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Pilot-scale soda pulping of wheat straw using continuous pulp digester

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A pilot-scale three tube continuous pulping digester was designed and fabricated to convert wheat straw into pulp for paper-making using the soda pulping delignification process. The physical and chemical properties of wheat straw were investigated to characterize wheat straw. The bulk density, porosity, moisture content, and fiber diameter were observed as 0.18 g/cm^3, 65.30%, 2.01% (w/w), 15–20 µm, respectively. Chemically, the wheat straw is contained 40% cellulose, 68.1% holo-cellulose, 18.30% lignin and 8.85% ash (w/w). The influence of various process variables on delignification process was studied at 117–159°C and 0.5–5.88 kg$_f$/cm^2. The white liquor concentration, white liquor flow rate, and screw feeder speed (RPM) were considered as process variables. The obtained pulp was analyzed in terms of kappa number and residual alkali as a measure of pulp quality and extent of delignification. The minimum value of kappa number and residual alkali were obtained as 19.58 and 54 g/L. It was observed that the high concentration and high white liquor flow rate results in a high value of residual alkali. The higher pressure and temperature inside the digester tubes were found suitable for better delignification operation.

Keywords: Wheat straw, white liquor, soda pulping, delignification, continuous pulp digester.

Introduction

The fibers obtained from plant origin (cellulose) are commonly used to manufacture the product like paper, pulp, cardboard, building material, and furniture to facilitate our daily life. The cellulose-rich plant based raw materials are the major source of fibers[1,2]. It includes soft/hard wood, agriculture waste, bagasse, grasses, molasses, etc. Traditionally, the most of the paper industries are using plant stem (wood) as a source of cellulose fibers[1,2]. Hence, there is a huge demand for wood (softwood or hardwood) to meet the global requirement of the paper. Presently, almost ~69% of the total paper is producing from recycle and argo-waste[1,2]. In India, the projected demand for paper is increasing continuously with a compound annual growth rate of 6.7% per annum[2]. The high amount of raw material for paper manufacturing is majorly fulfilling from the forest wood. Conversely, the forests are sinking due to increase in population, civilization, technological development, and many more. The continuous depletion of the forests is also affecting our environment and leading to global warming and other serious issues. So, it is necessary to see the alternative raw material for paper production and other cellulose products[3]. The alternate source may be bagasse, grasses, bast fibers, cereal straws, canes, molasses, etc. The straws are the most abundant agricultural by-product obtained worldwide after the cultivation of barley, oat, rice, rye, and wheat and similar crops[3]. Wheat is one of the popular crops grown all over the world. In India, wheat production covers almost 25% of the total crop production and it is the subsequent largest wheat producer after China[4]. Hence, an abundant quantity of wheat straw is available in India and it is a potential alternative cellulose-rich raw material for paper manufacturing due to its low lignin content[4].

In India, there are near about 850 units for paper, newsprint and cardboard production and the total installed capacity for paper production is 24 million tons. The 850 units for pulp production used wood, recycled paper, and agricultural residue as raw material with a production share of 20%, 70%, and 10% respectively[3–5].

The pulp/paper is produced by various mechanical, chemical, and combinations of both mechanical and chemical processes. The mechanical pulping process separates the cellulose fiber using mechanical energy by breaking the bonds between fibers bundle[6]. The lignin content is maintained in the pulp at such a level so that the strength and brightness of the paper do not affect. Lignin is a natural cellulose binder and polymeric material with high molecular weight and viscosity. The separation of cellulose fibers from the natural binder lignin is known as delignification/cooking[7]. The high residual lignin in the paper is not desirable in good quality paper. The lignin residue makes the product yellowish and this yellowness increases with time[6–8].

Chemical pulping processes are another pulping process for paper production and it is the most favorable commercial process used all over the world. In chemical pulping, lignin is removed from cellulose fiber using different chemical solutions. The chemical pulping is further classified in kraft, sulfite, and soda pulping. It has low yield as compared to mechanical pulping due to the chemical degradation of the cellulose and hemicellulose fibers in the presence of pulping solution[6,7]. Among the available chemical pulping process, the kraft pulping process provides a good quality of paper in terms of strength, fiber length, etc. A solution of sodium hydroxide and sodium sulfide (white liquor) is used in the kraft pulping process. In kraft pulping, at high temperature (155–175°C) and pressure (5–7 bar), the cellulose-rich raw material reacts with the white liquor and delignification process take place. It removes the lignin by breaking the bond between cellulose and lignin[6,7]. The sulfidity of white liquor is used to maintain the viscosity of cellulose mixture during cooking process[6–8]. In the soda process, sodium hydroxide is used as the main constituent of white liquor. Almost, 80% of the lignin, 10% of cellulose and 50% of the hemicellulose are dissolved in the white liquor by the soda process[6–8].

The main driving force for the pulping is the concentration difference of white liquor at elevated temperature and pressure. Every 10°C increase in the temperature doubles the reaction rate within the temperature range of 155–175°C. During the pulping process, a little amount of cellulose fibers is degraded and it became significant beyond 180°C. Hence, beyond 180°C, the pulp quality and yield affected[6–8]. In industry, there are some compounds that are also used as pulping aids for effective pulping e.g. dimethyl formamide[9], anthraquinone[10].

Soda pulping is the alternate process to produce sulfur-free pulp with low pollution than kraft pulping[10]. The researchers are trying to shift on the soda pulping process from kraft pulping to reduce environment load[11]. A significant amount of work is available in the literature on soda pulping[6–11]. The soda pulping of bagasse[12,13], lemongrass[14], and citronella grass[15], citrus waste[16] are performed recently as alternate cellulose sources[17].

Specifically, soda pulping of wheat straw was studied[8] at effective alkali concentrations of 12–18% at the operating temperature of 145–165°C and shown the significant effect of these parameters on the viscosity of wheat pulp[10,11]. It is also concluded that the viscosity of wheat pulp increases with a decrease in the value of kappa number (degree of delignification)[11]. In another study of Montane *et al.*[18], the two-stage cooking sequence was performed involving the steam impregnation of wheat straw with white liquor and stream treatment of impregnated material at 160–210°C. The presented impregnation method/process of wheat straw pulping took less time than the traditional pulping method with comparable pulp properties[18].

In the present work, soda pulping of wheat straw at different operating conditions has been performed in three tube continuous pulping digester at the pilot-scale. The alkali concentrations, white liquor flow rates, wheat straw screw RPM, temperature and pressure have been considered as process variables for the pulping operation. The physical and chemical properties of wheat straw were estimated to characterize the wheat straw. The product quality of obtained pulp has been analyzed as kappa number (extent of delignification), residual alkali, total solids and viscosity of the black liquor.

Experimental procedure

Soda pulping is the most acceptable/favored process used in the pulping of agriculture residue/waste due to its superior pulp quality. Wheat straw is being selected as a potential source that is under-utilized in the paper industry. Although, it is the most suitable raw material for good quality pulp/paper manufacturing due to its low lignin content.

Material:

Wheat straw and white liquor are the main constituents of the soda pulping. The dried fresh wheat straw is collected from the fields of Punjab and Haryana. The physical and chemical properties of the wheat straw were estimated and tabulated in Tables 1 and 2. These physicochemical analyses are including the determination of ash content of wheat straw[19], acid-insoluble lignin, acid-soluble lignin[20], cellulose percentage[21] and holo-cellulose[21] (Tables 1 and 2).

Table 1. Physical properties of wheat straw

Bulk density (g cm^{-3})	0.18
Moisture content (%)	2.01
Porosity (%)	65.30
Fiber length (μm)	30–40
Fiber diameter (μm)	15–20

Table 2. Chemical composition of wheat straw

Ash content (%)	8.85
Cellulose (%)	40.00
Holocellulose (%)	68.10
Acid soluble lignin (%)	1.30
Acid insoluble lignin (%)	17.00
Total lignin (%)	18.30

Another main constituent is the solution of sodium hydroxide at different concentrations and known as is white liquor[22]. The sodium hydroxide (NaOH) concentration used in each experiment are given in Table 3. The commercial-grade sodium hydroxide (94% pure) is used for the preparation of white liquor.

Table 3. Operating conditions for laboratory scale soda pulping

Set no.	Exp no.	NaOH (%)	W. L. flow rate (L/min)	RPM	Temp. (°C)	Pressure (kg$_f$/cm^2)
S_1	R_1	16.7	1	24	148	3.5
	R2	16.7	1.24	24	135	2.2
S_2	R_3	13	1.34	24	159	5.80
	R_4	13	1.34	30.9	159	5.8
S_3	R_5	9.2	1.36	34	117	1
	R_6	9.2	1.37	39.4	123	1.5
	R_7	9.2	1.71	39.4	123	1.5

Pilot-scale pulp digester:

The pilot-scale three tube continuous pulping digester was designed and fabricated for the pulping of the wheat straw. The design volume of the digester is 20 L with a total length of 3 m (1 m each tube). The three digester tubes are mounted horizontally and connected in such a manner that the digester slurry passes from one tube to another digester tube as shown

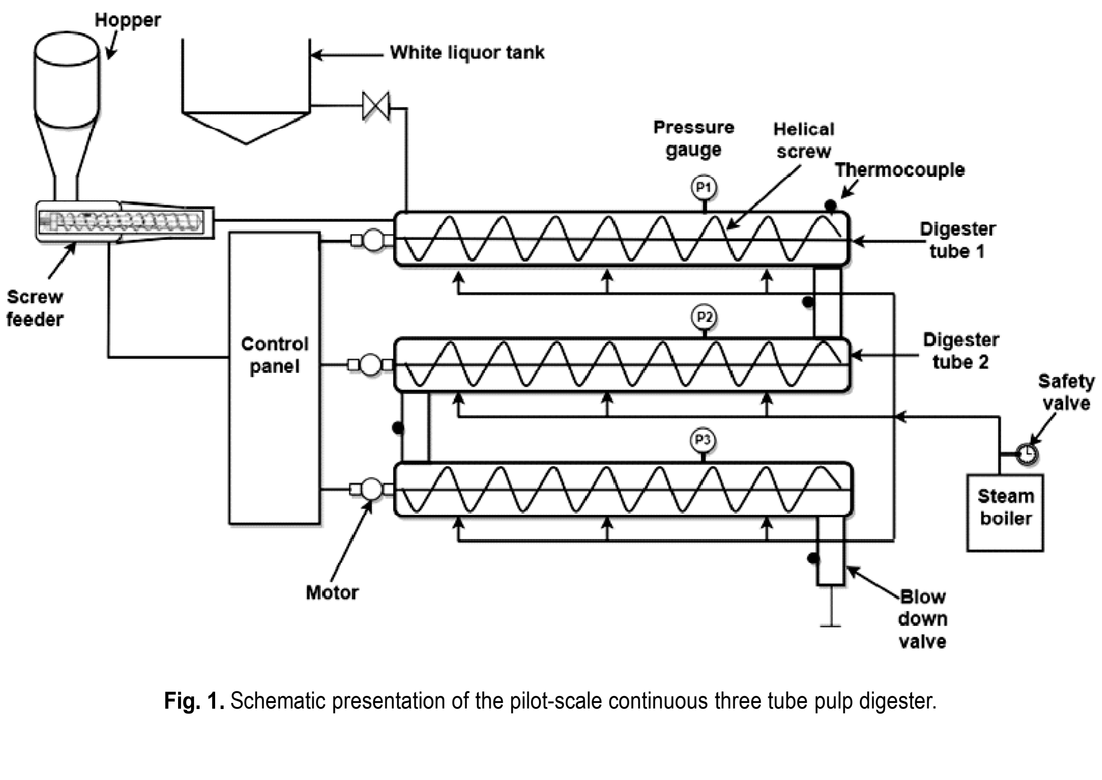

Fig. 1. Schematic presentation of the pilot-scale continuous three tube pulp digester.

in Fig. 1. The helical screw conveyor is fitted inside each digester tube to facilitate the unrestricted movement of the pulping mixture from inlet to outlet of the digester. The screw conveyors are rotated with the help of three-phase induction motor fitted with a speed controller through the control panel. The washed wheat straw is fed at the inlet of the first digester tube through the screw feeder as shown in Fig. 1. The feed rate of wheat straw is controlled with the help of screw feeder RPM. The other pulping constituent, white liquor is stored in a storage tank fitted with a stirrer to maintain the consistency of the mixture. The white liquor was fed to the first digester tube using a plunger pump by the separate inlet line near to the wheat straw screw feeder. The flow rate of white liquor is controlled with the help of a flow controller. Thermocouples and pressure gauges are being mounted in each tube to measure the temperature and pressure respectively. The high-pressure safety valves are installed on each tube of the digester to ensure the safety of the equipment. The whole set up was operated and controlled through the controlled panel. The schematic diagram of the digester is shown in Fig. 1.

Process description:

The pulping process refers to the delignification of cellulose-rich raw material. In the delignification process, the corrosive white liquor is used to dissolve the lignin content (natural cellulose binder) of raw material[22–25]. The washed and dried wheat straw was kept in a water tank for 48 h for the soaking and softening of biomass. The softened wheat straw is fed to the digester inlet through the wheat straw screw feeder. The process steam is generated in an electrically heated steam boiler and supplied at various locations of the digester tubes to maintain the desired temperature and pressure. The operating conditions are tabulated in Table 3.

The wheat straw was fed to the digester at 110°C at a constant rate. The wheat straw and white liquor mixture moves horizontally in tubes with the help of the screw conveyer and reached at the inlet of the second digester tube. Then the mixture moves from the second digester tube to the third digester tube. The delignification process[25] starts in the first digester tube and continues during the movement of the mixture inside the digester. At the end of the third digester tube, the pulp with black liquor is obtained from the blowdown valve fitted at the outlet of the third digester tube.

Results and discussion

The physical and chemical properties of the wheat straw are depending on the wheat crop species and its cultivation conditions. The obtained pulp quality and fiber size are greatly depending on the physical and chemical properties of the wheat straw. Additionally, higher fiber dimensions are always desirable for better quality pulp and paper production. The fiber dimensions are also important to classify the raw material/pulp for paper production. Hence, the estimation of physiochemical properties of the wheat straw and fiber dimensions has been performed. The physical and chemical properties of wheat straw are presented in Tables 1 and 2 respectively. The bulk density, moisture content, and porosity of the wheat straw were obtained as 180 kg/m^3, 2.01% (w/w) and 65.30% respectively (Table 1).

For the estimation of fiber dimensions, the SEM investigation of wheat straw fiber has been performed. The SEM images shown in Fig. 2 of the powdered wheat straw showed that the length of wheat straw fibers is ~30–40 μm and the average diameter of the fiber is ~15–20 μm (Fig. 2). Chemically, the fresh wheat straw was contained 8.85% ash, 40% cellulose, 68.10% holo-cellulose and 18.30% lignin (Table 2). The obtain physical and chemical properties are consistent with the literature values[18,21,23].

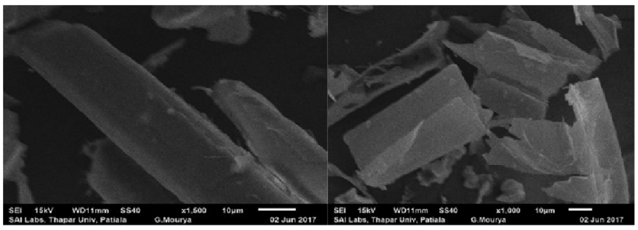

Fig. 2. SEM images for powdered wheat straw at 10 μm.

A series of soda pulping experiments were divided in 3 sets of experiments based upon NaOH concentration in white liquor as 16.7%, 13%, and 9.2. The white liquor flow rate is varied in the range of 1 to1.71 L/min as shown in Table 3. The RPM of wheat straw feed screw is varied from 24 to 39.4 to obtain effective feed for pulping and the white liquor flow rate and/or concentration were varied accordingly. The obtained pulp is investigated qualitatively for kappa number, residual alkali, total solids in black liquor and viscosity of black liquor. The obtained results are summarized in Table 4.

Table 4. Results of soda pulping

Set No.	Exp no.	Kappa no.	Residual alkali (g/L)	Total solids (g/L)	Viscosity of black liquor (cP)
S_1	R_1	33.12	21.82	82	1.283
	R_2	52.65	21	106	3.099
S_2	R_3	19.58	33	54	2.039
	R_4	39.71	45	68	1.873
S_3	R_5	27.16	25	56	2.105
	R_6	23.18	42	61	2.119
	R_7	28	38	58	2.109

The degree of delignification is usually estimated in terms of kappa number. The kappa number is obtained as the volume of 0.1 *N* potassium permanganate consumed by 1 g of moisture-free pulp[26].

In the first set of experiment S_1, two experiments (R_1-R_2) have been performed. In these experiments, the NaOH concentration was maintained at 16.7% and the white liquor flow rate was varied from 1–1.24 L/min. In the first experiment R_1, the temperature and pressure inside the digester were maintained at 147°C and 3.5 kg_f/cm^2 respectively. The white liquor flow rate was maintained at 1 L/min. The value of kappa number was estimated 33.1 and the residual alkali was 21.8 g/L. The other parameters like total solids in black liquor and viscosity of black liquor were observed as 173 g/L and 1.323 cP respectively. It is always desirable that the amount of residual alkali should be as small as possible. The 21.8 g/L is a higher value of residual alkali which shows the excess amount of NaOH in white liquor or ineffective delignification at low temperature and pressure.

In the second experiment R_2, the NaOH concentration was maintained the same as in R_1 i.e. 16.7% and the white liquor flow rate was increased from 1 to 1.24 L/min by keeping the feed screw speed constant i.e. 24 RPM. The temperature and pressure were maintained at 135°C and 2.2 kg_f/cm^2 respectively. The estimated kappa number of obtained pulp was 52.65, which was very high in comparison to the previous experiments R_1. The value of residual alkali, total solids, and viscosity, in this case, was 21.8 g/L, 106 g/L, and 3.09 cP respectively. It shows a further low degree of delignification and most of the material collected in a blow tank was unreacted. The possible reasons are low temperature and pressure, at which the delignification process was not completed.

Then, in the second set S_2, two experiments (R_3-R_4) were performed. In these experiments, the NaOH concentration was decreased and maintained at 13% with a white liquor flow rate of 1.34 L/min. The feed screw speed was varied as 24 and 30.9 RPM. Particularly in experiment R_3, the temperature and pressure were increased and maintained as 159°C and 5.8 kg_f/cm^2 respectively (as given in Table 3). The obtained output pulp was analyzed and the value of kappa number was estimated as 19.5. The value of residual alkali, total solids and viscosity are found as 33 g/L, 54 g/L, and 2.039 cP respectively. The lower value of kappa number indicated the high degree of delignification as compared to the experiments R_1 and R_2. This means effective delignification takes place but the higher residual alkali was obtained as 33 g/L than experiments R_1 and R_2. The possible reason behind the higher value of residual alkali may be the higher white liquor flow rate than the required one. It also shows that the delignification process enhances with increased pressure and temperature. The cellulose fiber gets broken at a pressure greater than 7 kg_f/cm^2.

Further, in the case of experiment R_4, alkali concentration and white liquor flow rate were kept constant as in the previous experiment i.e. 13% and 1.34 L/min respectively. RPM of wheat straw screw feeder was increased from 24 to 30.9 (Table 4). The pressure and temperature were maintained as 5.8 kg_f/cm^2 and 159°C respectively. The kappa number was obtained 39.7 and residual alkali value was measured 45 g/L. The total solids in black liquor and viscosity of black liquor determined were 68 g/L and 1.87 cP respectively. The value of kappa number and residual alkali are again higher than the previous experiments, which may be due to uneven raw material flow rates. This shows that an increase in RPM to 30.9 and white liquor flow rate to 1.344 L/min is ineffective in delignification process.

In the third set S_3, three experiments (R_5-R_7) were performed. In these experiments, alkali concentration was maintained at 9.2% and the flow rate was varied from 1.37 to 1.71 L/min. In experiment R_5, the white liquor flow rate was maintained at 1.36 L/min and the wheat straw screw feeder speed was kept at 34 RPM. The observed pressure was 1 kg_f/cm^2 and the achieved temperature is 117°C. The value of kappa number obtained in this case was found as 27.16 and residual alkali was 25.2 g/L. A sharp decrease in kappa number from 39.7 to 27.16 from experiment R_4 to R_5 was observed even at low temperature and pressure.

Further, in experiment R_6, the white liquor flow rate and white liquor alkali concentration were kept constant at 1.37 L/min and 9.2% respectively. The wheat straw screw speed was increased from 34 to 39.4 RPM. The temperature and pressure were maintained at 123°C and 1.5 kg_f/cm^2 respectively. In this experiment, the obtained value of kappa number and residual alkali were 23.18 and 42.8 g/L respectively. In this case, the increased RPM of screw feeder allowed a large amount of wheat straw feed rate and increase inside movement of the pulping mixture inside the digester tubes. Hence, the material could not get sufficient time for required delignification and resulted in the increasing value of residual alkali.

Furthermore, in experiment R_7, the white liquor flow rate was increased from 1.37 L/min to 1.71 L/min. The other cooking conditions as RPM, white liquor concentration and flow rate are kept constant as in R_6. The pressure was maintained at 0.5 kg_f/cm^2. In this experiment, the obtained value of kappa number increased from 23.18 to 28 as compared to R_6. Further, the value of residual alkali decreased in this case even at high white liquor flow rate. It shows the impregnation of white liquor is decreased in this case due to less pressure which further affects the delignification process. The flow rate of white liquor is maintained higher than the required amount for delignification process. This excess amount of chemicals are required to maintains the driving force for delignification process.

Conclusions

Soda pulping of wheat straw has been completed successfully on pilot-scale three tube continuous horizontal pulping digester. In the present work, the operating conditions are optimized for optimum kappa number and residual alkali for the soda pulping process. It has been observed that the pressure inside the digester tubes plays a significant role in the delignification process. Also, the increase in the alkali concentration and white liquor flow rate results in the increase in the residual alkali. The experiment R_3 has 13% alkali concentration with 1.34 L/min flow rate of white liquor exhibits better delignification process with the lowest value of kappa number as19.58.

References

1. Bureau of Energy Efficiency, Government of India, Improving energy efficiency in pulp and paper sectror, GIZ, New Delhi, 2018.
2. M. Sabnavis, S. Bhalerao and Mishra, *Ind. Res.*, 2018, 1.
3. A. Ashori, *Poly. Tech. Eng.*, 2006, **45**, 1133.
4. R. W. Hurter, *Agri. Resi.*, 1997, **1**, 1.
5. H. Singh *et al., Energy*, 2007, **32**, 1848.
6. G. A. Smook, "Handbook for pulp and paper technologists", 2nd ed., Angus Wilde, 1992.
7. H. Sixta, "Handbook of Pulp", Wiley-VCH, 2006.
8. P. Bajpai, "Pulp and Paper Making Process, in Bleach Plant Effluents from the Pulp and Paper Industry", Springer, 2013, pp. 7-11.
9. Y. Z. Shirkolaee, A. Talebizadeh and S. Soltanali, *Biores. Tech.*, 2008, **99**, 3568.
10. D. P. De-Almeida and J. L. Gomide, *O'PAPEL*, 2013, **74**, 53.
11. Z. Feng and R. J. Alen, *Appita J.*, 2001, **54**, 217.
12. S. K. Shomeina, O. T. Alzaki, S. D. Mohieldin and S. M. A. Yagi, *J. Mat. Env. Sci.*, 2017, **8**, 894.
13. F. Yue, Wu Lan, Songnan Hu, Ke-Li Chen and Fachuang Lu, *Sust. Chem. Eng.*, 2016, **4**, 5311.
14. A. Naser, M. A. Haque, R. Ramadevi and M. Naebe, *Cellulose*, 2018, **25**, 5455.
15. N. Sharma, R. D. Godiyal, Bhawana, B. P. Thapliyal and K. Anupam, *Waste Bio. Val.*, 2018, **9**, 409.
16. M. Singh, N. Pandey, P. Dwivedi, V. Kumar and B. B. Mishra, *Biores. Tech.*, 2019, **293**, 122105.
17. P. Dwivedi, M. Singh, N. Sehra, N. Pandey, R. S. Sangwan and B. B. Mishra, *Biores. Tech.*, 2018, **250**, 621.
18. D. Montane, X. Farriol, J. Salvado and E. Chornet, *Biom. Bioen.,* 1998, **14**, 261.
19. TAPPI Test Methods, T 221 om-85, Atlanta, USA, 1994.
20. TAPPI Test Methods, T 222 om-88, Atlanta, USA, 1995.
21. D. M. Updegraff, *Analy. Biochem.*, 1969, **32**, 420.
22. TAPPI Test Method T 624 cm-00, Atlanta, USA, 2000.
23. L. E. Wise, *Paper Trade*, 1946, **122**, 35.
24. M. Sheoran, S. Goswami, H. J. Pant, V. K. Sharma, A. Chandra, H. Bhunia, P. K. Bajpai, S. M. Rao and A. Das, *Appl. Rad. Iso.*, 2016, **111**, 10.
25. H. Sixta, "Handbook of Pulp", Wiley-VCH, 2006.
26. TAPPI Test Method T 236 om-99, Atlanta, USA.