Effect of Refining on Physical Properties and Paper Strength of *Pinus massoniana* and China Fir Cellulose Fibers

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To obtain a suitable refining process for *Pinus massoniana* cellulose fibers (PMCF) and China fir cellulose fibers (CFCF), the effects of the beating gap and the pulp consistency on the physical properties and the morphology of the two cellulose fibers were investigated. The results showed that the physical properties of the PMCF and the CFCF were well affected by the beating gap and the pulp consistency. The CFCF showed a smaller weight-average length and width than that of the PMCF. The CFCF exhibited smaller weight-average length, width, and kink index than the PMCF. It is easy to get the high beating degree, indicating it is more easily to be refined. Additionally, the tensile index and burst index of PMCFP and CFCFP increased with increasing beating degree, while the tear index decreased. Compared to the CFCF, the paper made from PMCF had superior strength properties. Consequently, the PMCF was suitable for refining with a high pulp consistency and a medium beating gap, whereas the CFCF had a medium pulp consistency and a big beating gap.

Keywords: Beating degree; Mechanical properties; Papermaking; Refining process; Wood fiber

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INTRODUCTION

Cellulose fiber is an important raw material for preparing paper and fiber composites. As a polymer, the cellulose fibers consist of a long flat chain of cellobiose base units, which expose the abundant hydroxyl groups, allowing the forming of hydrogen bonds (Clark 1985; Xie *et al.* 2011; Chen *et al.* 2012). Paper is a bonded random network of natural cellulosic fibers that can adhere together as a mat by forming intermolecular hydrogen bonds between adjacent fibers surfaces. The strength of paper depends on the tensile strength of individual fibers in the network and the bond strength (Page 1969). To obtain the desired strength properties of paper, the original qualities of the fibers and sufficient bonding strength among the fibers should be considered (Bhardwaj *et al.* 2004, 2007; Molin and Daniel 2004). According to Page's study, the breaking length expressing the tensile strength of strip increases linearly with the increasing bond strength per unit area and relative bonded area, and the relative bonded area-tensile strength relationship is independent of whether the strength is enhanced by mechanical beating or wet pressing (Page 1969). So, increasing the surface area through means of a mechanical refining treatment is an important method to improve the fiber's bonding strength and to form a fibrous network (Kang and Paulapuro 2006; Hubbe 2007; Kappel *et al.* 2009; Marais and Wågberg 2012; Gulsoy 2014).

Mechanical refining is a complex process, which is mainly dependent upon the function of the friction among fibers and the mechanical shear force. This process produces heavy changes in the fiber structure, such as cutting, swelling of fiber walls, fibrillation, flexibility increase, curling, *etc.* (Molin and Daniel 2004; Chen *et al.* 2012). During beating, most of the specific fiber surface areas are exposed with the breaking of swollen fibers. Due to linkages between fibers, the cohesion of the paper is improved, which is beneficial to the retention of cellulosic fines and additives (Rouger and Mutje 1984; Hou *et al.* 2011; Gao *et al.* 2012). However, mechanical beating also damages fibers. Refining will shorten the fibers, producing fiber fines, external and internal fibrillation, and changes in their shape (Page 1989). For example, refining leads the external fibrillation and the formation of fiber fines when part of the outermost cell wall is removed. According to studies of PFI-beaten pulps, the P/S1 layers of fibers are removed in the refining process. And the S2 layer of fibers is damaged in extensive refining (Giertz 1958; Kibblewhite 1972; Molin and Daniel 2004). The physical properties of fibers are always affected by beating factors, such as disc gap and pulp consistency. For example, the medium consistency refining process, depending on the friction between the fibers, can improve the fibrillation of fibers and maintain their length (Wei *et al.* 2010; Hou *et al.* 2011; Chen *et al.* 2012; Gao *et al.* 2012; Alcala *et al.* 2013). There is also difference in the various types of fibers during refining. To obtain the degree of 40 °SR, tobacco stalk and eucalyptus kraft pulp need a PFI mill revolution number of about 4500r, whereas the organosolv pulp from olive tree trimmings only need approximately 1500r (Mutje *et al.* 2005; Gao *et al.* 2012). Therefore, a refining treatment should improve the bonding strength without an excessive decrease in the fiber strength, which is very essential for obtaining products with the desired properties. The goal of this study was to study the different effects of refining on *Pinus massoniana* cellulose fibers (PMCF) and China fir cellulose fibers (CFCF). To obtain suitable refining processes for PMCF and CFCF, the morphology and physical properties of the cellulose fibers and the corresponding mechanical properties of paper were tested and analyzed.

EXPERIMENTAL

Materials and Pretreatment

Paperboard made of *Pinus massoniana* pulp and China fir pulp were purchased from the Qingzhou Papermaking Plant in Sanming (China), and used to prepare the PMCF and CFCF, respectively. They were pretreated with water at 15.0 wt.% for 120 min at ambient temperature and dissociated by the beating machine (ZQS2, Northwest Institute of Light Industry Machinery Factory, Xianyang, China). After the pretreatment, the pulps and water were separated. *Pinus massoniana* pulp and China fir pulp of 8 °SR and 12 °SR, respectively, were obtained for the mechanical pulping.

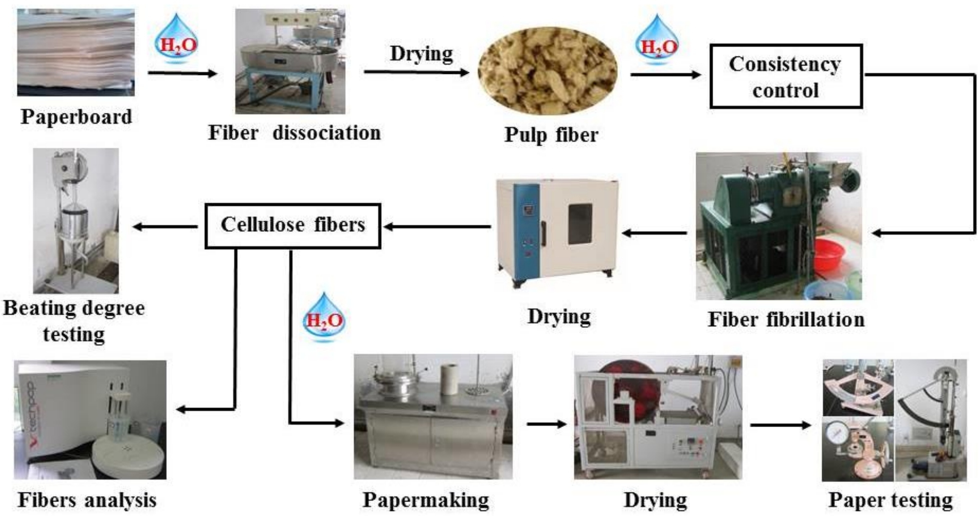

Fig. 1. The preparation process of cellulose fibers and papers

Mechanical Pulping

The pulps were refined in a disc refiner (ZDP-32, Jilin Paper Machinery Manufacturing Factory, Jilin, China). The preparation process of the cellulose fibers and papers is described in Fig. 1. The nomenclatures of papermaking from the PMCF and CFCF are PMCFP and CFCFP, respectively. The highest pulp consistency and smallest beating gap of disc refiner were 10% and 10 μm, respectively. To study the effects of refining on PMCF and CFCF by single factor (pulp consistency or beating gap), the refining process was divided into two parts with different parameters. First, beating gap used in the refining process were under various levels of 10, 20, 30, 40, 50, 150, 250, and 350 μm with a constant pulp consistency of 8%. In this part, a suitable beating gap of 20 μm was obtained. So, the other part of the refining process occurred under various pulp consistencies (4, 6, 8, and 10%) with a constant beating gap of 20 μm.

Testing of beating degree and physical properties of cellulose fibers

The beating degree of the pulps was determined by the Schopper Riegler method, using a beating degree tester (Z-DZY-100, Sichuan Changjiang Papermaking Equipment Co., Ltd., Yibin, China). A total of 2 g of pulp was diluted in 1000 mL of distilled water. The beating degree value is calculated in Eq. 1 according to GB/T 3332 (2004),

$$\text{Beating degree (°SR)} = (1000 - V_{\text{water}}) / 10 \qquad (1)$$

where V_{water} is the volume of transudatory water.

The properties of the cellulose fibers including the fiber weight-average length, fiber width, curl index, degree of fibrillation, and fiber fines were characterized by a fiber morphology analyzer (Morfi Compact, Techpap Co., Ltd., Saint Martin d'Hères, France).

Morphology of cellulose fibers

The morphology *of Pinus massoniana* cellulose fibers was analyzed by optical microscopy (JXB-D, Shanghai Rectangular Optical Instrument Ltd., Shanghai, China). A small specimen of cellulose fibers taken from refined *Pinus massoniana* fibers was placed on the glass slide with a drop of water (disperse fibers). Then, when the sample was dried at ambient temperature, it was tested by optical microscopy with magnification of ×100.

Papermaking and testing of their properties

The refined pulps were dispersed by water at a standard pulp disintegrator of 5 wt.% to 10 wt.%. The papers were prepared on a basis weight of 160 g/m^2. When the papers were prepared, they were placed at a constant temperature and relative humidity, according to standard TAPPI T222 om-11 (2011), for 24 h. The physical properties of the papers, including tensile index, tear index, and burst index, were tested in accordance with GB/T 12914 (2008), GB/T 455 (2002), and GB/T 454 (2002), respectively. The results reported are the average of 10 replicates.

RESULTS AND DISCUSSION

Effect of Beating Gap on the Beating Degree and Physical Properties of Cellulose Fibers

The beating degree and physical properties of PMCF and CFCF were obtained from various beating gaps with a pulp consistency of 8% (Fig. 2 and Table 1).

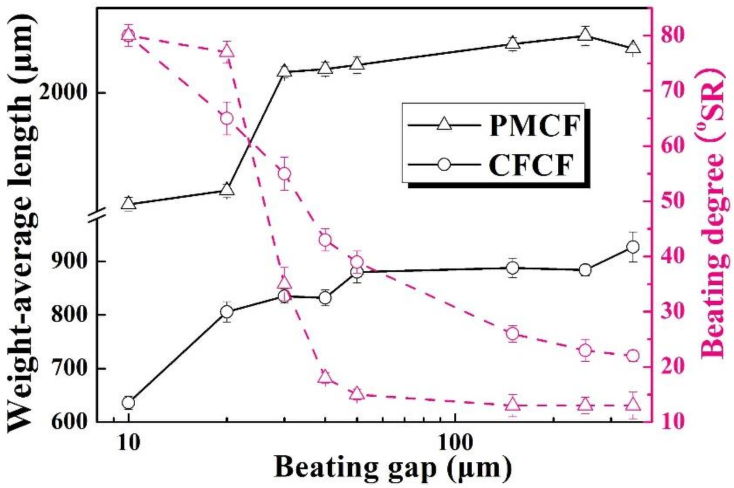

Fig. 2. Effect of various beating gaps on the beating degree and weight-average length of cellulose fibers

Table 1. Properties of *Pinus massoniana* and China Fir Cellulose Fibers Obtained with Different Beating Gaps

Beating Gap (μm)	*Pinus massoniana* Cellulose Fiber				China Fir Cellulose Fiber			
	Width (μm)	Kink index (%)	Degree of fibrillation (%)	Fiber fines (%)	Width (μm)	Kink index (%)	Degree of fibrillation (%)	Fiber fines (%)
10	29.0	32.1	1.064	7.87	26.2	23.4	1.255	26.94
20	28.6	39.6	1.158	5.38	29.8	25.8	1.263	19.19
30	29.1	34.8	0.696	6.59	29.9	23.4	1.287	17.97
40	31.0	44.7	0.472	4.30	29.0	26.9	1.445	18.69
50	31.1	43.2	0.505	2.88	29.4	26.0	1.271	15.03
150	31.2	49.4	0.342	3.37	30.8	29.3	1.174	15.68
250	31.4	49.2	0.347	2.93	30.4	23.7	1.225	14.84
350	31.8	58.5	0.331	2.94	30.7	27.0	1.249	15.81

As shown in Fig. 2, the beating degree of PMCF and CFCF sharply decreased with an increase in the beating gap, ranging from 10 μm to 50 μm. The properties of fibers were changed significantly when the beating gap became less than 30 μm. This is because the average widths of the PMCF and CFCF are approximately 31 μm and 30 μm, respectively (Table 1). A smaller beating gap increases the probability for fibers to suffer from shear force and friction force (Clark 1985; Gao *et al.* 2012). As the beating gap decreased, the fibers were more likely to suffer the function of friction force which removes part of the outermost cell wall and damages the S2 layer of the cell wall (Fig. 3a). However, the fibers are just through the disc refiner without sufficient shear force and friction force when there is a big beating gap. Therefore, the properties of the PMCF and CFCF are not greatly changed when the beating gap is more than 50 μm (Chu and Ou 2001).

The beating degree represents the susceptibility of pulp to refining. As shown in Fig. 2 and Table 1, the beating degree and the degree of fibrillation of the CFCF were higher than that of the PMCF, when the beating gap was more than 20 μm. It is easier to refine CFCF to 39ºSR (with a beating gap of 50 μm) than it is to refine PMCF to this level (with a beating gap of 30 μm), indicating the CFCF became more swollen and soft compared with PMCF. But, the beating degree of the PMCF was higher than that of the CFCF, when the beating gap was less than 20 μm. PMCF is more easily refined to 80°SR than that of CFCF. This is likely because PMCF experiences more friction force than CFCF.

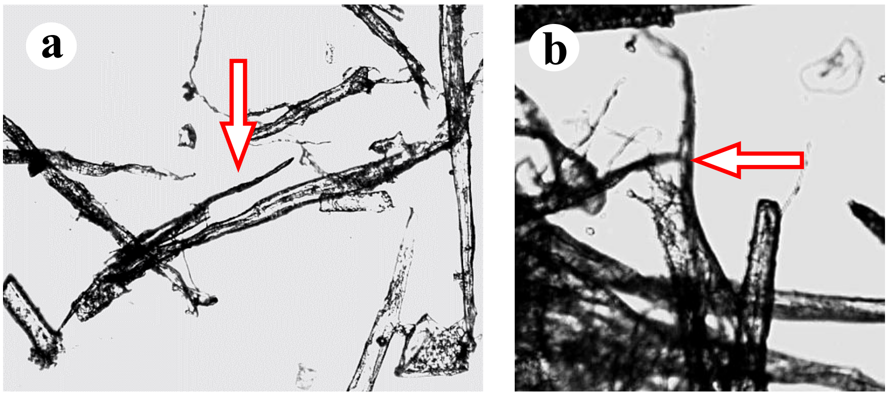

Fig. 3. The morphology of refined *Pinus massoniana* fibers

Similarly, the changes in the weight-average length of the PMCF and CFCF developed between the beating gap of 10 and 50 μm. When the beating gap was changed from 30 μm to 20 μm, the length of the PMCF decreased by 20.4%, whereas the CFCF only decreased by 3.6%. In contrast, when the beating gap changed from 20 μm to 10 μm, the length of the CFCF decreased by 26.7%, whereas the PMCF only decreased by 2.5%. This may be caused by the length and width of the cellulose fibers. The fibers with a bigger length and width are more likely to suffer adverse effects of shear force and friction force between two discs, which can cause the fibrillar structure at the end of the fiber and a fracture pattern (Molin and Daniel 2004; Chen *et al.* 2012). The fiber ends showed increased fiber fibrillation and a high level of fiber rupture (Fig. 3b).

The kink index refers to the fiber cell wall that is damaged by abrupt torsion. As shown in Table 1, the kink index of the PMCF and CFCF decreased as the beating gap decreased, whereas the fiber fines increased. This is because the probability of the PMCF and CFCF to suffer the function of shear force and friction force increased in a small beating gap. As the beating gap decreased, due to the over-cut fibers, the ratio of the fiber fines increased. And the kink index decreased because the bending of the swollen fibers improved (Lai *et al.* 2003; Liu *et al.* 2009; Dong *et al.* 2011). Consequently, the CFCF had a lower length, width, and kink index than the PMCF, except the parameters of the degree of fibrillation and fiber fines. To obtain good quality in PMCF and CFCF, a suitable and different beating gap should be considered.

Effect of Pulp Consistency on the Beating Degree and Physical Properties of Cellulose Fibers

The beating degree and the physical properties of the PMCF and CFCF obtained from various pulp consistencies with a constant beating gap of 20 μm are presented in Fig. 4 and Table 2.

Table 2. Properties of *Pinus massoniana* and China Fir Cellulose Fibers Obtained with Different Pulp Consistencies

Pulp Consistency (%)	*Pinus Massoniana* Cellulose Fiber				China Fir Cellulose Fiber			
	Width (μm)	Kink index (%)	Degree of fibrillation (%)	Fiber fines (%)	Width (μm)	Kink index (%)	Degree of fibrillation (%)	Fiber fines (%)
4	30.6	39.2	0.901	5.78	30.5	25.0	1.217	16.72
6	30.9	39.6	0.991	5.58	29.8	23.6	1.181	17.44
8	28.3	41.8	1.080	5.30	29.8	25.8	1.263	19.19
10	28.4	42.3	0.887	6.99	27.4	25.2	1.229	21.99

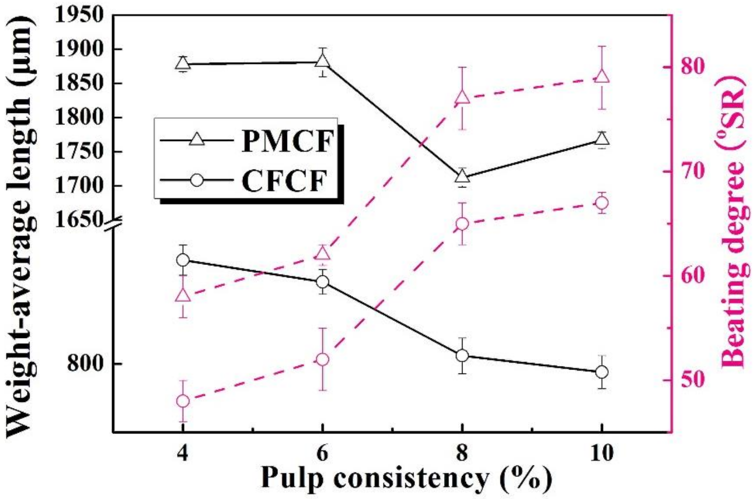

Fig. 4. Effect of various pulp consistencies on the beating degree and weight-average length of cellulose fibers

As presented in Fig. 4, the beating degrees of PMCF and CFCF increased as the pulp consistency increased, whereas the fiber weight-average length decreased. The beating degree of the CFCF with pulp consistency of 4% was higher by 39.6% than the fibers with pulp consistency of 10%, while their fiber weight-average length only decreased by 10.3%. Similarly, the beating degree of the PMCF with pulp consistency of 4% was higher by 36.2% than the fibers with pulp consistency of 10%, while their fiber weight-average length only decreased by 6.3%. This is because the high consistency refining process is mainly dependent upon the function of the friction among the fibers. It not only reduces the fiber cut-off and maintains their length, but it also improves the fibrillation of the fibers (Clark 1985; Liu *et al.* 2008; Gao *et al.* 2012).

Due to the fact that fibers have greater difficulty in passing through the discs in a higher pulp consistency, their physical properties are seriously affected. As shown in Table 2, the degree of fibrillation and the kink index of the PMCF and CFCF increased first, and then decreased with increased pulp consistency. The fiber fines content of the PMCF and CFCF increased as the pulp consistency increased (Lai *et al.* 2003; Han *et al.* 2008; Liu *et al.* 2009). CFCF had a higher level of fibrillation degree and fiber fines than PMCF, indicating CFCF is easier to be refined. Consequently, the high pulp consistency is beneficial to improve the properties of the PMCF, while the CFCF is fit for the relatively low pulp consistency.

Effect of Beating Degree on Mechanical Properties of Papers from *Pinus massoniana* and China Fir Cellulose Fibers

The different physical properties of fibers affect the mechanical properties of papers. Therefore, standard handpapers made from PMCF and CFCF were used to study the effects of refining on the properties of the PMCF and CFCF. The burst index, tensile index, and tear index of PMCFP and CFCFP, as a function of beating degrees, are presented in Fig. 5.

The tensile index and burst index of PMCFP increased with an increase of beating degree, especially within the beating degree ranging from 15 °SR to 40 °SR (Fig. 5). The tensile index and burst index of PMCFP with the beating degree of 39 °SR were increased by 588.3% and 332.4%, respectively, relative to the paper with the beating degree of 13 °SR. This is because the tensile index and burst index of papers are always affected by the combination of fibers (Gao *et al.* 2012; Yuan 2004). As the beating degree is increased, more and more hydroxyl groups are fractionated on the surface of fibers, improving the bonding ability among fibers. Additionally, the increasing ratio of small non-fiber fragments and tiny fibers fills in the paper, which can weave the fiber and reduce porosity, are good for the tensile index and burst index of papers (Zhang *et al.* 2011; Chen *et al.* 2012). On the contrary, the tear index of PMCFP decreased with an increase in the beating degree, demonstrating that excessive refining was not beneficial to the tear index of papers. Because the tear index is closely linked to the strength and length of fibers, the increasing ratio of tiny fibers and the decreasing of the fiber length leads to decreasing tear index (Wei *et al.* 2010).

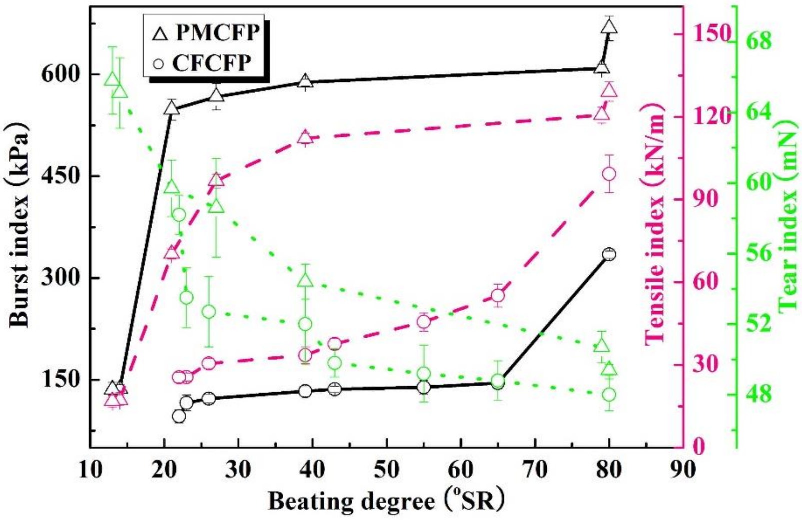

Fig. 5. Effect of various beating degrees on the properties of papers from *Pinus massoniana* and China fir cellulose fiber

The tensile index and burst index of CFCFP showed a similar trend to PMCFP, but the changes mainly developed within the beating degree range of 40 °SR to 80 °SR. The tensile index and burst index of CFCFP with the beating degree of 80 °SR increased by 164.1% and 146.1%, respectively, in comparison to paper with the beating degree of 43 °SR. The tear index of CFCFP showed a slightly decreasing trend with an increase in the beating degree. For instance, the tear index of CFCFP with the beating degree of 43 °SR was only higher by 3.8% than paper with the beating degree of 80 °SR.

Compared to the CFCFP, the PMCFP had superior strength properties. The tensile index and burst index of PMCFP were higher by 234.9% and 341.3%, respectively, than the CFCFP with the same beating degree of 39 °SR. This is mainly attributed to the longer length of the PMCF than the CFCF. In short, a suitable refining process should be considered for obtaining a good quality of papers and reducing the refining energy consumption. In this study, the CFCF is more easily refined than the PMCF. A high pulp consistency and medium beating gap is suitable for PMCF, while the CFCF should be refined on the conditions of a medium pulp consistency and a big beating gap.

CONCLUSIONS

1. In this study, the weight-average length of *Pinus massoniana* cellulose fibers (PMCF) and China fir cellulose fibers (CFCF) decreased with a decrease in the beating gap, whereas their beating degrees increased. Differently, the weight-average length of PMCF and CFCF increased with a decrease in the pulp consistency, whereas their beating degrees decreased.

2. The CFCF showed smaller weight-average length, width, and kink index than the PMCF. But, it exhibited higher degree of fibrillation and fiber fines. The CFCF more easily reached a high beating degree, indicating it is more easily refined. A suitable refining process for the PMCF is a high pulp consistency and medium beating gap, whereas for the CFCF is a medium pulp consistency and a big beating gap is recommended.
3. The tensile index and burst index of PMCFP and CFCFP increased with increasing beating degree, while the tear index decreased. And PMCFP has more superior strength properties than CFCFP. The tensile index and burst index of PMCFP with the beating degree of 39 °SR were higher by 234.9% and 341.3%, respectively, than CFCFP with the beating degree of 39 °SR.

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