



Process for coating of reconstituted tobacco sheet with citrates



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ABSTRACT

The combustion property and thermal pyrolysis behavior of reconstituted tobacco sheet (RTS) with combustion improvers added to the RTS coatings were studied. The RTS coatings comprised of water-soluble extracts from tobacco stems and leaves, carboxymethyl cellulose and sodium citrate/potassium citrate. The coating was mixed and stirred by a high-speed dispersion machine for 40 min, and then coated on the RTS base paper by papermaking method. Effects of citrates on the properties of RTS, such as major chemical compositions, surface microstructure, tar and carbon monoxide (CO) release per cigarette were investigated. The results showed that adding sodium citrate and potassium citrate in the coatings did not affect the main chemical contents and surface microstructure of RTS. With the ratio of potassium citrate increasing in the coating, the amount of tar and CO release per cigarette decreased significantly. Thermogravimetric (TG) analysis and the corresponding derivative thermogravimetric analysis (DTG) indicated that, compared to sodium citrated, potassium citrate accelerated the thermal degradation of RTS between 200 and 400 °C, and decreased the activation energy (E) according to the kinetic analysis results of Coats–Redfern model. Therefore, potassium citrate may become a potential burning additive controlling CO and tar release in mainstream smoke of RTS.

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1. Introduction

Tobacco (*Nicotiana tabacum*) is a widely cultivated plant in the world consisting of various organic and inorganic substances and biopolymers [1]. During tobacco harvesting and processing, a lot of tobacco wastes were produced that cannot be incorporated directly into cigarettes, such as tobacco stems, mid-ribs, leaf scraps and tobacco dust [2]. Under the development of biorefinery, it is a trend to utilize the biomass wastes or byproducts to develop a more integrated biorefinery system and improve the economy of biomass based industries [3–5]. In order to comprehensively utilize these tobacco wastes, the production of reconstituted tobacco sheet (RTS) has been gradually introduced into tobacco industry [6]. RTS is made of tobacco wastes and mixed with normal tobacco leaves to make cigarettes [7]. The tobacco wastes have less tar content than tobacco leaves, and thus incorporating RTS into the cigarettes could also reduce the harm to human body and the environment [8].

Cast sheet process, slurry process and paper making process are the main RTS preparation methods [9]. For paper making process, tobacco wastes are firstly extracted by water to obtain water-

soluble fraction and tobacco pulp. This special pulp is usually refined mechanically to increase fibrillation of fibers, which can improve the fiber bonding in the following RTS base paper forming process through a papermaking machine. The expanding process for tobacco stems is also used for developing high filling power products [10]. Bleached softwood pulp is always added to increase the RTS strength [7]. In a parallel operation, water-soluble fraction is concentrated to prepare the RTS coating and functional additives may be added. The coatings were coated or impregnated into the RTS base paper. After the following drying and slitting processes, reconstituted tobacco sheet (RTS) was made [9,11]. The coating/impregnating process in the papermaking process provides the possible opportunities to improve RTS properties. For example, by adding additives, coating process is able to improve bulk property, softening property, and liquid penetration performance of RTS [7].

Compared to natural tobacco leaf, reconstitution process of RTS results in a different chemical composition [11]. Therefore, some additives should be incorporated into RTS to modify its qualities reducing smoking harshness, irritation and hazardness. Flavors, mineral ash modifiers, humectants etc., are always added to RTS [12]. For many years, governments and public health authorities have been attempted to make cigarettes with lower tar and CO yields as a way to reduce the hazardness for smokers and the environment [13]. For examples, potassium malate [14], alkali-metal

Abbreviations: RTS, reconstituted tobacco sheet.

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Table 1

Effects of the ratio of sodium citrate and potassium citrate in the coatings on basic chemical compositions of reconstituted tobacco sheet.

The ratio of sodium citrate and potassium citrate	Water-soluble sugars (%)	Total alkaloid (%)	Total nitrogen (%)	Total chlorides (%)
1:0	9.86 ± 0.02	0.98 ± 0.03	1.60 ± 0.03	0.88 ± 0.04
1:1	9.78 ± 0.01	1.01 ± 0.04	1.61 ± 0.02	0.86 ± 0.02
1:2	10.11 ± 0.04	1.04 ± 0.02	1.61 ± 0.01	0.90 ± 0.03
1:3	9.92 ± 0.02	1.03 ± 0.02	1.61 ± 0.01	0.86 ± 0.02
0:1	9.96 ± 0.03	1.02 ± 0.01	1.59 ± 0.02	0.84 ± 0.05

salts (sodium or potassium) [15], $(\text{NH}_4)_2\text{HPO}_4$ and $(\text{NH}_4)_2\text{SO}_4$ [16], nano-manganese-based pigment [17], nano- Fe_2O_3 particle, urea phosphate [18], etc., are all documented to be able to modify the tobacco pyrolysis process or reduce the temperature of burning cone to reduce the level of carbon monoxide and tar in the mainstream smoke.

In this study, burning rate accelerating agents (sodium citrate and potassium citrate) were added in the RTS coating in order to improve the pyrolysis behaviors of RTS. Thermogravimetry analysis (TGA) was used to analyze the thermal degradation behavior. The combustion behavior (tar and CO delivery) of RTS was also investigated. This study might be able to offer theoretical foundation for improving the amount of RTS used in the cigarette and achieving comprehensive utilization of tobacco wastes.

2. Experimental

2.1. Materials

The tobacco stems and tobacco leaf scraps were obtained from Hunan Province, China. The carboxymethyl cellulose (CMC) was purchased from Danisco Corporation. Sodium citrate and potassium citrate were obtained from Sigma–Aldrich.

2.2. Methods

The reconstituted tobacco sheet (RTS) was made by the paper-making method introduced by Gao et al. [7]. In this RTS coating, 0.1% CMC and 1% combustion improvers was added in the concentrated tobacco water-soluble extracts, respectively (based on the dry weight of concentrated tobacco extracts). The combustion improvers consisted of sodium citrate or/and potassium citrate. To prepare reconstituted tobacco coating, 0.1% CMC and 1% combustion improvers were added in concentrated tobacco water-soluble extracts. In order to reach the coating solid content of about 39% additional distilled water was blended in the mixture. Then the mixture was stirred by a high-speed dispersion machine at a speed of 2000 r/min for 40 min. The weight ratio of sodium citrate and potassium citrate in the combustion improver was as follows: 1:0, 1:1, 1:2, 1:3, 0:1. The viscosity of prepared coatings was about 105.0 mpa s as tested by Brookfield DV-Viscometer (produced by Brookfield Cor., USA), and pH was 4.7~4.8 tested by pH-meter (H14221) at 25 °C. The coating method also followed the procedure by Gao et al. [7]. Briefly, the prepared coating was coated on the reconstituted tobacco sheet base paper by a K-Control Coater, and controlling the coating weight was about 36 g/m². The coated paper was dried in an oven and then equilibrated at 23 °C and 50% RH for 48 h for the following tests.

2.3. Physical properties of reconstituted tobacco sheet

The main chemical components of RTS, such as water-soluble sugars, total alkaloid, total nitrogen and total chlorides, were tested by a continuous flow analytical instrument (AA3, SEAL Analytical Co., UK) according to Chinese Tobacco Standards YCT 159-2002, YTC 160-2002, YTC 161-2002 and YTC 162-2002.

2.4. SEM analysis

The surface of RTS was investigated by a scanning electron microscope (SEM, S3700 Hitachi Ltd., Japan). The samples were coated with gold by means of a plasma sputtering apparatus prior to SEM evaluation. The accelerating voltage was 10 kV and working distance was 11 mm.

2.5. Mainstream smoke analysis

RTS was equilibrated at 22 °C and relative humidity (RH) 60% for 48 h. The RTS was then cut and prepared into RTS cigarette (cigarette contains only RTS) by handmade method for smoke analysis. A SM450 linear smoking machine was employed for cigarette mainstream smoke analysis following YC/T 29 and YC/T 30 standards. Tar and carbon monoxide delivery in the mainstream smoke per cigarette were tested. All of the tests were replicated two times.

2.6. Thermogravimetry analysis

The TG analysis was carried out by a thermogravimetric analyzer (TG Q500, TG Cor. USA). Approximately 10 mg of sample was placed in an aluminum pan and used for testing. The high purity nitrogen flow rate was 25 ml/min and the temperature rose from 30 °C to 700 °C with a heating rate of 10 °C/min [10].

3. Results and discussion

3.1. Chemical compositions of reconstituted tobacco sheet with combustion improvers

As shown in Table 1, the ratio of sodium citrate and potassium citrate in the coating hardly affected the chemical compositions of reconstituted tobacco sheet (RTS) including water-soluble sugars, total alkaloid, total nitrogen and total chlorides. When adding combustion improvers in the RTS coating without regard to the ratio of sodium citrate and potassium citrate, the contents of water-soluble sugars, total alkaloid, total nitrogen and total chlorides in the RTS were all close to 10%, 1.0%, 1.6% and 0.9%, respectively (Table 1). The results were consistent with the analytical results of Chinese commercial RTS [19]. It indicated that the combustion improvers would not alter the major chemical compositions of RTS. However, these values were much lower than those from tobacco leaves and tobacco stems [7]. Because in the process of making RTS, pulp fibers and flavors were added [11], and harmful components were removed [12], which slightly reduced the contents of some original chemical components.

3.2. SEM analysis of reconstituted tobacco sheet

In order to analyze the effects of combustion improvers on the surface microstructure of the RTS, SEM analysis were performed (Fig. 1). The different ratio of sodium citrate and potassium citrate in the coating did not bring much change to the microstructure of RTS (Fig. 1). The SEM images were similar to the micrographs of RTS without adding combustion improvers in the coatings, as

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