Review

Basic effects of pulp refining on fiber properties—A review

Samira Gharehkhani[a,**], Emad Sadeghinezhad[a], Salim Newaz Kazi[a,*], Hooman Yarmand[a], Ahmad Badarudin[a], Mohammad Reza Safaei[b], Mohd Nashrul Mohd Zubir[a]

[a] Department of Mechanical Engineering, University of Malaya, 50603 Kuala Lumpur, Malaysia
[b] Young Resarchers and Elite Club, Mashhad, Iran

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A B S T R A C T

The requirement for high quality pulps which are widely used in paper industries has increased the demand for pulp refining (beating) process. Pulp refining is a promising approach to improve the pulp quality by changing the fiber characteristics. The diversity of research on the effect of refining on fiber properties which is due to the different pulp sources, pulp consistency and refining equipment has interested us to provide a review on the studies over the last decade. In this article, the influence of pulp refining on structural properties i.e., fibrillations, fine formation, fiber length, fiber curl, crystallinity and distribution of surface chemical compositions is reviewed. The effect of pulp refining on electrokinetic properties of fiber e.g., surface and total charges of pulps is discussed. In addition, an overview of different refining theories, refiners as well as some tests for assessing the pulp refining is presented.

Contents

Abbreviations: C, C-factor; CSF, Canadian Standard Freeness; CTMP, chemithermomechanical pulp; FSP, fiber saturation point; FTIR, Fourier transform IR spectroscopy; HC, high consistency; LC, low consistency; MFA, microfibrillar angle; ML, middle lamella; NMR, nuclear magnetic resonance; P, primary wall; SEM, scanning electron microscopy; SR, Schopper Riegler; TMP, thermomechanical pulp; ToF-SIMS, time-of-flight secondary ion mass spectrometry; XPS, X-ray photoelectron spectroscopy; XRD, X-ray diffraction.

* Corresponding author. Tel.: +60 3 7967 4582.
** Corresponding author. Tel.: +60 3 7967 4582.
E-mail addresses: s_gharahkhani_248@yahoo.com (S. Gharehkhani), salimnewaz@um.edu.my (S.N. Kazi).

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Nomenclatures

Symbols

BEL	bar edge length (km)
CEL	cutting edge length (km/rev)
C_f	pulp consistency (%)
D	depth of grooves (m)
G	width of grooves (m)
IL	impact length of the bars (m)
l_A	length of fiber (m)
n_b	number of rotor and stator bars on circle $2\pi r$ in refiner
P_{net}	net power (kW)
P_{noload}	no-load power (kW)
P_{tot}	total power (kW)
$R1$	inner radius of refining zone (m)
$R2$	outer radius of refining zone (m)
SEL	specific edge load (W s/m)
SRE	specific refining energy (KW h/t)
SSL	specific surface load (W s/m^2)
w	coarseness of fiber (mg/m)
W_r	width of rotor (m)
W_{st}	width of stator (m)
α	average intersecting angle (deg)
ρ	density (kg/m^3)
ϕ	bar angle (deg)
ω	rotational speed (rpm)

1. Introduction

Modification of the pulp or fiber quality to improve the paper features is one of the most significant scientific challenges in the paper industries. Pulp consists of cellulose fibers which come from wood and non-wood plants, and it is the major raw material in papermaking. The main sources for wood pulps are softwood (e.g., spruce, pine) and hardwood (e.g., eucalyptus, aspen) trees, and for non-wood are crops and agriculture residues (e.g., cotton, kenaf). Chemical compositions and structural properties of wood (hardwood and softwood) and non-wood species along with some remarks are presented in Table 1.

The cell wall structure (Fig. 1) of different species is generally composed of cellulose, hemicelluloses and lignin. Cellulose is a polysaccharide consisting of glucose units (Pokhrel, 2010). The cellulose molecule with several chains organized into elementary fibrils, which are the narrowest fibrils (diameter 3.5 nm). Each elementary fibril can consist of as high as 40 cellulose chains. The aggregation of elementary fibrils forms the microfibrils having diameters between 10 and 35 nm (Chinga-Carrasco, 2011; Sixta, 2008). Finally, the macrofibrils are other units which are shaped by the microfibrils aggregations (Fig. 1b) (Abe & Yano, 2009; Donaldson, 2007). Macrofibrils are twisted around the cell wall axis and introduce the term microfibrillar angle (MFA), (Fig. 1a) (Barnett & Bonham, 2004; Pulkkinen, 2010; Wathen, 2006). A smaller fibril angle is beneficial for paper strength (Blomstedt, 2007; Courchene, Peter, & Litvay, 2006). Cellulose has crystalline structure while hemicellulose has amorphous structure (definition of crystallinity is discussed in Section 4.6). Hemicellulose surrounds cellulose microfibrils. Hemicellulose has a lower strength than cellulose and can be easily hydrolyzed. It is a polymer of neutral polysaccharides present in the plant cell wall matrix and can be divided into xylans, mannans, β-glucans with mixed linkages, and xyloglucans. It can be seen in Table 1, for the cases of the hardwood and softwood, the amount of hemicelluloses are more or less same. However, there are significant differences between the types of hemicelluloses in the hardwood and softwood species. Detailed information about the above mentioned polysaccharides and amount of them in different species can be found in literatures (Ebringerova, Hromadkova, & Heinze, 2005; Sixta, 2008). The third component of the cell wall is lignin. Lignin acts as glue and binds the different layers of cell wall (Sutton, Joss, & Crossely, 2000). Lignin is a hydrophobic substance and can be removed by chemical pulping and bleaching. Low amount of lignin in the raw material makes it a good candidate for paper making. All of these components are present in the different layers of cell wall. The cell wall can be divided into different layers: middle lamella (ML), primary cell wall, secondary cell wall and lumen (Fig. 1a; Sjöström, 1993). ML acts as a cementing substance between the cells and has highest concentration of lignin. (Shafiei Sabet, 2013). The middle ML surrounds the primary wall (P). P layer is thin and flexible. It consists mainly of hemicelluloses and lignin and a loose aggregation of microfibrils which are oriented randomly in this layer. Cellulose chains are twisted along the axis of glucan chains and are held by hydrogen bonds between the chains (Sixta, 2008; Thomas et al., 2013). Secondary wall is located between the P and lumen. It sometimes consists of three distinct layers: S1, S2 and S3 (Bergander & Salmén, 2002; Meier, 1962). Most of fiber mass belongs to the S2 layer, the MFA in the S2 is 10–30° while the S1 layer has a high microfibril angle (50–70°). The S3 layer is thin with fairly horizontal microfibrils (MFA is 70–90°) (Blomstedt, 2007). The last layer in the cell wall is lumen (w) which is the hollow core and can hold the moisture (water or water vapor). Fibres with large lumen tend to flatten to ribons during pulping which result in good strength properties.

To produce the pulp from the raw materials, different pulping processes can be performed on the chips or small parts which have been produced by the chipping of timber or other parts of plant. Depending on the pulping processes the wood pulps are categorized as mechanical pulp, chemical pulp (e.g., kraft, sulfite pulps) and chemithermomechanical pulp (CTMP). Mechanical pulps are produced from raw material by the application of mechanical energy. The mechanical pulps have good print quality. Thermomechanical pulps (TMP) are one of the popular types of mechanical pulps which are produced by processing the wood chips using the high temperature steam and mechanical refining. Chemical pulps are almost pure celluloses which are produced by heat and chemical treatment. During the treatment of raw material with chemicals, a large amount of lignin is extracted from the material. The chemical pulps can be classified to kraft and sulfite pulps. Kraft pulp or sulfate pulp, is obtained by the treatment of the chips with a mixture of sodium hydroxide and sodium sulfide and sulfite pulp is formed during the pulping process by using the various salts of sulfurous acid. CTMPs are produced by the combination of chemical and mechanical treatments. They need less mechanical energy,

Table 1
Chemical composition and structural properties of typical wood and non-wood species.

Properties	Wood		Non-wood			Remark
	Softwood	Hardwood	Wheat straw	Corn stalks	Bagasse	
Chemical composition						
Cellulose	40–45[a]	43–47[a]	30[a]	35[a]	40[a]	Fiber length is approximately 3 mm for softwood and 1 mm for hardwood. Most of the softwoods are softer than hardwoods. Softwood fibers are more flexible than hardwood fibers. In the case of non-wood, the average ratio of fiber length ranges from 1 mm to 30 mm. It depends on plant species and the plant part from which the fiber is derived. Non-woods have lower lignin content (compared to wood), so can be pulped in less time compared to woods.
Hemicellulose	25–29[a]	25–35[a]	50[a]	25[a]	30[a]	
Lignin	25–31[a]	16–24[a]	15[a]	35[a]	20[a]	
Extract	1–5[a]	2–8[a]	5[a]	5[a]	10[a]	
Structural properties						
Slenderness ratio (fiber length/fiber diameter)	95–12[b]	55–75[b]	86.76[c]	54.32[d]	70[e]	
Flexibility ratio (fiber lumen diameter/fiber diameter)×100	75[b]	55–70[b]	71.76[c]	44.03[d]	29[e]	

Data have been presented by [a] (Sixta, 2008), [b] (Smook, 1992), [c] (Singh, Dutt, & Tyagi, 2010), [d] (Usta, Kirci, & Eroglu, 1990) and [e] (Agnihotri, Dutt, & Tyagi, 2010).

and the chemical treatment is performed with lower temperature and shorter time. The CTMPs have good strength. Among different types of pulps, the chemical pulps have the highest share in pulp productions. For instance, in Europe, kraft pulps account for 65% of total pulp production (Abdul Khalil, Bhat, & Ireana Yusra, 2012).

A variety in pulp sources have made a demand for having a fundamental process for improvement of the fiber quality that can be applied on all types of pulps. One of the processes that is conducted in the stock preparation is so-called “pulp refining” or “beating”. Pulp refining or beating could be described as a mechanical treatment of the pulp by using the special equipment (refiner). Nowadays the term “refining” is more common than “beating” in the studies because of the appearance of different refiners which are widely used in the pulp industries. However both phrases have same meaning. The term “refining” is adopted in this study to keep the consistency in the text.

In the refining process, the fibers are under compression and shear forces which are causing several changes in specifications of fibers. Dependent on the initial fiber properties, pulp consistency (weight in grams of oven-dry fiber in 100 g of pulp–water mixture) and refiner specification, the changes in fiber result in higher bonding (Mohlin, Miller, Mohlin, & Miller, 1995).

This review focuses on the effects of pulp refining on the structural and electrokinetic properties of the fibers which are critical in

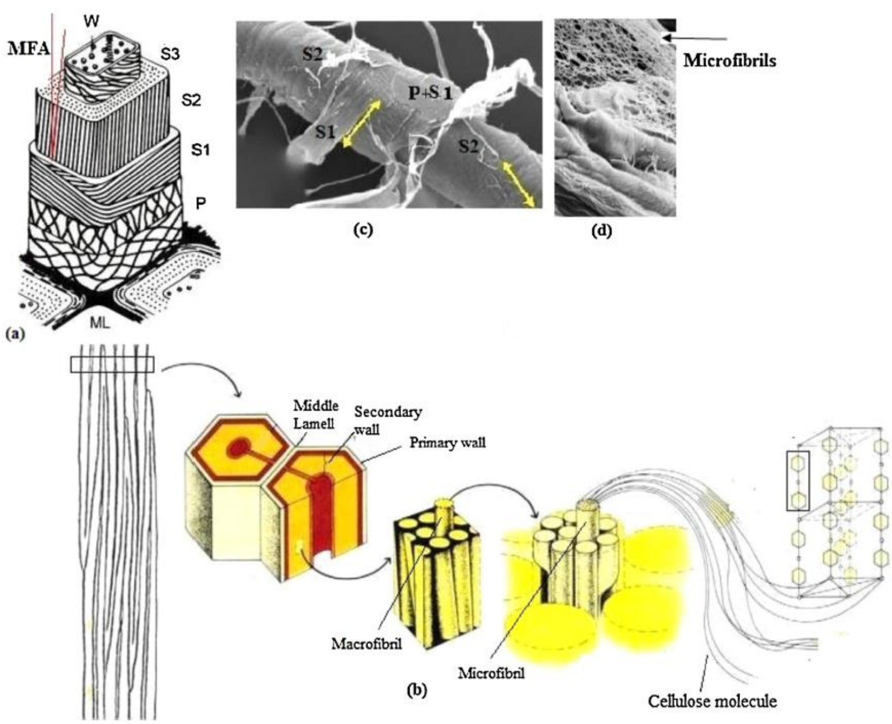

Fig. 1. Fibre unit structure: (a) schematic of cell wall layers (middle lamella (ML), primary wall (P), secondry wal (S1, S2, S3) and lumen (W); (b) fibrillar structure of cell wall; (c) scanning microscopic image of cell wall layers; and (d) scanning microscopic image of microfibrils (Chinga-Carrasco, 2011; Fardim, Liebert & Heinze, 2013; Fellers & Norman, 1998; Page, 1989a,b; Sixta, 2008).

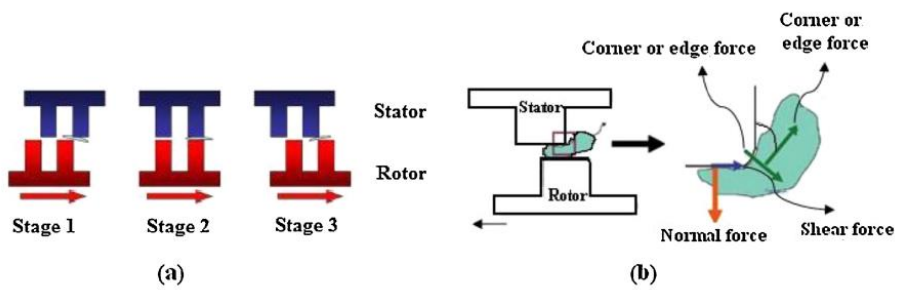

Fig. 2. (a) Refining mechanism (Nugroho, 2012). (b) Forces acting during refining (Cuberos-Martinez & Park, 2012).

the paper industries. In addition, a summary of different theories, refiners (laboratory and industrial refiners) and also measurement methods suggested for monitoring the refining are presented. The effects of refining on paper properties are out of scope of the present study, but it has been referred where it is related to the fiber properties.

2. Theories of refining

This section is presented to provide a common basis of the refining theories. The goal of refining theory is to predict changes in pulp properties from known refining conditions. Prior to discuss about theories, mechanism of refining is explained in this section.

In the refiners, fibers are treated between two parallel grooved plates; stator and rotor. The three dominant refining stages are shown in Fig. 2(a). The first one is pick-up stage fibers are accumulated and trapped between the edges of bars. In the next step, the trapped fibers are compressed by the surfaces of the moving and stationary bars. In the final stage, the fibers are affected by the shear forces. In this stage the fibers also hit the bars on the surface to edge and again edge to edge. During the bar crossing, two different forces act on the fibers, one of them due to the contact fibers to bars and another one due to the contact fiber to fiber (Lumiainen, 2000; Nugroho, 2012; Technology, 2001).

2.1. Specific edge load (SEL) theory

The SEL theory is a well known and a most reliable theory in papermaking industries (Genco, 1999; Karlström, Eriksson, Sikter, & Gustavsson, 2008; Kerekes, 2011; Mohlin, 2002; Olejnik, 2013). The SEL is the amount of effective energy spent per unit edge length of bar crossing and rigorously is presented as "machine intensity", meaning it represents the energy expenditure at bar crossings without reference about the pattern of energy distribution (Kerekes & Senger, 2006; Kerekes, 2011). SEL can be obtained from Eq. (1): (W s/m or J/m)

$$\mathrm{SEL} = \frac{P_{\mathrm{net}}}{\mathrm{CEL}}, \quad P_{\mathrm{net}} = P_{\mathrm{tot}} - P_{\mathrm{noload}} \tag{1}$$

where P_{tot} is the total power consumed, P_{net} is the net power (kW) consumed to change the pulp properties. P_{noload} is the initial power or no-load power which is defined as the power needed to rotate the rotor in the refiner. It is noticeable that this power has no contribution in pulp refining, and it is only consumed to overcome the losses, including loss due to shaft and bearings friction (mechanical losses), required energy to rotate the refiner plate in pulp suspension close to the stationary plate (hydraulic) and required energy consumed by refiner when pumping pulp suspension from inlet to outlet of refiner (pumping). Different studies have been reported to explore the no-load power in LC refining both experimentally and computationally (Dietemann & Roux, 2005; Rajabi Nasab, 2013).

As can be seen in Eq. (1) another parameter contributed in the SEL is cutting edge length (CEL) which is calculated by Eq. (2).

$$\mathrm{CEL} = \mathrm{BEL} \times \omega \tag{2}$$

where BEL is bar edge length and ω is rotational speed. Plate designs are typically characterized by BEL and BEL/CEL is given to paper makers by suppliers of refiner plates.

Typical SEL for softwood and hardwood is 1.5–3 J/m and 0.2–1.0 J/m respectively (Nugroho, 2012). The SEL theory works quite well with coarse fillings when bars are wider than the length of the fiber flocs (Lumiainen, 2000).

One parameter that contributes in the study of different theories is specific refining energy (SRE). SRE is used for calculating how much energy is given from the refiner to the fibers, and can be obtained by dividing the net power by the fiber mass flow rate. Typical SRE for chemical pulps is between 80 and 250 Kw h/t (Heymer, 2009; Loijas, 2010). The higher the SRE, the more refining is happening to the fibers. Nugroho (2012) found that at constant refiner speed, flow rate, and pulp consistency, the specific refining energy and refining intensity are influenced by net power only, so it makes a linear correlation between specific refining energy and refining intensity. In this case, the specific refining energy is proportional to refining intensity.

2.2. Specific surface load (SSL) theory

However SEL is widely used theory in industries, it is quite simple. As an example, in this theory, the width of bars has not been considered. To eliminate this lacking, Lumiainen (1990) proposed the SSL theory which consider the impact length of the bars (IL) in the SEL theory (Loijas, 2010). SSL (W s/m^2 or J/m^2) is calculated by Eq. (3).

$$\mathrm{SSL} = \frac{\mathrm{SEL}}{\mathrm{IL}} \tag{3}$$

IL can be calculated from the width of rotor (W_{r}) and stator bars (W_{st}), and from the average intersecting angle α of the rotor and stator bars.

$$\mathrm{IL} = \frac{W_{\mathrm{r}} + W_{\mathrm{st}}}{2} \times \frac{1}{\cos(\alpha/2)} \tag{4}$$

The SSL theory seems to work quite well when bars are so narrow. If bars are much narrower than the fiber floc, then they heavily cut the fibers (Lumiainen, 2000).

2.3. C-factor theory

Several scientists have described the SRE into two parameters which are the number of impacts (N), and the intensity of each impact (I), on pulp (Kerekes, 1990; Lewis & Danforth, 1962; Stevens, 1981). To provide a relationship among the net power and fiber mass flow rate with N and I, Kerekes (1990) has proposed the C-factor theory (Eq. (5)).

$$\mathrm{SRE} = \frac{P_{\mathrm{net}}}{\dot{m}} = N.I = \frac{C}{\dot{m}}\frac{P_{\mathrm{net}}}{C} \tag{5}$$

where C is described as the capacity of a refiner to inflict impacts on fibers. C has been given for different refiners (disk, conical and PFI mil) as a function of fiber length, coarseness (mass per unit length), plate geometry, rotation speed and pulp consistency (Heymer, 2009; Kerekes, 1990; Kerekes, Soszynski, & Doo, 2005). For example C for a disk refiner is calculated by Eq. (6):

$$C = \frac{8\pi^2 GD\rho C_F l_A n_b^3 \omega(1 + 2\tan\phi)(R_2^3 - R_1^3)}{3w(l_A + D)} \tag{6}$$

variables has been defined in the nomenclature.

Although, C is a more precise theory than SLE and SSL, it is not yet a widely used theory in the papermaking. One important factor that makes some difficulties in use of C especially in the industries, is the measurement of the initial pulp dimensions (length and coarseness). Another factor, refers to the basic effects of refining, as during refining, the length of the fiber is subjected to the change, so the C is not constant (Batchelor, Kjell-Arve, & Ouellet, 1999; Olejnik, 2013).

2.4. New concept in refining theories:

By reviewing the above mentioned theories, (SEL, SSI and C) it is found that all of them are energy-based intensities and energy is the commonly used variable in the paper mills. However, it is stated that force and not energy is responsible to change the fiber properties during refining. In the new concept, the attempts are focused to use force in characterizing refining action (Kerekes, 2010; Kerekes & Senger, 2006). To understand such theories, detailed knowledge of the applied forces, the fiber distribution in refiner gaps, and the amount of fibers, is needed (Heymer, 2009). In the pulp refining, the typical gap is 0.1–0.2 mm which is as big as the thickness of several single fibers. Therefore fibers are trapped between the stator and rotor bars as the flocs and the forces act on the flocs rather than single fibers (Andersson, 2011; Batchelor, Lundin, & Fardim, 2006). The forces which are transmitted to the flocs are: normal force, shear force and corner or edge force (Fig. 2(b)). The normal force is due to the compression of the floc between bars, shear force is produced by the movement of bar surface on the floc and corner force appears at the bar edge (Batchelor, Martinez, Kerekes, & Ouellet, 1997; Martinez & Kerekes, 1994; Roux, Bloch, & Nortier, 2007). To estimate the amount of flocs trapped between bars, Batchelor et al. (2006) conducted some experiments. He proposed that the number of fibers trapped under any segment of bar will determine the point at which the net refining power begins to rise and the net refining power is fitted with a negative exponential function.

Kerekes (2011) proposed a force based theory that the refining intensity is based on forces on bars which are linked to the forces on fibers through fiber distribution over bars and gap size. It should be noted that in Kerekes theory, the forces are predicted on the individual fibers. To develop such concept, more studies are needed.

3. Summary of refiners and control tests used in the stock preparation

In order to aid the future discussions, the present section provides an overview of different types of refiners, and tests applied on pulps for the stock preparation.

3.1. Different pulp refining machines

Refiners can be categorized into laboratory and industrial refines. Table 2 provides a summary of refiners along with the quantitative information about the different refiners which have been used in published research works since 2000, based upon the search in the Scopus website.

3.1.1. Laboratory refiners

In the laboratory scale, PFI Mill, Valley Beater, Jokro mill and pilot-refiners are used for refining the small pulp samples where the mechanisms of refining are different.

3.1.1.1. PFI mill. As illustrated in Table 2, the PFI mill is the most commonly used laboratory refiner. It is a high energy and low intensity refining device (Kerekes et al., 2005). Using this refiner, the pulps are refining between a stainless steel roll with bars and a rotating disk with a smooth bed where pulps are distributed over the disk wall uniformly. Both elements rotate in the same direction. However they have different speed. The refining specification is reported either in "revolution", or "PFI count" which is 10 times of revolution.

3.1.1.2. Valley beater. The Valley beater is another refiner that has a different shape in comparison to PFI mill, which consists of the roll bars and bedplate. Comparing to the PFI mill, the bigger amount of samples and longer time for refining is needed in the Valley beater (Yasumura, DAlmeida, & Park, 2012). The valley beater increases the fiber cutting and the fine formation, whereas PFI mill tends to increase the internal fibrillation and swelling, comparatively (Garcia et al., 2002; Jones, Venditti, Park, Jameel, & Koo, 2013; Park, Venditti, Jameel, & Pawlak, 2006).

After each run in the refining devices, the refining degree is assessed by the Canadian Standard Freeness test (CSF). Detail of the test is incorporated in this section. Regarding the calibration requirements, it is recommended for the valley beater that the refining should not continue when pulp reach at 250 CSF.

3.1.1.3. Jokro mill. The Jokro mill is not very common to the researchers in comparison to other laboratory devices. Jokro mill same as PFI mill consists of a refining roll with several bars within a container, where the groove dimensions in Jokor mill are smaller than PFI mill, especially in groove depth. The time is regulated for controlling the refining.

3.1.1.4. Small-scale pilot refiner. The small-scale pilot refiners are used to simulate the industrial scale refining. They have different types of geometries same as the industrial refiners. Details of different geometries are presented in Section 3.1.2.

3.1.2. Industrial refiners

The refiners with different types of geometries such as disk, conical and cylindrical are used in the industry. They consist of rotor and stator, which could have various patterns of plates with different grooves and bar dimensions. Nowadays, the manufacturers are proposing new refiners by considering the industrial demands.

One of the key objectives in developing the new refiners is the reduction of energy consumption. However, many suppliers have stated that their refiner can lead to more energy savings, this claim needs further investigation to know how much energy could be saved in comparison to the existing ways such as using of one large refiner instead of numerous small refiners.

Another parameter is homogeneity. Reduction in the amount of untreated fibers is an important challenge in fiber refining. Although different concepts such as multiple disk refining, low consistency (LC) and stable gap refining could slightly overcome this

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