A FACILE METHOD TO PRODUCE HIGH-BULK AND HIGH-STRENGTH ROLLED RECONSTITUTED TOBACCO SHEET WITH REFINED TOBACCO CELLULOSE FIBERS

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The rolling method is a pivotal means for reconstituted tobacco sheet (RTS) production due to its cost-effectiveness. However, the traditional rolling method is limited by its raw material grinding processes and yields of rolled RTS (RRTS) with subpar strength and bulk. Therefore, it is a challenge to develop a method to produce high-strength and high-bulk RRTS. Here, by replacing traditional raw materials grinding processes with papermaking refining processes, we present a facile and practical method to produce RRTS with elevated bulk and strength. This method separately refines tobacco leaves and stems into long and coarse leave and stem cellulose fibers. These fibers were subsequently reconstructed into RRTS. The detailed process parameters were optimized. The comparative RRTS with flax fibers instead of stem fibers was investigated. The optimal formula of new RRTS was determined. The updated processes, along with the use of refined cellulose fibers, led the RRTS to a significant improvement in strength and bulk, with a 5.3-fold increase in strength, and a 0.7-fold increase in bulk, while smoking qualities were preserved, which surpassed the smoking experience of RRTS with flax fibers. We anticipate this work will enhance the qualities of RTS and facilitate the transition of traditional tobacco industries toward healthier directions.

Keywords: bulk and strength, rolling method, reconstituted tobacco sheet, tobacco powder, tobacco cellulose fibers

INTRODUCTION

Reconstituted tobacco sheet (RTS) is an engineered tobacco product in the tobacco industry that is created by processing tobacco leaves, stems, scraps, or other tobacco by-products into a sheet-like form.[1–4] Because RTS can optimize its formula, alter flavor, improve qualities, lower tar and nicotine content, reduce health risks, increase yield and profitability, and allow manufacturers to make the process more efficient and potentially more sustainable, it becomes a growing trend in tobacco industry.[5,6] Rolled reconstituted tobacco sheet (RRTS) is a specific type of RTS produced using a roller pressing method. For typical RRTS fabrication, the dried tobacco leaves, stems, and scraps are totally ground into tobacco powder by a grinder machine first. Then, the tobacco powder is blended with other ingredients to form a mixture, which subsequently undergoes a rolling process to form a thin sheet.[7,8] The sheet can be incorporated into various tobacco products, such as traditional cigarettes, cigars, and heat-not-burn cigarettes. Due to the simple, efficient, and low-cost advantages, the rolling method has become an attractive option for tobacco manufacturers.[6] The performance of rolled RTS has a vital impact on the product quality, as well as the production cost and economic benefits. Therefore, it is of great importance to conduct investigations on the rolling methods and the obtained products.

Among rolled RTS qualities, mechanical

strength, and bulk properties are crucial.[9,10] Adequate mechanical strength of RRTS is required for processability, such as machine cutting and shredding.[11] Desirable bulk properties are closely related to the filling performance and the qualities of cigarettes. High bulk leads to high filling power, high air permeability, and high economic benefits.[12] The mechanical strength and bulk properties are mainly determined by the processing processes of RRTS. Limited to the grinding processes and the obtained short or rounded tobacco powder, the RRTS produced by traditional rolling methods are constrained by inherent drawbacks of weak mechanical strength and poor bulk properties. To solve this problem, various modified roller pressing methods have been designed. The most notable strategies can be classified into six categories:

(1) Pre-heating[13] – heating the tobacco mixture with steam or microwave before pressing can soften fibers and facilitate better bonding, improving sheet strength and uniformity, and potentially altering smoke characteristics;

(2) Multi-stage roller pressing – this strategy involves passing the tobacco mixture through multiple sets of rollers with varying pressures and speeds; this allows for more controlled compression and density variation within the sheet, improving mechanical strength but potentially decreasing bulk properties;

(3) Roller surface modifications – grooved rollers, perforated rollers with textured surfaces can create grooves or channels within the sheet, improving bulk properties and influencing smoke flow and taste, and potentially reducing mechanical strength;

(4) Scraper-induced wrinkling – wrinkling is a useful method and the degree of wrinkling can be controlled by adjusting the speed of the sheet and the pressure of the scraper against the sheet; this strategy can efficiently improve bulk, but decrease mechanical strength;

(5) Adding multifunctional additives,[14] fillers[15,16] or additional fibers – additives like binders,[17] plasticizers, lubricants, flavorings, and foaming agents can tailor sheet properties for specific needs; adding binders can improve inter-fiber cohesion and improve sheet strength, but potentially decrease bulk properties;[14] incorporating wood fibers can enhance mechanical strength, but smoking taste may be affected;

(6) Hybrid methods – combining multiple strategies from different categories to create synergistic effects and tailor sheet properties.

Overall, the strategies above demonstrate some irrefutable advantages, but some strategies are complex and hard to implement. Meanwhile, they cannot fully solve the problem of weak mechanical strength and poor bulk properties of RRTS. Therefore, there still remains a big challenge to develop a facile and practical method to produce RRTS with high mechanical strength and high bulk properties.

Considering the papermaking mechanical refining process as a substitute for traditional grinding processes to prepare raw materials may solve the problem. Mechanical refining is a method used in the papermaking industry to separate fibers from wood through mechanical processes.[18] In this process, wood chips are passed through a refiner with rotating discs, mechanically refining them into mechanical pulp fibers.[19] The process has a high yield, is cost-effective, easily scalable, and environmentally friendly. Compared with short or rounded tobacco powder produced by the traditional grinding process, fibers produced by the mechanical refining process exhibit several unique characteristics:

(1) Longer and more intact pulp fibers, which provide better inter-fiber bonding and enhance fiber entanglement and network formation within the sheet, leading to improved tensile strength and tear resistance;[20] long fibers also create a more open and porous structure, increasing bulk properties;

(2) High individual fiber tensile strength – the natural strength of wood fibers is retained, contributing to strong sheets;

(3) High fiber fibrillation – refined fibers can lead to fibrillation, where individual fibers are partially split or frayed at the ends; this fibrillation can improve fiber-to-fiber bonding area and enhance strength;[21]

(4) High roughness and coarseness – mechanical refining tends to produce fibers with high roughness and coarseness, contributing to increased sheet thickness and porosity;[22]

(5) Retention of raw material characteristics – refined fibers preserve the inherent properties of raw materials; for tobacco raw materials, nicotine, flavors, and other bioactive components can be retained.[23]

Overall, using the papermaking mechanical refining process to produce tobacco fibers to enhance RRTS mechanical and bulk properties is promising.

Here, by replacing raw materials grinding processes with papermaking refining processes, we developed a facial, and practical method to produce high-bulk and high-strength RRTS. Different from the traditional grinding process of grinding all tobacco materials into short and rounded powder, the improved method takes mechanical refiners to individually separate tobacco leaves and stems into long cellulose fibers. The obtained refined cellulose fibers were blended again and reconstructed to form RRTS. The detailed process parameters, such as beating degrees and loading content, were optimized. The optical formula of the new RRTS was obtained. The results show the mechanical strength and bulk properties were significantly improved. Also, the desirable sensory and smoking qualities were maintained. We anticipate this work may help tobacco manufacturers to improve product quality and enhance consumer satisfaction.

EXPERIMENTAL

Preparation of tobacco powder through grinding processes

An amount of dried tobacco raw material was put into the multi-function high-speed grinder (800Y, Yongkang Boou, China) and was ground at a speed of 34000 r/min for 4 min, and then the obtained materials were screened with a 100-mesh metal mesh sieve to obtain tobacco powder with particles above 100 mesh.

Water extraction of tobacco leaves and tobacco stem

Dried tobacco leaves (30 g, D.W.) were put into a beaker with 180 mL of water (solid-liquid ratio, 1:6). The mixture was heated and the temperature was kept at 65 °C for 60~70 min. Then, the mixture was poured into a filter bag to squeeze out the water. The tobacco extract was collected in a glass bottle with a sealed cap, and then it was refrigerated at 4 °C for use. The process for tobacco stem extraction was the same as that of tobacco leaves.

Preparation of tobacco fibers through papermaking refining processes

Plain water was added to the extracted tobacco leaves (30 g, D.W.) and the weight was adjusted to 300 g. The wet tobacco leaves were stuck on the wall of PFI refiner discs (Mark VI, No.621, Hamjern Maskin, Norway). The refining pressure was set to 1.5 N/mm and refining processes began. When the refining processes stopped, a pulp slurry (2 g, D.W.) was taken out to measure beating degrees with a Schopper type beating degree tester (YQ-Z-13, Hangzhou Nuoding, China) and diluted to 1000 mL. The refined tobacco leaf pulp was placed into a sealed bag with a sealing strip and stored in the refrigerator at a temperature of 4 °C. For tobacco stems, the procedure was similar to that described above for tobacco leaves, with the difference that the tobacco stem needs pre-refining with a papermaking round disc refiner (2500-II, Uchiyama Kikai Co., Ltd., Japan).

Preparation of flax fibers

Because long flax fibers are often used as reinforcing fibers in traditional RRTS, this research comparatively investigated RRTS reinforced with both stem fibers and traditional flax fibers. The preparation of flax fibers was done as follows. A flax pulp sheet (20 g, D.W.) was placed into a beaker with 1 L of water to be soaked for 12 h, and then, put into a deflaker (Lorentzen & Wettre, 260, Sweden) with a speed of 10,000 rpm and subjected to refining. When refining was finished, the flax pulp was taken out and its concentration was readjusted to 10%, then, it was collected in a sealed bag and placed in the refrigerator to balance the moisture.

Slurry mixing

According to the required material ratio, the tobacco leaf fibers and stem fibers or flax fibers were added into a beaker and then transferred to the stirring container of the hand-held electric mixer with 200 mL of water. The mixer was operated at a speed of 10000 r/min for 30 s. After uniformly dispersed, the slurry was poured into a pulp bag and the water was squeezed out. A uniformly mixed fiber paste was obtained.

Preparation of other ingredients

A tobacco extract (8 mL) was put in a beaker. According to the ratio of required materials, glycerol, 1,2-propylene glycol, and carboxymethyl cellulose sodium (CMC) were added and stirred for 1 min with a magnetic rotor stirrer. Then, the solution was poured into the uniformly mixed fiber paste. The paste and solution were mixed uniformly with a glass rod to form a wet paste.

Roller pressing

A 25×25 cm filter cloth was placed on a plastic plate. The wet paste was put in the center of the filter cloth and covered with a piece of plastic film. A glass rod was used to press the wet paste evenly by hand, then subjected to a kitchen roller pressing machine to form the RRTS sheet.

Dry processes

After pressing, the wet RRTS with filter cloth was put into a drying oven together and dried at 60 °C for 4 hours. After drying, the RRTS was separated and placed in a room with a constant temperature of 23 °C and humidity of 50% for 12 hours to balance the moisture content. Finally, it was collected in a sealed bag for storage.

Characterization and calculation

The morphology of tobacco powder, tobacco fibers, and flax fibers was observed by a Paper Fiber Measuring Instrument (XWY-VII, Zhuhai Warren, China). The microstructure of RRTS was observed by SEM (Hitachi, SU5000, Japan). Fiber analysis was performed by a Fiber analyzer (Morfi Compact, Techpap, France).

After measuring the thickness of the RRTS with a thickness measurer (Lorentzen & Wettre, 250, Sweden), the base weight and the bulk of RRTS were calculated.

After measuring the tensile force of RRTS with a tensile testing machine (RH-KZY300, Guangzhou Runhu, China), the tensile index of RTS was calculated.

RESULTS AND DISCUSSION

Preparation of RRTS through papermaking refining processes

Figure 1 illustrates the preparation processes of RRTS by two types of pressing methods. The upper process flow represents the preparation processes of RRTS by the conventional roller pressing method. The lower process flow is the preparation process of RRTS by the improved roller pressing method.

In the conventional roller pressing method, the dried tobacco materials (Fig. 1a) were entirely ground into short and round tobacco powder (Fig. 1b) through grinding processes. The obtained ground tobacco powder was blended with flax fibers (Fig. 1c), water, and other ingredients, including a binder of CMC and white smoke-generating agents of glycerol or 1,2-propylene glycol (Fig. 1d) to form a wet paste, which was subsequently subjected to a roller pressing treatment. Conventional RRTS was obtained and it demonstrates weak strength (Fig. 1e) and poor bulk (Fig. 1f), because of the densely packed, short and round tobacco powder.

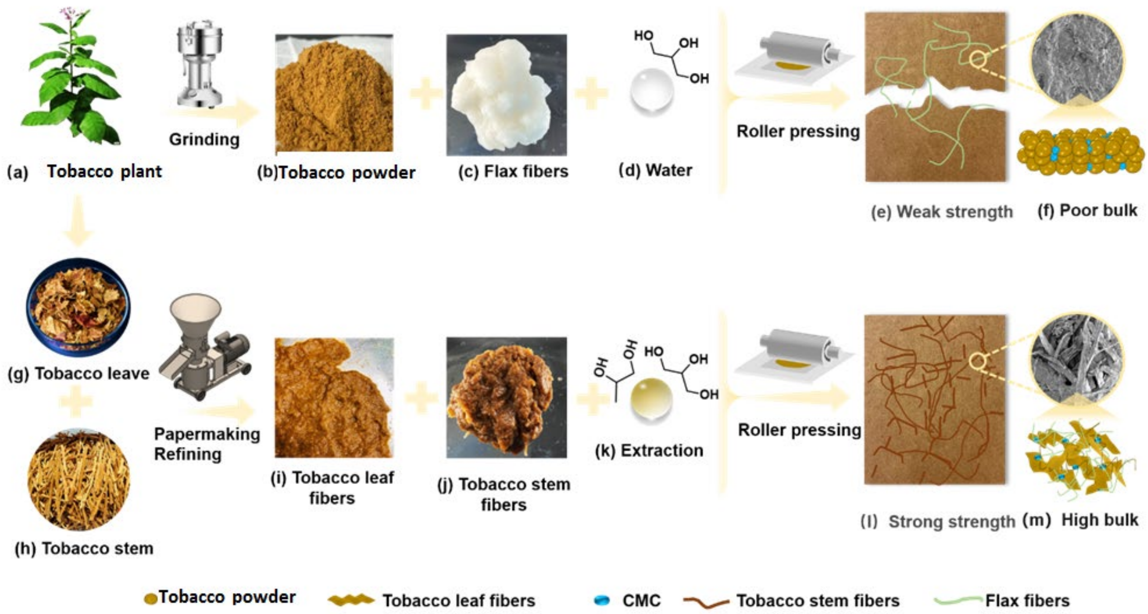


Figure 1: Preparation of RRTS by traditional (upper process flow) and improved roller pressing (lower process flow) methods; (a) starting tobacco materials, (b) dried tobacco entirely ground to tobacco powder through grinding processes, (c) flax fibers, (d) water and other ingredients, (e) weak-strength RRTS prepared by traditional roller pressing methods, (f) SEM image and schematic structure of conventional RRTS with weak strength and poor bulk properties; (g) dried tobacco leaves and (h) dried tobacco stems separately refined into (i) tobacco leaf fibers and (j) tobacco stem fibers through papermaking refining processes, (k) tobacco extracts and other ingredients, (l) high-strength RRTS prepared by improved roller pressing methods, (m) SEM image and schematic structure of improved RRTS with high bulk and high strength proeprties

In the improved method, the dried tobacco materials were sorted into tobacco leaves and scraps (Fig. 1g), and tobacco stems (Fig. 1h). Through papermaking refining processes, tobacco leaves and scraps were refined into long and coarse tobacco leaf fibers (Fig. 1i); tobacco stem was refined into long and coarse tobacco stem fibers (Fig. 1j). The separately refined tobacco leaf fibers and stem fibers were blended with tobacco extracts, binder, and another ingredient to form a wet paste. The paste was pressed and the RRTS sheet was formed, showing high strength (Fig. 1l) and high bulk (Fig. 1m) due to the long and coarse tobacco fibers. The long and coarse tobacco fibers provide better inter-fiber bonding and enhance fiber entanglement and network formation within the sheet, leading to improved strength. Meanwhile, the long and coarse tobacco fibers create a more open and porous structure, resulting in enhanced bulk.

Microscopic morphology characterization

Figure 2 shows the microscopic morphology of tobacco powder, tobacco leaf and stem fibers, and flax fibers. The information on the refining kinetics for the three types of fibers is listed in Table 1. Figures 2a-c are the digital and microscopic images of tobacco powder, illustrating tobacco powder is short and almost round. Figures 2d-f show the top and side view digital images of tobacco leaf fibers at different refining or beating degrees. With the increase of beating degrees from 20, 45 to 52 °SR, the tobacco leaf fibers become more and more smooth.

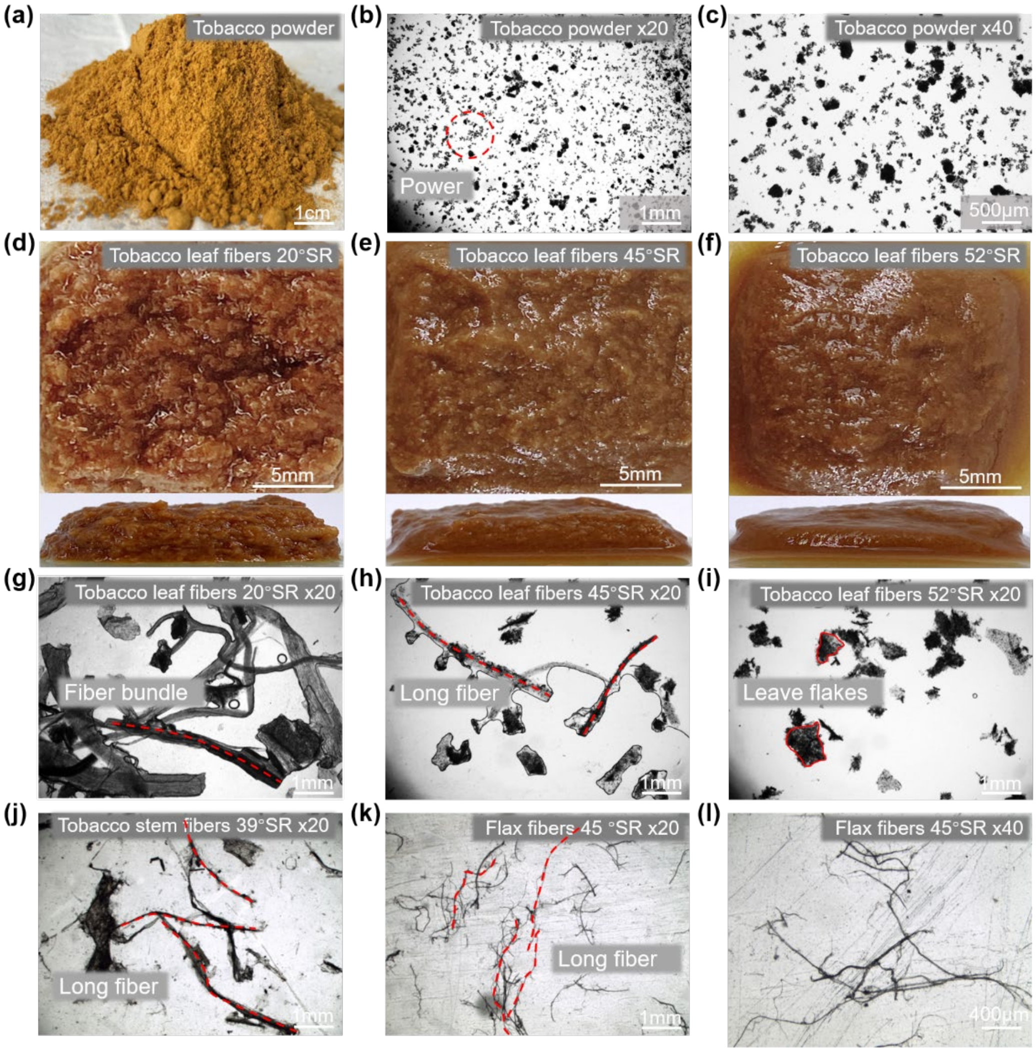


Figure 2: Morphological characterization of tobacco powder, tobacco leaf fibers, tobacco stem fibers, and flax fibers; (a) digital photo image of tobacco powder; microscopic images of tobacco powder at 20× magnification (b) and 40× magnification (c); (d)-(f) digital photo images of tobacco leaf fibers at 20, 45, and 52 °SR; (g)-(i) microscopic images of tobacco leaf fibers at 20, 45, and 52 °SR (20×); (j) microscopic images of tobacco stem fibers at 39 °SR (20×); (k)-(l) microscopic images of flax fibers at 45 °SR (20 and 40×)

Table 1
Refining kinetics of three type of fibers

Type of fiber	Beating degree, °SR						
	0 r, 0 min	4000 r, 3 min	7000 r, 5 min	10000 r, 7 min	15000 r, 10 min	20000 r, 14 min	25000 r, 21 min
Tobacco leaf fibers	-	20	34	40	45	52	55
Tobacco stem fibers	-	-	24	25	39	-	-
Flax fibers	45	66	74	76	-	-	-

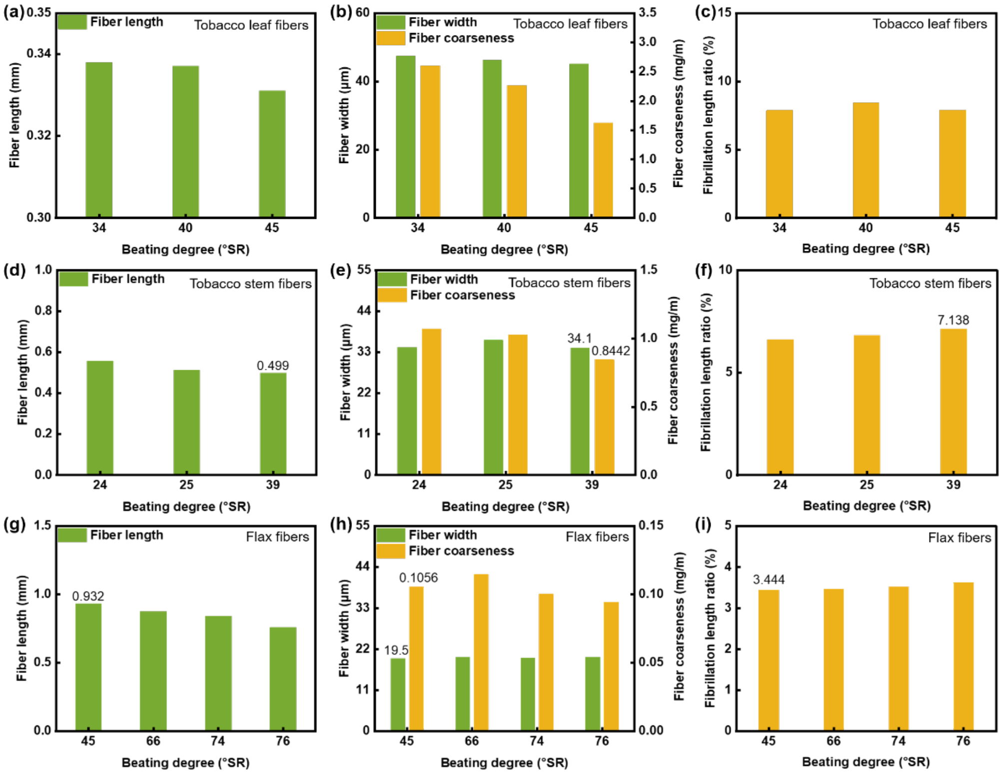


Figure 3: Fiber morphology measurement; (a) fiber length, (b) fiber width and coarseness, and (c) fiber fibrillation length ratio of tobacco leaf fibers with different beating degrees; (d-f) fiber length, width, and coarseness and fibrillation measurement of tobacco stem fibers; (g-i) measurement of flax fibers

Their microscope images are shown in Figure 2g-i. When tobacco leaves were subjected to a beating degree of 20 °SR, the obtained leaf fibers had fiber bundles and not many fines (Fig. 2g). When the beating degrees were increased to 45 °SR, the fiber bundles were broken into fibers and some tobacco fines appeared (Fig. 2h). When the beating degrees were increased to 52 °SR, the tobacco leaf fiber became shorter and more fines can be observed (Fig. 2i). The results show that refining has a vital influence on the morphology of tobacco leaf fibers. Figure 2j shows the microscope images of tobacco stem fibers at 39 °SR (20×), illustrating tobacco stem can be separated into long stem fibers by refining processes. Figures 2k and l present the microscope images of flax fibers at 45 °SR, showing flax fibers have outstanding length. Overall, compared with tobacco powder, tobacco leaf and stem fibers and flax fibers demonstrate better morphology, such as longer length that is beneficial for the improvement of the mechanical properties of the RRTS.

Fiber analysis of tobacco fibers and additional fibers

The morphology of fibers is one of the basic characteristics of fiber raw materials, and it has a vital influence on the properties of RRTS. Table 1 presents the refining kinetics for three types of fibers. The morphology of fibers, including length, width, coarseness, and fiber fractionation length ratio, was measured (Fig. 3). Figures 3a-c show the results of tobacco leaf fibers at 34, 40, and 45 °SR. With the increase of beating degrees, the fiber length, width, and coarseness decrease, while the fibrillation increases. It is likely that refining cuts fiber, breaks fiber walls, and frays fiber ends and surfaces, leading to short, narrow, and fibrillated fibers. The influence of beating on tobacco stem fibers (Fig. 3d-f) and flax fibers (Fig. 3g-i) is similar to that on tobacco leaf fibers. Among tobacco leaf fibers, stem fibers, and flax fibers, flax fibers have the longest length, then stem fibers, while leaf fibers have the shortest length. Also, the tobacco leaf fibers have the highest width and coarseness, followed by stem fibers and flax fibers. The results show flax fibers have the highest length and width ratio, illustrating outstanding flexibility. The tobacco leaf fibers have the lowest length and width ratio, implying they are coarse and stiff. Compared with the long and flexible flax fibers, tobacco stems and leaf fibers are easily fibrillated. Fiber fibrillation can increase surface area and promote inter-fiber bonding, improving mechanical strength. To obtain high fiber fibrillation, we selected tobacco leaf fibers of 45 °SR and stem fibers of 39 °SR to carry out the next step experiment. For flax fibers, we selected flax fiber of 45 °SR to carry out the next step experiment because beating has little influence on the fiber fibrillation.

Effect of fiber proportion and beating degrees on the bulk and tensile strength of RRTS

Instead of tobacco powder, tobacco leaf fibers were the main materials to fabricate RRTS. To improve the RRTS properties, some long fibers are normally added into RRTS. In this work, long fibers of tobacco stem fibers were added. In parallel, flax fibers were also used to construct RRTS. The former is denoted as $RRTS_{(leaf+stem)}$, and the latter is denoted as $RRTS_{(leaf+flax)}$. The meaning for the name of all RRTS as well as the variables in each RRTS are listed in Table 2. The specific recipes and variables for all formulations in this research are listed in Table 3.

Figure 4a presents the effects of the loading content of tobacco stem fibers on the bulk and tensile strength of the $RRTS_{(leaf+stem)}$, wherein tobacco leaf fibers hold a constant beating degree of 45 °SR, and tobacco stem fibers hold a constant beating degree of 39 °SR.

With the increase of the loading content of stem fibers from 0.4% to 0.8%, the bulk and tensile strength all increase first, reach the highest value at 0.6%, and then decrease gradually. A similar trend appears in $RRTS_{(leaf+flax)}$ when the loading content of flax fibers increases (Fig. 4b). The bulk and tensile strength of $RRTS_{(leaf+flax)}$ obtain a high value at 0.6%. These results indicate that the addition of long fibers, including stem and flax fibers, can increase the bulk and tensile strength of RRTS.

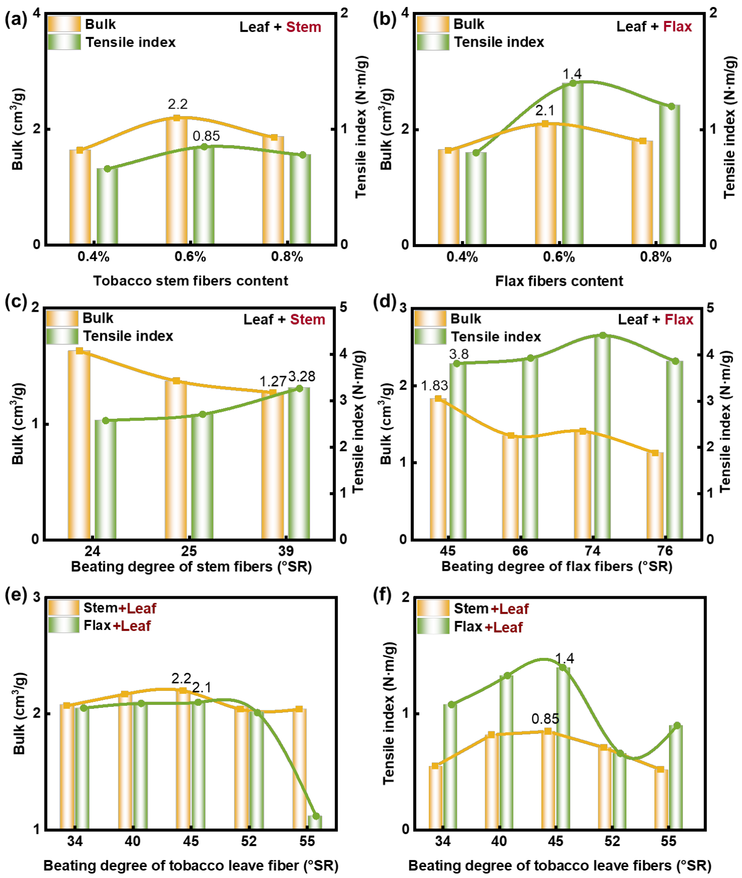


Figure 4: (a) Effect of the loading content of stem fibers on the bulk and tensile strength of the RRTS with tobacco leaf fibers (45 °SR) and stem fibers (39 °SR); (b) Effect of the loading content of flax fibers on the bulk and tensile strength of the RRTS with tobacco leaf fibers (45 °SR) and flax fibers (45 °SR); (c) Effect of various beating degrees of stem fibers on the bulk and tensile strength of the $RRTS_{(leaf+stem)}$; (d) Effect of various beating degrees of flax fibers on the bulk and tensile strength of $RRTS_{(leaf+flax)}$; (e) Effect of various beating degrees of tobacco leaf fibers on the bulk of the $RRTS_{(leaf+stem)}$ and $RRTS_{(leaf+flax)}$; (f) Effect of various beating degrees of tobacco leaf fibers on the tensile of the $RRTS_{(leaf+stem)}$ and $RRTS_{(leaf+flax)}$

Table 2
Denotation of RRTS samples

Denotation	Meaning
$RRTS_{(leaf+stem)}$	RRTS consists of leaf and stem fibers, where stem is the variable
$RRTS_{(leaf+flax)}$	RRTS consists of leaf and flax fibers, where flax is the variable
$RRTS_{(stem+leaf)}$	RRTS consists of leaf and stem fibers, where leaf is the variable
$RRTS_{(flax+leaf)}$	RRTS consists of leaf and flax fibers, where leaf is the variable
$RRTS_{(leaf+stem+CMC)}$	RRTS consists of leaf and stem fibers, and CMC, where CMC is the variable
$RRTS_{(leaf+flax+CMC)}$	RRTS consists of leaf and flax fibers, and CMC, where CMC is the variable

*Glycerol and 1,2-propylene glycol were included in all RRTS

Table 3
Recipes and variables in the formulation

	Formulation						Variable (VAR)	
	Leaf	Stem		Flax		CMC		
	Beating degree, °SR	Beating degree, °SR	Content, %	Beating degree, °SR	Content, %	Content, %	Details	Best
Fig. 4a	45	39	VAR	w/o	w/o	w/o	0.4,0.6,0.8	0.6%
Fig. 4b	45	w/o	w/o	45	VAR	w/o	0.4,0.6,0.8	0.6%
Fig. 4c	45	VAR	0.6	w/o	w/o	0.5	24,25,39	45°SR
Fig. 4d	45	w/o	w/o	VAR	0.6	0.5	45,66,74,76	45°SR
Fig. 4e	VAR	39	0.6	w/o	w/o	w/o	34,40,45, 52, 55	45°SR
Fig. 4f	VAR	w/o	w/o	45	0.6	w/o	34,40,45, 52,55	45°SR
Fig. 6a	45	39	0.6	w/o	w/o	VAR	0.3,0.5,0.6	0.6%
Fig. 6b	45	w/o	w/o	45	0.6	VAR	0.2,0.3,0.5	0.5%

*"VAR" represents the variable in this formulation; "w/o" means that content was not included; glycerol and 1,2-propylene glycol were all included in all formulations

It can be explained by the fact that long fibers, such as stem and flax fibers, can provide better inter-fiber bonding and enhance fiber entanglement and network formation, leading to enhanced tensile strength. Long fibers of stem and flax fibers also create a more open and porous structure, resulting in increased bulk properties.

Figure 4c presents the effect of various beating degrees of tobacco stem fibers on the bulk and tensile strength of the $RRTS_{(leaf+stem)}$, wherein stem fibers were added with a content of 0.6% and tobacco leaf fibers held a beating degree of 45 °SR. With the increase of the beating degree of stem fibers from 24 to 39 °SR, the bulk of $RRTS_{(leaf+stem)}$ decreases gradually, while the tensile strength increases gradually, demonstrating beating has a positive effect on bulk and an adverse effect on tensile strength. Figure 4d presents the effect of various beating degrees of flax fibers on the bulk and tensile strength of the $RRTS_{(leaf+flax)}$, wherein flax fibers were added with a content of 0.6% and tobacco leaf fibers held a beating degree of 45 °SR. With the increase of the beating degree of flax fibers from 45 to 76 °SR, the bulk of $RRTS_{(leaf+flax)}$ generally decreases gradually, while the tensile strength increases first and then decreases gradually, showing a similar trend to that of $RRTS_{(leaf+stem)}$. The results show the increase in the beating degree of long fibers is beneficial for the improvement of tensile strength, but harmful for the improvement of bulk.

Figure 4e presents the effect of various beating degrees of tobacco leaf fibers on the bulk of the $RRTS_{(leaf+stem)}$ and $RRTS_{(leaf+flax)}$, wherein stem fibers were added with a content of 0.6% and held a beating degree of 39 °SR, and flax fibers were loaded with a content of 0.6% and held a beating degree of 45 °SR. With the increase of the beating degree of leaf fibers from 34 to 55 °SR, the bulk of $RRTS_{(leaf+stem)}$ and $RRTS_{(leaf+flax)}$ almost keeps constant first and then decreases gradually, implying beating likely increases the flexibility of fibers and leads to decreased bulk. Meanwhile, $RRTS_{(leaf+stem)}$ shows a better bulk than

RRTS$_{(leaf+flax)}$. It is likely attributed to the fact that stem fibers have a higher coarseness than flax fibers. Figure 4f presents the effect of various beating degrees of tobacco leaf fibers on the tensile strength of the RRTS$_{(leaf+stem)}$ and RRTS$_{(leaf+flax)}$, wherein stem fibers were added with a content of 0.6% and held a beating degree of 39 °SR, and flax fibers were loaded with a content of 0.6% and held a beating degree of 45 °SR. With the increase of the beating degrees of leaf fibers from 34 to 55 °SR, the tensile strength of RRTS$_{(leaf+stem)}$ and RRTS$_{(leaf+flax)}$ increases first, reaches the highest value at 45 °SR and then decreases gradually, indicating fiber fibrillation predominately contributes to the increase of tensile strength before 45 °SR, and after 45 °SR, fiber cutting predominately contributes to the decrease of tensile strength. Meanwhile, RRTS$_{(leaf+flax)}$ shows a better tensile strength than RRTS$_{(leaf+stem)}$. It should be ascribed to the fact that flax fibers have a longer length than stem fibers. Figure 5 shows the digital photo and SEM images of RRTS$_{(stem+leaf)}$ and RRTS$_{(flax+leaf)}$ with leaf fibers refined at various beating degrees.

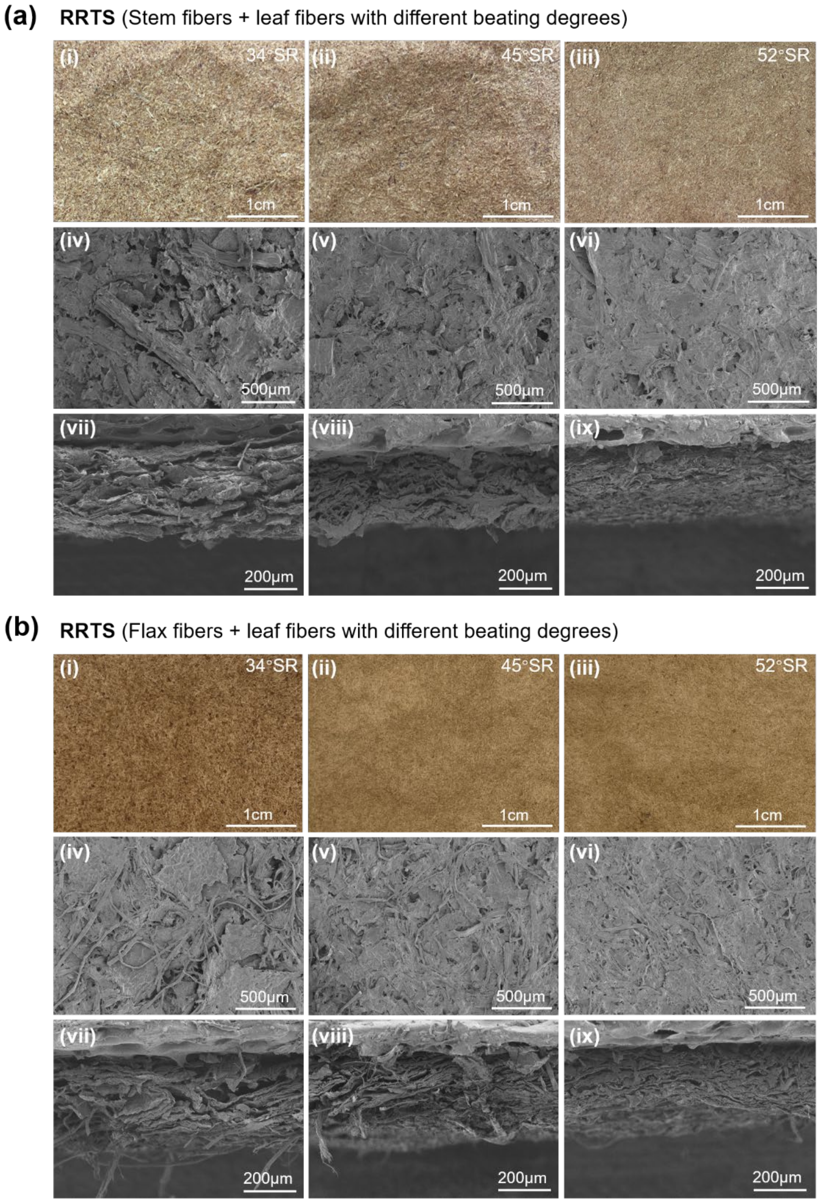


Figure 5: Digital photo and SEM images of RRTS$_{(stem+leaf)}$ and RRTS$_{(flax+leaf)}$ with leaf fibers of various beating degrees. (a) Digital photo images of RRTS$_{(stem+leaf)}$, including top view digital photo image (i)-(iii), top view SEM image (iv)-(vi) and cross-section SEM image (vii)-(ix); (b) Digital photo images of RRTS$_{(flax+leaf)}$, including top view digital photo image (i)-(iii), top view SEM image (iv)-(vi) and cross-section SEM image (vii)-(ix)

With the increase of the beating degrees of leaf fibers, the surface of both $RRTS_{(stem+leaf)}$ and $RRTS_{(flax+leaf)}$ becomes smoother and smoother, and the inner structure becomes denser and denser, implying the beating of leaf fibers decreases the bulk and potentially increases the strength of RRTS due to the dense structure.

Overall, to achieve optimal and balanced bulk and tensile strength, we chose the following formula to subsequently construct RRTS. For $RRTS_{(leaf+stem)}$, tobacco leaf fibers (45 ºSR) and stem fibers (0.6%, 39 ºSR) will be used. For $RRTS_{(leaf+flax)}$, tobacco leaf fibers (45 ºSR) and flax fibers (0.6%, 45 ºSR) will be used.

Fabrication of RRTS and smoking quality evaluation

Besides the loading content and beating degrees of long fibers, and beating degrees of tobacco leaf fibers, the loading content of CMC has an important influence on the bulk and tensile strength. Figure 6a shows the effect of the CMC content on the bulk and tensile index of $RRTS_{(leaf+stem+CMC)}$ with tobacco leaf fibers (45 ºSR), stem fibers (0.6%, 39 ºSR), and CMC. With the increase of CMC from 0.3% to 0.6%, the bulk of RRTS decreases, while the tensile strength increases sharply, indicating the addition of CMC can efficiently increase the tensile strength of RRTS. Figure 6b shows the effect of the CMC content on the bulk and tensile index of $RRTS_{(leaf+flax+CMC)}$ with tobacco leaf fibers (45 ºSR), flax fibers (0.6%, 45 ºSR), and CMC. With the increase of CMC from 0.2% to 0.5%, the bulk of RRTS decreases first, and then increases slightly, while the tensile strength increases sharply. The results of Figures 6a and b illustrate the addition of CMC has a positive effect on tensile strength and a negative effect on bulk. With the results in Figure 4, we finally obtained the best formula for two new RRTS: (1) $RRTS_{(leaf+stem+CMC)}$: tobacco leaf fibers (4.8 g, D.W., 45 °SR); tobacco stem fibers (0.4 g, D.W., 39 °SR), CMC (0.4 g, 0.6%), 1,2-propylene glycol (0.3 g), glycerol (0.7 g), tobacco extract (8 g), water (46.8 g); and (2) $RRTS_{(leaf+flax+CMC)}$: tobacco leaf fibers (4.8 g, D.W., 45 °SR); flax fibers (0.4 g, D.W., 45 °SR), CMC (0.3 g, 0.5%), 1,2-propylene glycol (0.3 g), glycerol (0.7 g), tobacco extract (8 g), water (46.8 g). The best formulation was summarized in Tables 4 and 5. For $RRTS_{(leaf+stem+CMC)}$, the bulk is 1.32 cm^3/g and the tensile index is 3.69 N·m/g. For $RRTS_{(leaf+flax+CMC)}$, the bulk is 1.87 cm^3/g and the tensile index is 4.26 N·m/g. These bulk and tensile strength values are all far better than those of traditional RRTS, with a bulk of 0.77 cm^3/g and tensile index of 0.58 N·m/g, demonstrating the exceptional advantages of our improved RRTS method.

Figures 6c and d show the digital photo and SEM top view images of traditional RRTS and improved $RRTS_{(leaf+stem+CMC)}$. A smooth and dense surface can be observed for traditional RRTS due to the densely packed tobacco powder (Fig. 6c). After improvement, the long and coarse tobacco leaf and stem fibers replaced tobacco powder and created a rough and porous surface (Fig. 6d). The updated structure also leads to improvements in bulk and tensile strength. Figures 6e and f show a comparison of the performance of RRTS of this work and previously published studies. Compared with the traditional RRTS in sample 3, our improved RRTS obtained a significant enhancement in bulk and strength. For $RRTS_{(leaf+stem+CMC)}$, the bulk increases by 70%, and the tensile strength increases by 530% (sample 1, Fig. 6e and 6f). For $RRTS_{(leaf+flax+CMC)}$, the bulk increases by 143%, and the tensile strength increases by 634% (sample 2, Fig. 6e and 6f). The data of samples 4, 5, and 6 are the results of previous literature (Fig. 6e and 6f). Roll pressing methods were used in samples 4 and 5 and CMC loading content was optimized, while in sample 6 papermaking methods were used. The bulk and tensile strength of samples 4 and 5 produced by roll pressing methods are much lower than that of our work.

Table 4
Best formulation for $RRTS_{(leaf+stem+CMC)}$

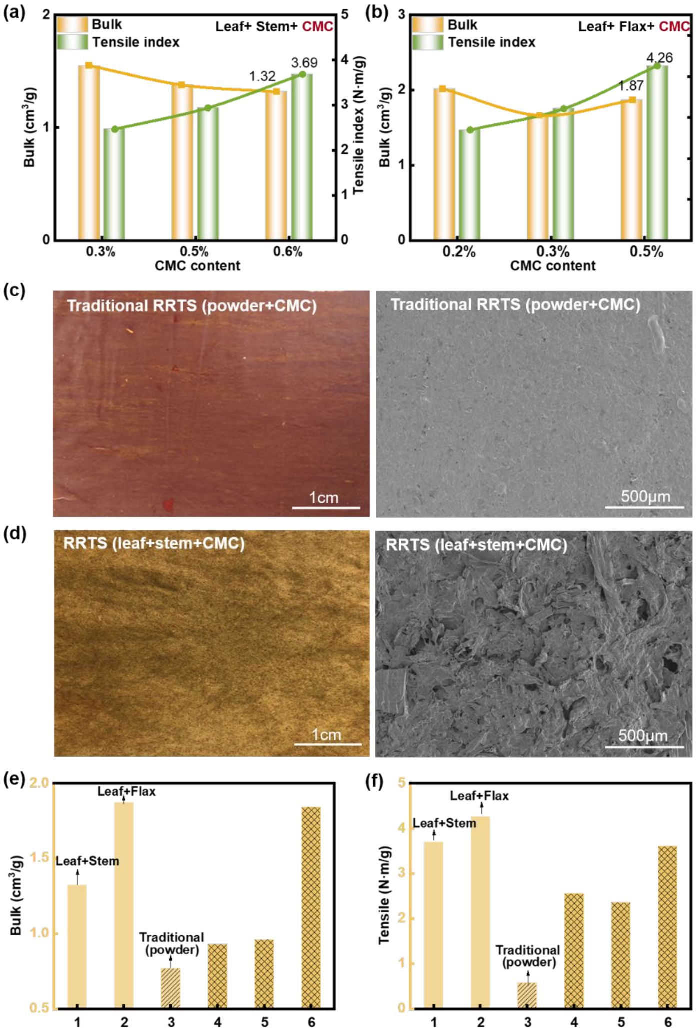
Leaf		Stem		CMC	1,2-Propylene glycol, g	Glycerol, g	Tobacco extract, g	Water, g
Beating degree, °SR	Content, g	Beating degree, °SR	Content, g	Content, g				
45	4.8	39	0.4	0.4	0.3	0.7	8	46.8

* The weight in "Content, g" refers to dried weight

Table 5
Best formulation for RRTS(leaf+flax+CMC)

Leaf		Flax		CMC/g	1,2-Propylene glycol, g	Glycerol,g	Tobacco extract, g	Water, g
Beating degree, °SR	Content, g	Beating degree, °SR	Content, g	Content, g				
45	4.8	45	0.4	0.3	0.3	0.7	8	46.8

* The weight in "Content, g" refers to dried weight



(g)

No.	Items	Sheet wettability	Bulk	First smoke volume	Overall smoke volume	Consist-ency[b]	Practicab-ility
1	Traditional RRTS (powder+CMC)	Dry	P[a]	M–	G	M	G–
2	RRTS(leaf +stem+CMC)	Slightly Wet	G	G–	G–	G–	G–
3	RRTS(leaf+ flax+CMC)	Slightly Wet	G	G–	M	M	M

Figure 6: Fabrication of RRTS and smoking quality evaluation; (a) Effect of CMC content on the bulk and tensile index of RRTS(leaf+stem+CMC); (b) Effect of the CMC content on the bulk and tensile index of RRTS (leaf+flax+CMC); (c) Digital photo and SEM top view images of traditional RRTS with tobacco powder; (d) Digital photo and SEM top view images of RRTS(leaf+stem+CMC); (e) Bulk and (f) Tensile strength of RRTS of this work and previously reported study; (g) Smoking quality evaluation of the traditional RRTS with tobacco powder, and improved RRTS(leaf+stem+CMC) and RRTS(leaf+flax+CMC) Note: [a] G is for good, M is for moderate, and P is for poor; [b] Consistency is the fluctuation of each puff of smoke, the smaller the fluctuation, the better the consistency

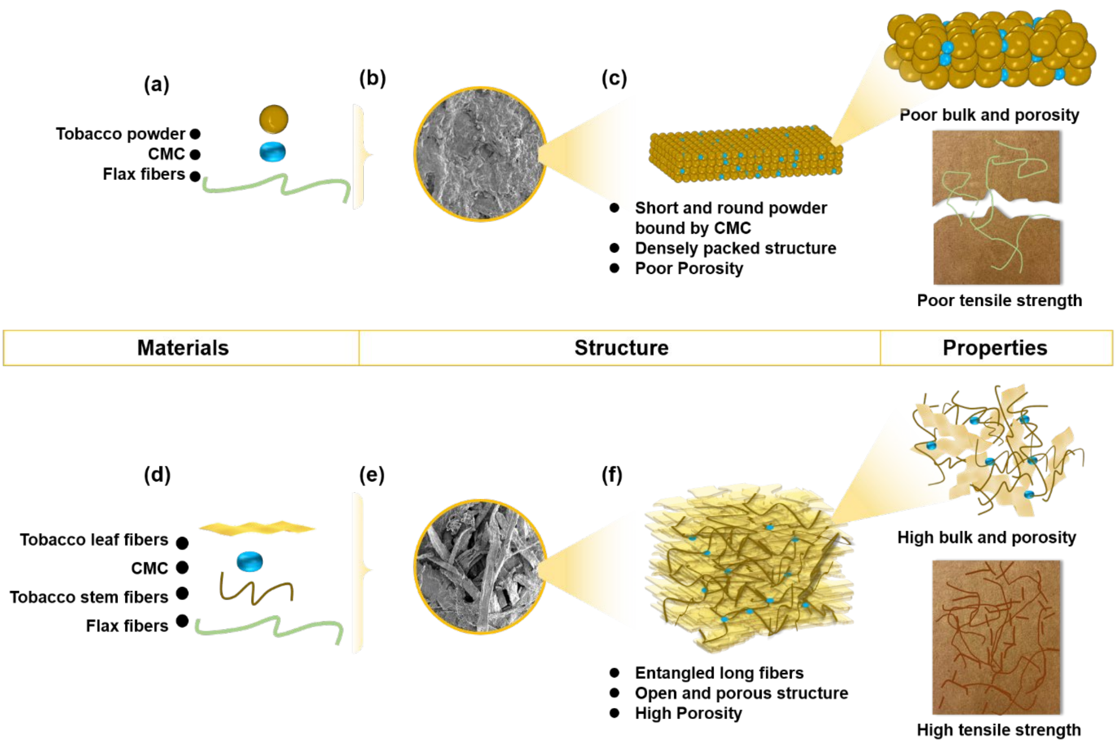


Figure 7: Schematic illustration of the mechanism of the improvement in bulk and strength; (a) Raw materials of traditional RRTS with tobacco powder (b) Surface SEM image of traditional RRTS (c) Proposed generation mechanism of the bulk and strength of traditional RRTS; (d) Raw materials of improved RRTS with tobacco leaf and stem fibers; (e) Surface SEM image of improved RRTS with tobacco leave and stem fibers; (f) Proposed improvement mechanism of the bulk and strength of RRTS, theoretically verifying tobacco leaf and stem fibers to replace tobacco powder can improve the bulk and strength

Although the bulk and tensile strength of sample 6 produced by papermaking methods are comparable with that of our rolled tobacco sheets, our sheets are low-cost and the processes are simpler and more efficient. The papermaking methods require specialized equipment and expertise and the processes are far more complex than roller pressing methods, making the produced sheets reach high production costs.

Finally, we performed a smoking quality evaluation with traditional RRTS, improved $RRTS_{(leaf+stem+CMC)}$ and $RRTS_{(leaf+flax+CMC)}$. For traditional RRTS, the sheet is dry, the bulk is poor, and the consistency is moderate. For $RRTS_{(leaf+stem+CMC)}$, the sheet is slightly wet, the bulk is good, and the smoke volume and consistency are good. For $RRTS_{(leaf+flax+CMC)}$, the sheet is slightly wet, the bulk is good, and the smoke volume and consistency are moderate. Meanwhile, the traditional RRTS and $RRTS_{(leaf+stem+CMC)}$ have similar smoking sensory qualities that are better than that of $RRTS_{(leaf+flax+CMC)}$. Thus, from the standpoint of the bulk and tensile strength, $RRTS_{(leaf+flax+CMC)}$ is better than $RRTS_{(leaf+stem+CMC)}$. But from the standpoint of smoking sensory quality, $RRTS_{(leaf+stem+CMC)}$ is better than $RRTS_{(leaf+flax+CMC)}$. It is likely attributed to the tobacco stem fibers, which preserve most of the inherent properties of the raw materials, such as nicotine, flavors, and other bioactive components, leading to the smoking experience being enriched.

Schematic illustration of the mechanism of the bulk and strength improvement

Figure 7a shows the main raw material of the traditional RRTS. Traditional RRTS is mainly constructed with small-size tobacco powder. The small-size powder is closely bound by CMC, leading to a smooth surface (Fig. 7b). Meanwhile, the powder is densely packed within the sheet, resulting in poor bulk and weak strength (Fig. 7c).

In this work, the tobacco powder was replaced with refined tobacco leaf and stem fibers (Fig. 7d). The long and coarse tobacco fibers generate a rough surface (Fig. 7e) and create a porous structure and enhanced fiber entanglement (Fig. 7f), leading to high bulk and high strength properties. The improved bulk and strength are attributed to papermaking refining processes, which produce long and intact, high-tensile strength, highly fibrillated, and coarse fibers. Moreover, refined fibers retain more of the inherent properties of tobacco raw materials and improve the smoking experience.

CONCLUSION

In summary, we have developed a facile and practical method to produce high-bulk and high-strength RRTS. The traditional raw materials grinding process was replaced by the papermaking refining process. Long and coarse tobacco fibers were obtained. The process parameters were optimized, and the optimal formula of the new RRTS was achieved. The comparative investigation of the RRTS with flax fibers instead of stem fibers was also conducted. Benefiting from the updated refining process and the long and coarse tobacco fibers, the strength and bulk of RRTS were significantly improved, while its sensory and smoking qualities were retained. The results indicate the developed RRTS exhibits superior overall characteristics. With the facile, practical, and potentially scalable method, coupled with the elevated RTS qualities, this work may offer tobacco manufacturers a viable path to improve product quality and shift from traditional tobacco manufacturers towards a greener and healthier direction.

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