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Design of long-chain branched copolyesters and manufacture as well as physical properties of their extrusion films

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A R T I C L E I N F O

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A B S T R A C T

A long-chain branched copolyester (i.e. *p*-hydroxybenzoic acid (HBA)/2-hydroxy-6-naphthoic acid (HNA)/1,1,1-tris(4-hydroxyphenyl)ethane (THPE) copolyester) and its extrusion film with superior physical properties to commercial Ticona A950 and Vecstar CTZ have been manufactured by the design of appropriate prescriptions, T-Die extrusion, and thermal treatment of crystal transformation. In order to investigate the fabricating feasibility for flexible copper clad laminate (FCCL) of fourth/fifth generation long-term evolution (4G/5G LTE), we have also agglutinated lab-made copolyester extrusion film with the copper foil by hot compression. Experimental results manifest that lab-made copolyester extrusion film is a highly potential FCCL substrate of 4G/5G LTE because its thickness, dielectric constant (D_k), dielectric loss (D_f), hygroscopicity, yellowness index (YI), anti-static capability, flammability, melting temperature (T_m), coefficient of thermal expansion (CTE), and peel strength to copper foil are 50 μm, 2.90, 0.00140, 0.04%, 2.7, 6.5 × 10^9 Ω/□, UL-94 V0, 305 °C, 24.8 ppm/°C, and 29.9 lb/in, respectively.

1. Introduction

Copolyester is a well-known recycled engineering material with extraordinary physical performances (e.g. high thermal resistance [1], low hygroscopicity [2], high dimensional stability [3], low dielectric constant (D_k) as well as dielectric loss (D_f) [4,5], high processibility [6], good nonflammability [7], high chemical resistance [8], low coefficient of thermal expansion (CTE) [9], outstanding mechanical properties [10,11], etc.) and has been applied for industrial fibers [12], displays [13], flexible printed circuits (FPCs) [14], electrical switches [15], optoelectronic devices [16], and connectors for electronic products as well as automobiles [17]. In recent years, the flexible copper clad laminate (FCCL) of fourth/fifth generation long-term evolution (4G/5G LTE) has attracted much attention because the transmission speed of 4G/5G LTE is 10/100 times faster than 3G LTE, whose FCCL substrate is made of polyimide (PI) [18]. However, PI with high thermal stability is unqualified for FCCL substrate of 4G/5G LTE due to its high hygroscopicity, D_k, and D_f [19]. Since copolyester exhibits lower hygroscopicity, D_k, and D_f than PI, it has become a candidate for FCCL substrate of 4G/5G LTE. Although the commercial copolyester pellet (i.e. Ticona A950) prepared by *p*-hydroxybenzoic acid (HBA), 2-hydroxy-6-naphthoic acid (HNA), and 1,1,1-tris(4-hydroxyphenyl)ethane (THPE) has been found to exhibit high thermal stability [20–22] and excellent mechanical properties [23,24], its film (i.e. commercial Vecstar CTZ) is manufactured by expensive blow-molding technique [25] rather than cost-effective extrusion molding technique because of low melt strength [26] and similar rheological behaviors of Newtonian fluids under high shear rate [27].

In this paper, long-chain branching copolyester pellets with excellent physical properties have been firstly prepared with the modification of proper recipe. Then the copolyester film with competent D_k, D_f, hygroscopicity, melting temperature (T_m), and CTE has been manufactured with T-Die extrusion and heat treatment. Eventually, we have evaluated its applying feasibility for FCCL substrate by hot compression. Therefore, the novelty brought by the paper is the preparation of a long-chain branched HBA/HNA/THPE copolyester with better physical performances (e.g. higher melt strength, lower yellowness index (YI), higher tensile strength, greater antistatic capability, higher elongation, lower CTE, etc.) than commercial Ticona A950 by the effective modification of prescriptions, the manufacture of its film with superior D_k,

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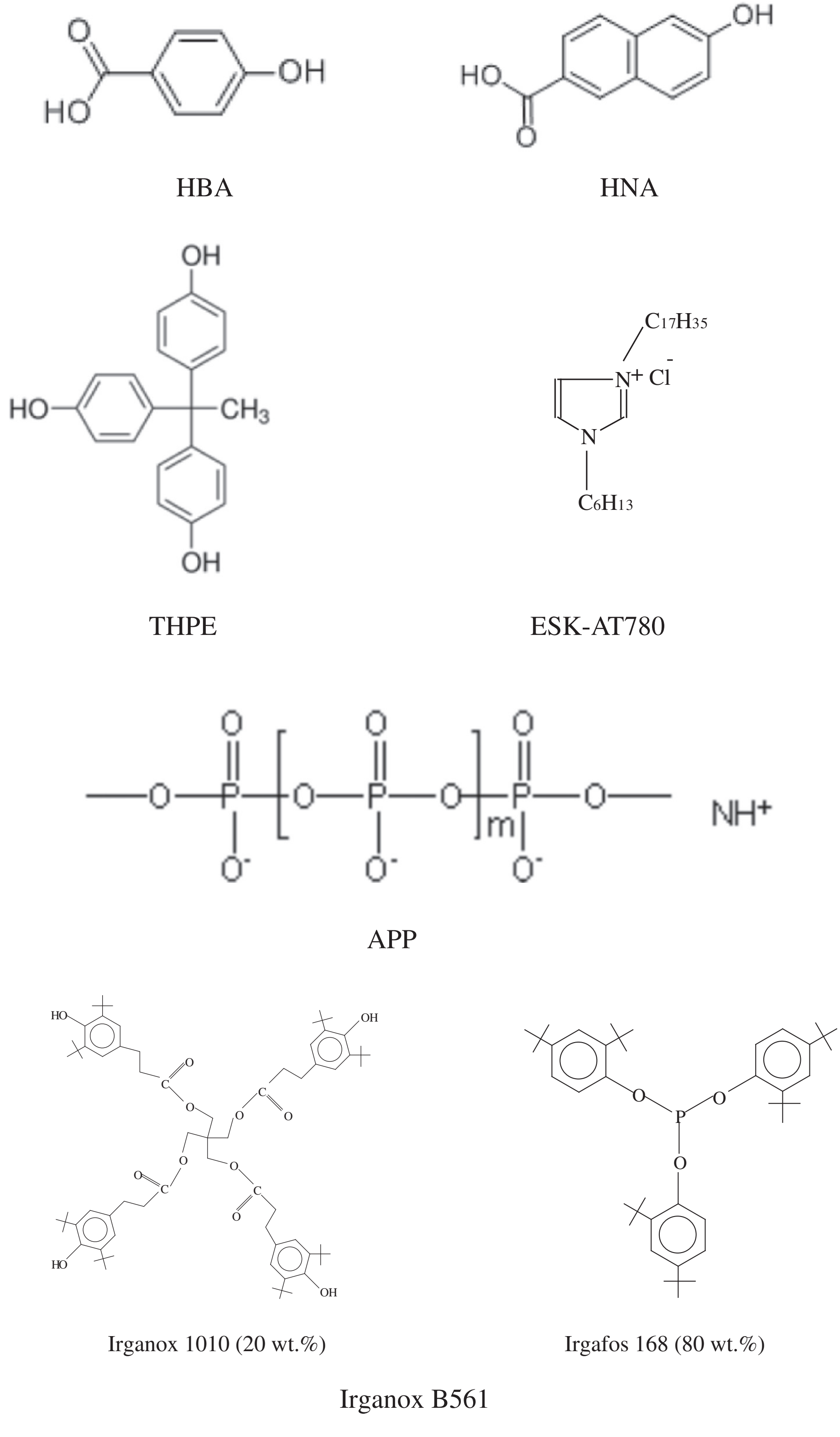

Fig. 1. Chemical structures of HBA, HNA, THPE, ESK-AT780, APP, and Irganox B561.