% elongation at break.

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1. Introduction

In recent years, the synthetic biodegradable aliphatic polyesters derived from renewable resources have always been spotlighted with tremendous interest from the standpoint of conserving our petrochemical resources and overcoming the environmental issues associated with conventional plastics [1–3]. Because of their superior biodegradable properties compared to those of conventional polymers, biodegradable aliphatic polyesters are one of the most promising alternative substances for solving the serious environmental problems. As a synthetic polyester, poly(butylene succinate) (PBS), together with its copolymers and blends, has been attracting a considerable practical attention for its biodegradability, thermal properties, and acceptable mechanical properties [4]. Packaging, food containers and waste bags are some of its applications [5,6]. Furthermore, its raw materials, 1,4-butanediol and succinic acid, both can be available either from fossil fuel feedstock or from bio-based renewable resources [1–3,7]. For these advantages, their crystallization behaviors, crystal structure and morphology have been extensively investigated by using various techniques from the academic viewpoint as well as from the viewpoint of practical applications.

However, as a semi-crystal polyester, PBS suffers from some defects such as low degradation rate which mainly caused by its high relative crystallinity [8]. In order to improve such properties, up to now, a series of copolymers of PBS with other comonomers such as adipic acid, terephthalic acid, ethylene glycol, diethylene glycol and ε-caprolactone have been investigated [8–12]. These copolymers have offered opportunities in understanding the effect of comonomers on the structures and properties of the copolyesters, and developing novel polymer materials with excellent properties.

This paper will present the synthesis of aliphatic copolyester P(BS-co-NPGS)s as well as aliphatic PBS obtained by polycondensation reaction from succinic acid, 1, 4-butanediol and/or neopentyl glycol in the presence of the effective co-catalysts stannous chloride and 4-Methylbenzenesulfonic acid. Furthermore, the non-isothermal crystallization behavior is important since most polymers are practically processed under non-isothermal conditions, so systematic investigations on the effect of NPGS content on the non-isothermal crystallization kinetics of the P(BS-co-NPGS)s compared with that of neat PBS are presented in this paper, and the kinetics analysis of non-isothermal crystallization is performed with the Avrami equation

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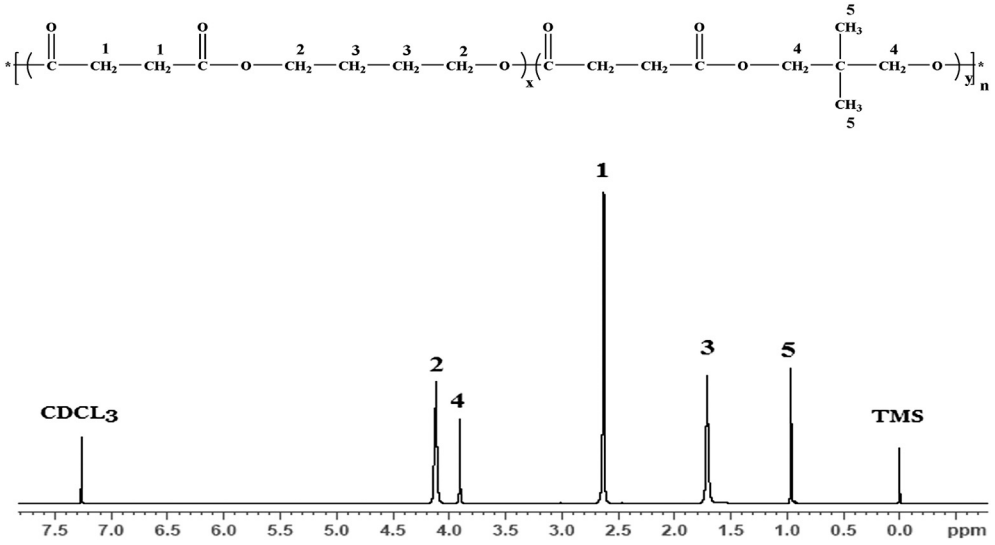

Fig. 1. ^{1}H NMR spectrum of P(BS-co-20 mol% NPGS).

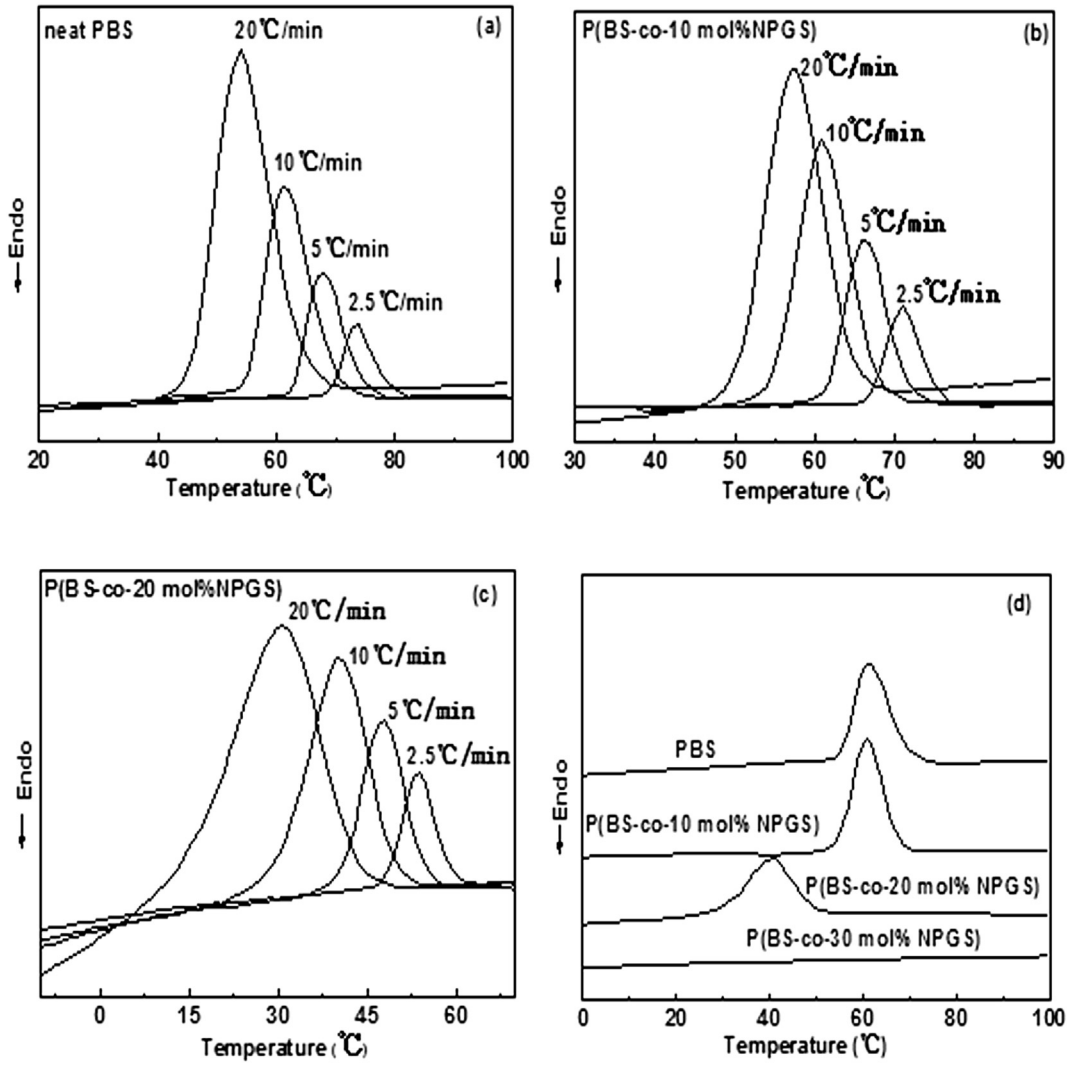

Fig. 2. Crystallization peaks on cooling for neat PBS (a), P(BS-co-10 mol% NPGS) (b) and P(BS-co-20 mol% NPGS) (c) at various rates. (d) Crystallization peaks of neat PBS and its copolyesters at cooling by 10 °C/min.

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