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Elaboration of AA-BB and AB-type Non-Isocyanate Polyurethanes (NIPUs) using a cross metathesis polymerization between methyl carbamate and methyl carbonate groups

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ABSTRACT

AA-BB and AB-type Non-Isocyanate Polyurethanes (NIPUs) were synthesized through a cross metathesis polymerization between methyl carbamate and methyl carbonate groups within AA-BB and AB monomers. These monomers were prepared by reaction of diamines, diols or amino-alcohols with an excess of dimethylcarbonate (DMC), using 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD) as catalyst. Due to a slight chain extension, dimethyl dicarbonate monomers as well as AB-type monomers contained few carbonate groups, which were present within the final NIPUs. The polymerization was performed in bulk at 200°C, using K₂CO₃ as catalyst. Except for AB-type monomers obtained from amino-alcohols with 3 and 4 methylene groups, the polymerization was successful leading selectively to the expected NIPUs. These materials exhibited good thermal properties with T_g ranging from -35 to 10°C, T_m ranging from 96 to 256°C and degradation temperature above 200°C. The polyurethane properties were generally intermediate comparing to those of polyurea and polycarbonate analogues prepared in the same conditions.

KEYWORDS: Polyurethane, NIPU, dimethylcarbonate, TBD, K₂CO₃.

INTRODUCTION

Polyurethanes (PUs) are an important family of polymer materials. PUs display versatile properties and chemical structures implying their use in various application fields. In the industry, PUs are typically prepared by the reaction of polyols with toxic diisocyanates, which are synthesized from amines and toxic phosgene.¹⁻⁴

During the last few years, efforts have been undertaken in order to develop environmentally friendly alternatives to prepare polyurethanes, using non-isocyanate procedures and biosourced monomers. Three main routes have been described: (i) the preparation of poly(hydroxylurethane)s by aminolysis of five-membered bicyclocarbonates with diamines;⁴⁻¹⁷ (ii) The polymerization of AB-type fatty monomers possessing acyl azide and hydroxyl groups, with or without catalyst. Isocyanate groups are formed *in-situ* via Curtius rearrangement after heating of the acyl azide groups;¹⁸⁻²¹ (iii) The transurethane polycondensation involving a reaction between dialkyl dicarbamates²²⁻²⁵ or dihydroxyethyl dicarbamates²⁶⁻²⁹ with diols, in presence of organo-metallic or organic catalysts, especially 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD).

Other non-isocyanate and non-phosgene methods to prepare PUs exist but not fulfill entirely the above requirements. Indeed, NIPUs can be prepared by cationic ring opening polymerization

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