Effects of Reaction pH and Hardener Type on Reactivity, Properties, and Performance of Urea-Formaldehyde (UF) Resin

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ABSTRACT

This study was conducted to investigate the effects of reaction pH conditions and hardener types on the reactivity, chemical structure and adhesion performance of UF resins. Three different reaction pH conditions, such as traditional alkaline-acid (7.5 → 4.5), weak acid (4.5), and strong acid (1.0), were used to synthesize UF resins which were cured by adding three different hardeners (ammonium chloride, ammonium citrate, and zinc nitrate) to measure adhesion strength. Fourier transform infrared (FT-IR) and carbon-13 nuclear magnetic resonance (13C-NMR) spectroscopies were employed to study chemical structure of the resin prepared under three different reaction pH conditions. Adhesion strength of the resins cured with three different hardeners was determined with lap shear specimens in tension. The gel time of UF resins decreased with an increasing in the amount of both ammonium chloride and ammonium citrate added in the resins. However, the gel time increased for zinc nitrate. Both FT-IR and 13C-NMR spectroscopies showed that the strong reaction pH condition produce uronic structures in UF resin, while both alkaline-acid and weak acid conditions produce quite similar chemical species in the resins. The maximum adhesion strength was occurred with the resin prepared under strong acid pH condition. However, this study indicated that the weak acid reaction condition provide a balance between increasing resin reactivity and improving adhesion strength of UF resin. The measurement of formaldehyde emission from the panels bonded with the UF resins prepared is planned for future work.

Keywords: UF resin, reaction pH, gel time, FT-IR, C13-NMR, hardener types, shear adhesion strength.

1. INTRODUCTION

To a large extent, aminoresin adhesives or aminoplastic adhesives include urea-formaldehyde (UF), melamine-formaldehyde (MF) resin, melamine-urea-formaldehyde (MUF) resin, melamine-fortified UF resin, and melamine-urea-phenol-formaldehyde concondensation (MUPF) resin. The world wide production of UF resins in 1998 was estimated to be approximately 6 billion tons per year based on 66% resin solids by mass (Dunky, 1998).

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Wood-based composite panel industry is a major consumer of UF resins. For example, the consumption of amino resin, including UF resins, MF resins and MUF resins in North America was about 59% of wood-based adhesives in 1997 (Sellers, Jr., 1999). Among these aminoplastic resins, UF resin is a polymeric condensation product of the chemical reaction of formaldehyde with urea. UF resin is the most important type of adhesive in the wood-based panel industry, such as particleboard (PB), medium density fiberboard (MDF), and partly oriented strandboard (OSB), plywood, and some other types of boards.

Compared to other wood adhesives, such as phenol-formaldehyde (PF) resins and diphenylmethane diisocyanate (MDI), UF resin possesses some advantages such as fast curing, good performance in the panel, water solubility and lower price. Disadvantages of using the UF resin are lower resistance to water and its formaldehyde emission from the panels. Lower resistance to water limits UF resin-bonded panels to interior applications. Formaldehyde emission was one of the most important aspects of UF resin in last few decades (Myers 1986). The reversibility of the aminomethylene link and hence the susceptibility to hydrolysis explains lower resistance against the influences of water and moisture, and subsequently formaldehyde emission (Dunky, 1998). Thus, the use of UF resin bonded wood-based composite panels is limited only to non-structural applications due to the lack of water resistance.

Thus, much attention has been paid to reduce or control the formaldehyde emission from UF resin-bonded panels. The presence of free formaldehyde in the UF resins prepared is one of the reasons for formaldehyde emission (Pizzi, 1983). One of the approaches of reducing formaldehyde emission was to lower F/U molar ratio of the resin synthesized (Marutzky, 1986).

In addition, the number of urea additions also influences the properties of prepared UF resin (Steiner, 1973). However, lower F/U molar ratio reduced formaldehyde emission at the expense of poor mechanical properties such as internal bond (IB) strength and modulus of rupture (MOR) (Marutzky, 1986). In order to overcome this problem, many attempts have been made to modify the resin synthesis methods, hardener types, additives, etc (Dunky, 1998).

Until the mid-sixties, most UF resins were synthesized by the two-step reaction procedure, i.e., methyololation and condensation. In other words, the methyololation reaction was done under alkaline condition followed by the condensation reaction under acidic condition (Hse et al., 1994). This synthesis method was widely employed for UF resin preparations for a long time. In early the seventies, however, this method faced the serious problem of formaldehyde emission. Thus, lower F/U molar ratios from 1.1 to 1.2 started to be used in preparing resin. Obviously, these lower F/U molar ratio resins produced poor IB strength of the panel.

In addition to lowering the F/U molar ratio, a number of studies have focused on modifying UF resin properties by manipulating resin synthesis parameters, such as reaction pH condition (Tomita and Hatono, 1978; Gu et al., 1995; Tohmura, 2000; Hse et al., 1994), introduction of second urea addition (Tomita and Hatono, 1978), and the use of additives (Dutkiewicz, 1984; Pizzi, 1994). In particular, a Japanese group (Tomita and Hatono, 1978; Gu et al., 1995; Tohmura, 2000) and Hse et al. (1994) have studied the change of chemical structure of UF resins prepared under different reaction pH conditions. One of the common findings of these studies was the detection of uronic structures in the UF resin prepared under a strong acid condition. Furthermore, Hse et al.