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Paper making potential of *Amaranthus hybridus* stalks; Sustainable raw material in paper industries

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Abstract
Stalks of non-wood herbaceous species *Amaranthus hybridus* was investigated by soda pulping to investigate the future of *Amaranthus hybridus* species stalks as a auspicious renewable cradle in pulp and paper industry, which is profusely accessible in Kashmir (India) along road sides, foot paths, wastelands, pastures, hilly areas as unwanted weed. In the opportunity of current study, the element characterization of *Amaranthus hybridus* stalk dust, chemical characterization of liquor analysis and paper properties were carried out. The elemental analysis of test species bared that Ash%, Lignin%, Hot water solubility%, 10% NaOH solubility, Alcohol-benzene solubility% and Holocellulose % content were detected in the array of 11.03, 17.55, 20.25, 40.40, 6.64 and 62.41 respectively. Liquors obtained after digestion of test species results what is called as black liquor and the chemical analysis of the liquor includes pH, total solids, chemical oxygen demand and color which were chronicled 8.84, 6.82%, 226240 ppm and 115505 PCU respectively and for wash liquor the corresponding values were recorded 8.45, 1.90%, 113100 ppm and 31684 PCU. The cooked *Amaranthus hybridus* stalks subjected to soda pulping process and were no chemical was added (control) results in pulp and the strength properties of standard resultant sheets of 60 gsm of beaten pulp at 300 ml csf were assessed in terms of tensile strength (Nm/g), tear index (mN.m^2/g), burst index (Kpa m^2/g), double fold number. The overall results revealed that pulp obtained from *Amaranthus hybridus* stalks at 8% soda pulping has a auspicious potential to be used in pulp and paper industry.

Keywords: *Amaranthus hybridus*, Kashmir valley, liquor, paper properties, proximate analysis; soda pulping

1. Introduction

Herbaceous species *Amaranthus hybridus* L. a healthy and flourishing vegetable a member of the Amaranthaceae family having wide range of uses of which the common use, is as a vegetable. Leaves of *A. hybridus* is used in the treatment of intestinal bleeding, diarrhea and excessive menstruation (Foster and Duke, 1990) [11]. Stem epidermis is composed of a single row of cells flattened tangentially and fitting closely along their walls. A well-defined cuticle extends over the epidermal cells (Omosun *et al*., 2008) [25].

A sector of paper industry is one of the high demand sectors in the world of industrial production. It has been seen that total global intake for paper-making increase from 316 million tons in 1999 and 351 million tons in 2005 to about 425 million tons by 2010 (Garcia *et al*., 2008) [12]. Presently fabrication of non-wood pulp mainly takes place in countries with a shortage of wood, such as China and India (Oinonen and Koskivirta, 1999) [24].

Consumption of non-wood fibers is virtuously comprehensive way of production of pulp and paper compared to the clear-cutting of woody trees or prehistoric forests (Rousu *et al*., 2002) [29]. Other benefit of herbaceous plants as a fiber source includes their fast annual growth (Kissinger *et al*., 2007) [18]. As the impending of the wood-based fibre supply being questioned worldwide, growing interest is being shown to use several alternate fibrous sources for papermaking. Shortage and diminishing wood resources, collectively with augmented paper demand are the lashing forces to use herbaceous materials such as sugarcane bagasse (De Lopez *et al*., 1996) [6] bamboo, (Madakadze *et al*., 1999) [23], esparto grass (Jun and Tschirner, 2010) [17], sisal and kenaf (Ribas Batalha *et al*., 2012) [26]. Herbaceous fibers are copiously accessible and have turned out to be one of the significant substitute and complementary homes for fibrous material for pulp and paper making in developing countries including China, India, Thailand, and Indonesia (Ashori 2006) [3]. Additional non-woods that had been reflected consists of abaca (Jiménez *et al*. 2005) [16], *leucaena* varieties (Díaz *et al*. 2007) [7], rice straw (Rodríguez *et al*. 2008a) [27], various agricultural residues (Jiménez *et al*. 2008) [27-28], empty fruit bunches (Rodríguez *et al*. 2008b) [28], *hibiscus* species (Dutt *et al*. 2009) [8, 9],

canola stalks (Enayati *et al*. 2009) [10], rice stem fibers (Alireza and Pejman 2010) [2] and palm fruit fibers (Sridach 2010) [33].
Himalayan province of Jammu and Kashmir is gorgeous in its plant treasure due to inimitable topography and climatic conditions. A number of non-wood wild plants having well developed stem or stalk system are available in this region which may afford a hefty biomass as raw materials for pulp and paper. With this viewpoint, an endeavour has been made to study physiognomies of *Amaranthus hybridus* for suitability of pulp and paper making.

2 Materials and Methods

2.1 Pre-treatment of the Raw Material

Amaranthus hybridus stalks was collected from peripheries of the Kashmir valley and was carried to Kumarappa National Handmade Paper Institute Jaipur (KNHPI) for further investigation. All the leaf and root portion of *Amaranthus hybridus* was shredded and only the stem portion was used in the present analysis. The procured *Amaranthus hybridus* stalks were washed with tap water to eradicate unnecessary dirt, sand, nodes, and other foreign materials. The washed material was air-dried and then chopped into 1-1.5 inches size with the help of chopper and kept in polyethene bags under shady conditions for further experimental work.

2.2 Chemical Characterization of *Amaranthus hybridus* stalks

The sliced *Amaranthus hybridus* stalks were withered in oven at 103 ± 2ºC overnight and then oven dried shreds were converted to dust powder by dust machine of 0.4 mm slot size. The proximate analysis carried out included Hot water solubility (T207 om-99), 10% NaOH solubility (T212 om-98), Alcohol-benzene solubility (1:2 v/v) (T204 cm-97), Lignin (T222 om-02), Holocellulose (T249 cm-00), and Ash (T211 om-93). The pH, conductivity, total solids chemical oxygen demand and color, were determined as per standard APHA testing methods.

2.3 Pulping Studies

Stem or stalk portion of *Amaranthus hybridus* was digested in electrically heated rotary digester of 0.02 m^3 capacity having six bombs, each of 1 L capacity. Pulping was carried at dosage of 8 % soda (NaOH) and without chemical (control) at a temperature of 120º C for 3 hours with raw material to liquor ratio of 1:10.The condition of soda pulping is given as under.

Table 1: Cooking conditions of soda pulping

S. No.	Parameters	8% sulphite pulping	Without any chemical
1	Sodium hydroxide @ 8%	8.0 g	-
2	Temperature, º C	120	120
3	Time, h	3	3
4	Bath ratio	1:10	1:10

2.4 Washing studies

Washing of the beated pulp was carried in Buchner funnel to remove the residual matter in the cooked pulp and results in the formation of wash liquor. The distilled water was used for washing until the pH of the wash liquor was reduced to 8.3.

2.5 Preparation of Laboratory Handsheets and Evaluation of Paper Properties

The unbleached pulp of at 8 % soda dosage and without chemical was beaten to different beating levels. Laboratory handsheets of 60 g/m^2 were prepared by standard TAPPI testing method (T221 cm-99) and tested for Tear index (T414 om-98), tensile index (T494 om-01), Burst index (T403 om-97), Double fold numbers (T423 cm-98) and Brightness (%).

3 Results & Discussions

3.1 Proximate Analysis of *Amaranthus hybridus* stalks

Proximate analysis of the oven dried dust of *Amaranthus hybridus* stalks (% OD basis) was determined out as per TAPPI testing methods and the results are given in the table 2

Table 2: Proximate analysis of *Amaranthus hybridus* stalks dust

Parameters	*Amaranthus hybridus* stalks	Testing Method
Ash%	11.03	T211 om-93
Lignin%	17.55	T222 om-02
Hot water solubility,%	20.25	T207 om-99
10 NaOH solubility,%	40.40	T212 om-98
Alcohol-benzene solubility,%	6.64	T204 cm-97
Holocellulose,%	62.41	T249 cm-00

Proximate investigation (Table 2) indicated *Amaranthus hybridus* stalks contains considerable amount of holocellulose and lignin (17.55 & 62.41%) content which is analogous to straws and grasses. The higher concentration of holocellulose content of test species compared to agro residues and woody raw materials makes it suitable for production handmade paper, thus providing an opportunity for the production of pulp and also can blending these fibrous pulps in various proportions with different non wood fibers, depending on the desired end- products. This will help not only in tackling the issue of rare woody material scarcity but also help in decreasing the budget of the handmade paper. Hollocellulose is the total polysaccharide fraction of wood or straw or total content of carbohydrate materials. High holocellulose, therefore, is considered desirable for pulp and paper industry because it is linked with higher pulp yield and healthier strength properties by Mabilangan and Estiudillo (1996) [22]; (Shakhes *et al*., 2011) [31]. Greater the holocellulose content present in raw materials for paper production, superior they are considered for paper production. The lignin content of *Amaranthus hybridus* stalks samples is comparable or more than other non-wood raw materials and was found lower than that woody pulp. The low lignin content than woody species indicates that these fibres require very mild pulping conditions of alkaline sodium hydroxide. The data presented in Table-2 revealed that *Amaranthus hybridus* stalks showed lignin concentration of the order of 17.55 per cent. Lignin is considered to be detrimental polymer and its elimination during pulping and bleaching necessitates high quantities of energy and chemicals (Shakhes *et al*., 2011) [31]. High lignin content also advocates long cooking cycle. Additional usage of chemicals may degrade lignin and can afford virtuous pulp but at the same time there will be higher pollution of environment by the production of black liquor and more requirements of fuel, electricity and time which in the long run affect the cost economics in the paper industry. Therefore, lesser the lignin content in non-wood herbaceous raw material superior it will be appropriate for paper fabrication. Lignin content of *Amaranthus hybridus* stalks was comparable or even lower than annual plants and hardwoods (17-26%) especially with Eucalyptus (23.3%); it is however, substantially lower than softwoods (25-32%) (Ates *et al*., 2008). The acceptable levels of lignin content in *Amaranthus* *hybridus* stalks recommends that this material needs, milder pulping conditions (lower temperatures and chemical charges) than those of softwoods and hardwoods. The results also revealed that the potential of test species to undergo bleaching more easily and with application of less chemicals (Vervis *et al.*, 2004) [36]. The results showed that the ash content of the *Amaranthus hybridus* stalks (11.03 per cent) was found less than that of rice straw (16.6 per cent) but higher than that of canola (6.6 per cent), wheat straw(4.7 per cent), corn (7.5 per cent), bngkot (0.7 per cent) and almost similar to that of sunflower. The higher content of ash in the raw material does not depict good for the strength properties of paper, hence forth the species should contain lesser concentration of ash.

The examination of the data in Table-2 indicated that the mean alcohol benzene solubilty of *Amaranthus hybridus* stalks was of the order of 6.64 per cent. The higher alcohol-benzene solubility values indicate the presence of considerable amount of light weight aliphatic and aromatic compounds. These may decrease the pulping yield of the plant wastes and contribute higher chemical consumption in pulping and higher load in pulping effluent (Enayati *et al*., 2009) [10]. Non wood plant materials have also substantially higher alcohol-benzene solubility when compared with Bamboo, Eucalyptus, coniferous and deciduous wood which are the main fibrous raw materials for papermaking which leads to lower pulp yield and probably higher biological oxygen demand (BOD) load in effluents (Ates *et al*., 2008) [4].

A perusal of the data in Table-2 revealed that the hot water solubility was found 20.25 per cent. The plant material used for papermaking contain higher content of soluble compounds in hot water decrease the pulping yield and thus contribute higher chemical consumption in pulping and higher load in pulping effluent (Enayati *et al.,* 2009) [10]. The water solubility estimates are part of extraneous components, such as inorganic compounds, tannins, gums, sugars, and colouring matter present in the wood and hot water estimates, in addition to starches (Dutt *et al.*, 2009) [8, 9].

The results in Table-2 revealed that the 10% sodium hydroxide solubility content was of the order of 40.40 per cent. The high value of caustic soda solubility may also be due to the easy penetration and degradation of the cell wall by alkali as found by Kristova *et al.* (1998) [19]. But at the same time the alkali solubility values were lower than the other commonly used non wood plant materials and grasses (Enayati *et al.,* 2009; Shakhes *et al.* 2011) [10, 31].

3.2 Chemical (Black and wash liquor) analysis

Chemical characterization of the black and wash liquor of pulp *Amaranthus hybridus* stalks was determined out as per APHA standards testing methods and the results are given in the table 3.

Table 3: Chemical (Black and wash liquor) analysis

S. No	Parameters	Black liquor	Wash liquor
1	pH	8.84	8.45
2	Total solids, %	6.82	1.90
3	Chemical oxygen demand, ppm	226240	113100
4	Color, PCU	115505	31684

The data obtained in Table-3 showed that the black liquor and wash liquor of test species exhibited pH of the order 8.84 and 8.45 respectively, however in case of control (without chemicals (C_0) both black and wash liquor was found lower than that of black and wash liquors which was obtained by chemically treated pulpings as in the later no alkaline chemical was used. The values of pH of black liquor were found higher than that of wash liquors. The results of our study were in accordance with the findings of Kumar *et al*., 2013 [20, 21]. The pH of the black liquor was found to be high when compared to recommended discharged levels as listed in Minimal National Standards, prescribed by the Central Pollution Control Board, New Delhi (Pollution Control Department).

The data presented in Table- 3 indicated that the black liquor and wash liquor which was extracted after digestion exhibited chemical oxygen demand (COD) in the order of 226240 and 113100 ppm respectively. The values of chemical oxygen demand (COD) of black liquor were found higher than that of wash liquors. The overall results revealed that with the increase in concentration of alkaline dosage chemical oxygen demand (COD) was increased. The results of our study were in accordance with the findings of Kumar *et al*., 2013 [20, 21]. These results are in conformity with those of Dafinov *et al.* (2004) [5].

The results in Table-3 indicated that the total solids of black liquor and wash liquor obtained after digestion with the application of 8 per cent soda, was 6.82 and 1.90 per cent for black and wash liquors respectively.

Total solid content was found highest in chemical pulping and lowest in case of control pulping. The overall results showed that the total solids concentration was much higher in the black liquor when compared with recommended discharged levels as listed in Minimal National Standards, prescribed by the Central Pollution Control Board. But at the same time the total solid concentration was much lower when compared with black liquor effluent of conventional paper mill effluents where paper is being made from softwood and hardwood species of trees (Saravanan and Sreekrishnan, 2004) [30].

The results in Table-3 indicated that the Color of black liquor and wash liquor obtained after digestion with the application of 8 per cent soda pulping was of the order of 115505 and 31684 PCU. The overall results showed that the Color concentration of the black liquor of pulp produced was much lower when compared with the colour of black liquor of conventional paper mill effluents (Saravanan and Sreekrishnan, 2004) [30].

3.3 Physical strength properties of unbleached paper

Paper properties are considered as the characteristic properties which actually determine the quality of the paper. The strength properties of test species at soda pulping process with chemical dosages of 8% beaten at 300 ml freeness level was evaluated out as per TAPPI standards and the results are given in the table 4.

Table 4: Strength properties of unbleached and bleached pulp of *Amaranthus hybridus*

S. No	Parameters	Without any chemical	8% soda paper
1	Tensile index, Nm/g	3.95	12.43
2	Tear index, $mN.m^2/g$	0.92	3.94
3	Burst index, $Kpa.m^2/g$	0.02	0.93
4	Double fold number	3	12

Application of soda chemical pulping at the dosage of 8 % NaOH had significant effect on tensile strength of paper of *Amaranthus hybridus*. Among the pulping, highest strength properties tensile strength, tear index, burst index and double

fold no. was found in soda pulping than control in pulping
Characteristic strength properties of the paper achieved from non-wood herbaceous weed are presented in Table 4. Results of the present study revealed that paper obtained from soda pulp showed the higher strength properties than controlled paper (without chemical) which might be probably due the better delignification of pulps in sulphite pulping by higher alkaline nature of chemicals (Jahan *et al*., 2007) [13]. It has been found that fibre cell wall became swollen greatly due to the high charge of alkaline pulping doses and most of the lignin is removed from the fibres (Vainio *et al*., 2007) [35] which results in higher strength properties with alkaline nature. The tensile strength of test species was found higher than other nonwood plant fibres viz; 8 % soda palmyra palm fruit fibres (9.20 Nm/g) Sridach (2010) [33]. The tear index of paper of the test species was higher than other nonwood plant fibres viz; vine shoots kraft pulp (0.31 $mN.m^2/g$) (Jimenez *et al*., 2007), wheat straw (3.07 $mN.m^2/g$) (singh *et al*., 2011) [32]. The burst index of test species was higher than other nonwood plant fibres viz; holm Soda–Aq (0.42 $KPa.m^2/g$) and Holm kraft (0.53 $KPa.m^2/g$) (Alaejos *et al*., 2006) [1]. However double fold characteristics of papers were higher than other non-wood plant fibres viz. viz; *Acacia auriculiformis* soda - AQ (02), *Acacia auriculiformis* soda (01) and *Acacia auriculiformis* kraft (03) Jahan *et al*. (2008) [14].

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