Oct. 31, 1939. U. POMILIO 2,178,266

CONTINUOUS PROCESS FOR THE MANUFACTURE OF SEMIPULP

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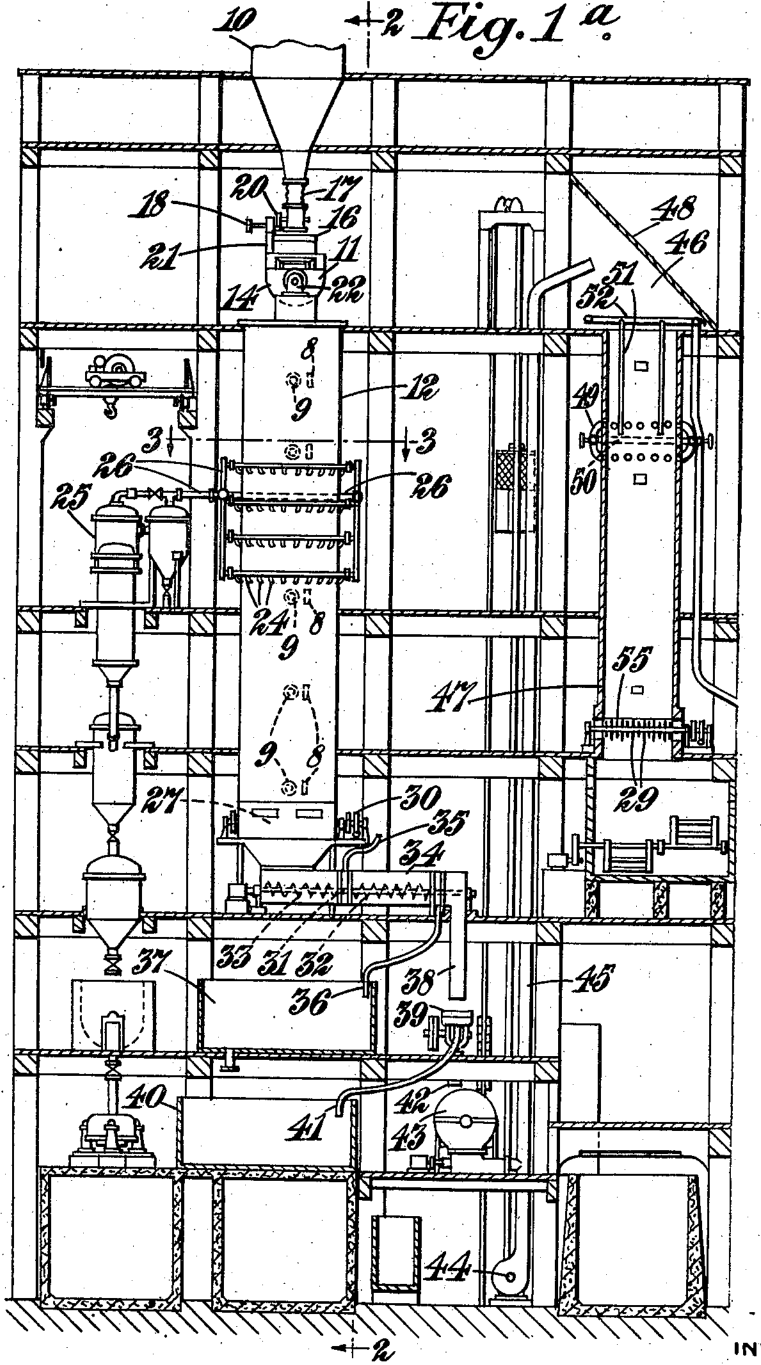
Fig. 1a



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CONTINUOUS PROCESS FOR THE MANUFACTURE OF SEMIPULP

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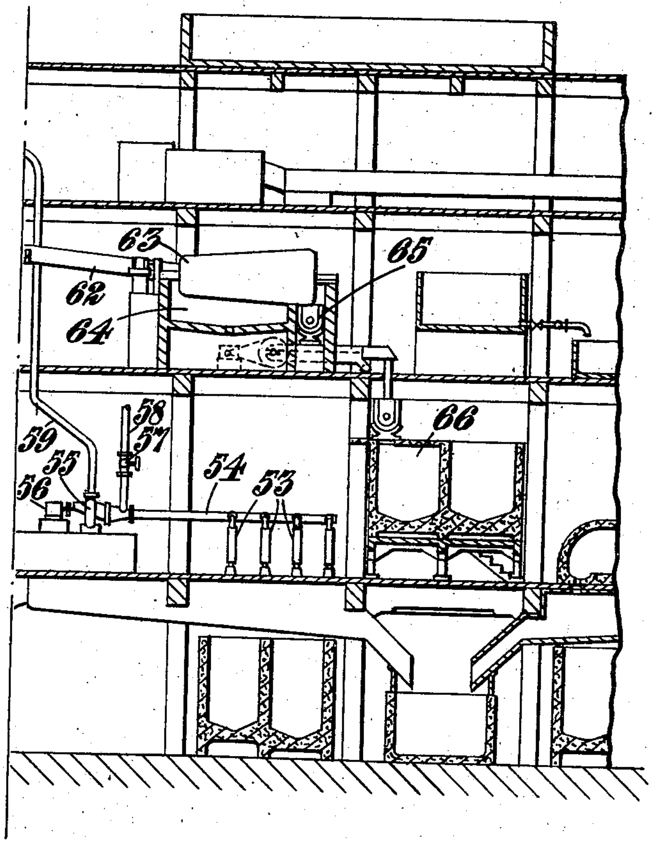
Fig. 1b.



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CONTINUOUS PROCESS FOR THE MANUFACTURE OF SEMIPULP

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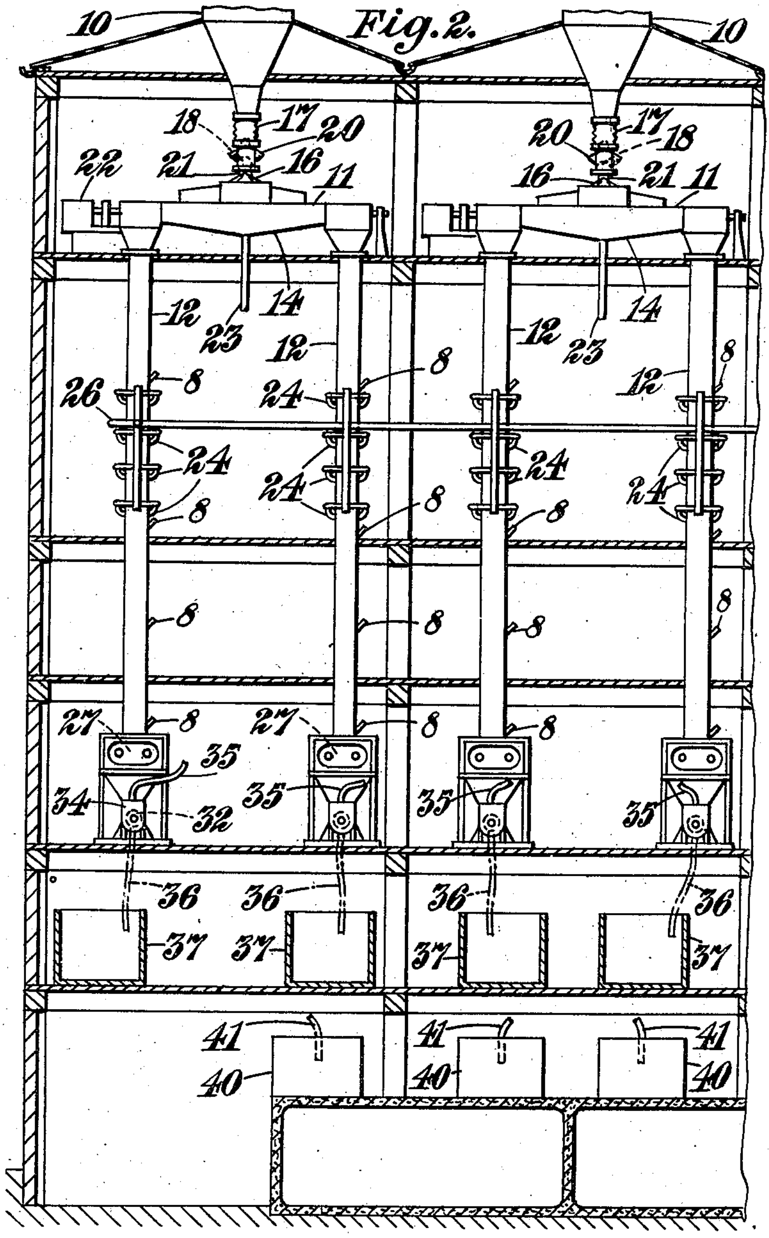
Fig. 2.



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CONTINUOUS PROCESS FOR THE MANUFACTURE OF SEMIPULP

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Fig. 4.

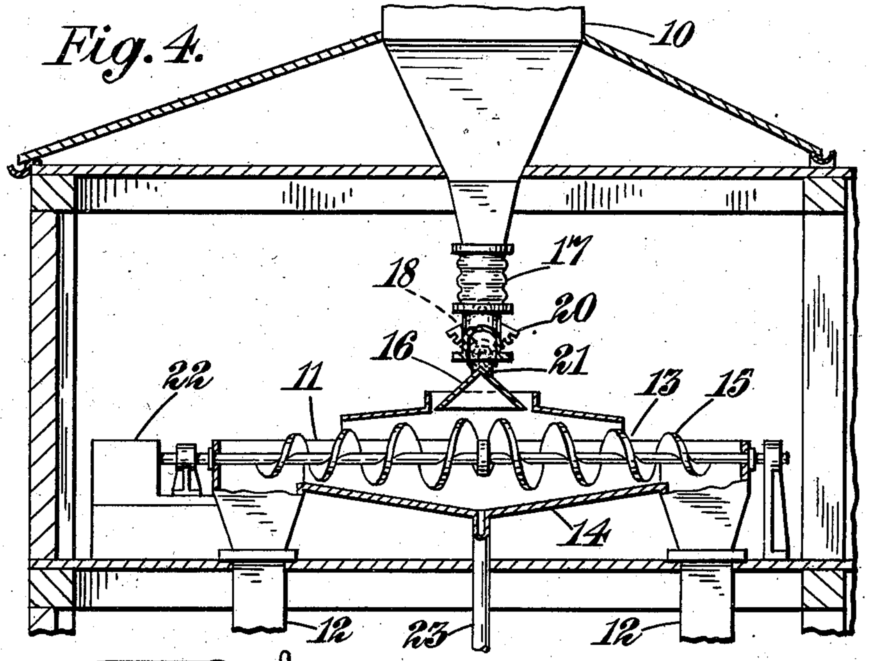

Fig. 3.

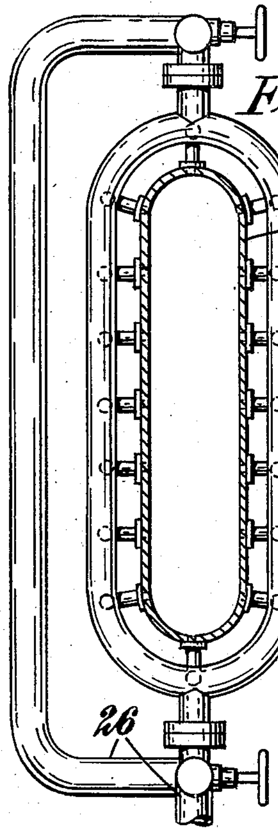

Fig. 5.

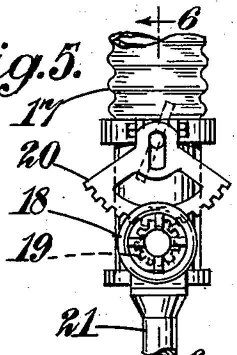

Fig. 6.

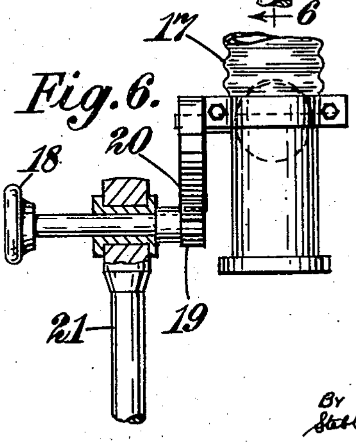

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Fig. 7.

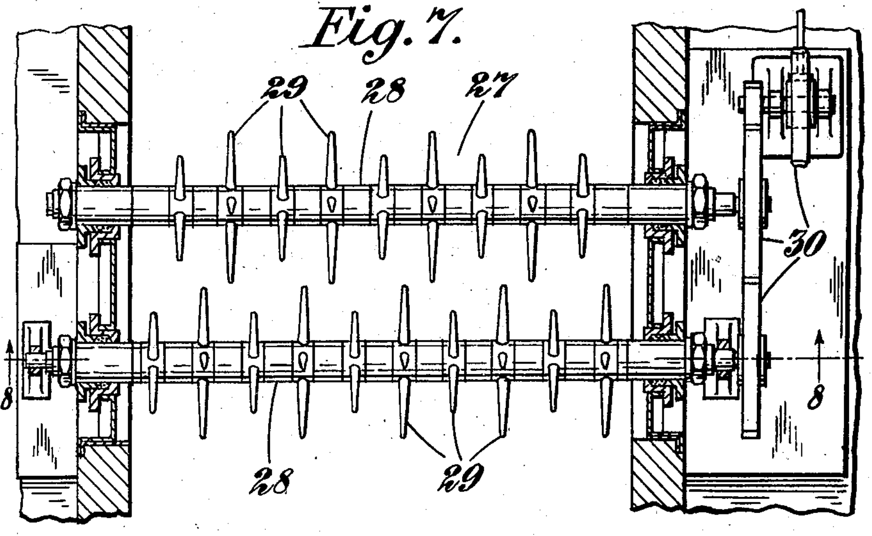
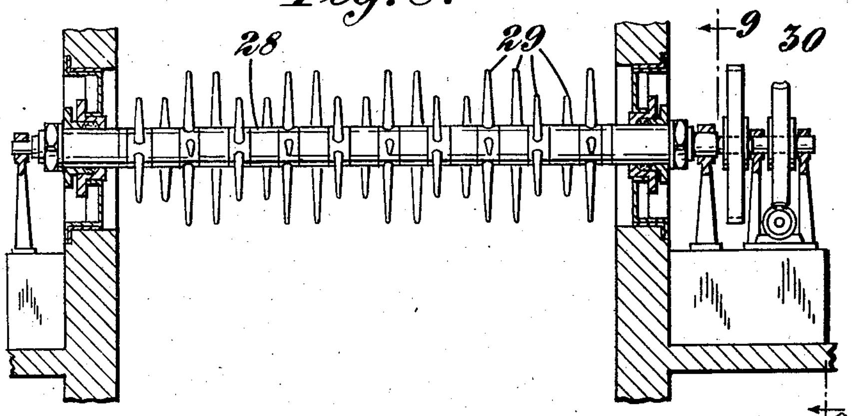


Fig. 8.



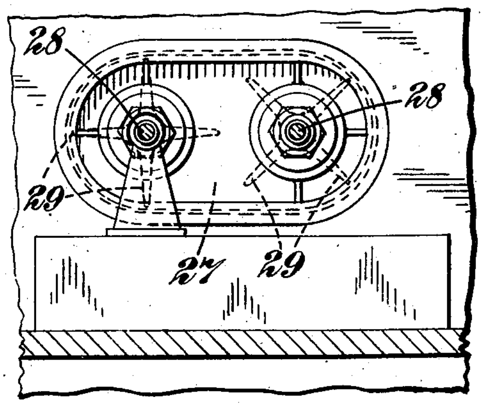


Fig. 9.

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UNITED STATES PATENT OFFICE

2,178,266

CONTINUOUS PROCESS FOR THE MANUFACTURE OF SEMIPULP

Umberto Pomilio, Rome, Italy, assignor to Pomilio Corporation Limited, London, England

Application July 16, 1937, Serial No. 153,995
In Great Britain January 23, 1937

1 Claim. (Cl. 92—13)

This invention is for improvements in or relating to processes and apparatus for the extraction of cellulosic fibres from vegetable material such as the straws of wheat, barley, rice, rye and other cereals, the stalks of flax, cotton or hemp, canes such as sugar cane, bagasse, Danube canes, bamboo, grasses such as esparto, sisal, tambucki, Tatching, elephant grass and other wild grasses, or wood, or other fibrous materials such as tow residues and residues from the manufacture of natural textile materials, all hereinafter referred to as "fibrous vegetable matter".

The object of the invention is to produce the cellulosic fibres in the state of a semi-pulp suitable for the manufacture of paper (a semipulp being a pulp from which the original noncellulosic materials have not been entirely removed).

In general the known chemical processes of making paper pulp for fine or white papers aim at substantially complete removal of the noncellulosic materials and these processes are relatively expensive.

There are two well-known methods of producing cheap semipulp. In the Masonite process wood of certain types cut into small pieces is subjected in an autoclave to the action of steam under high pressure which acts both thermally and chemically on the encrustants of the cellulose fibres. When the pressure is suddenly released the loosened vegetable material is broken up into filaments. In this process there is no appreciable removal of the encrustants and the pulp is of a brown colour. It is used for making boards but is unsuitable for making paper since the pulp is not resolved into separate papermakers' fibres.

Mechanical pulp is made by grinding wood with grinding stones (generally the whiter woods). The wood may be steamed before grinding and again the steaming operation may result in a certain degree of caramelization producing a brown colouration. The power required for the production of mechanical pulp is high, generally of the order of 1000 kilowatt hours per ton of ground wood or more. Mechanical pulp contains all the original encrustants but the filamentary character of the cellulose is destroyed. It is not normally used alone for the manufacture of paper. It is employed up to a proportion of about 80% in newsprint (generally in admixture with chemical pulp) and it is employed in a proportion of 25–40% in many other grades of cheap paper.

In the grinding operation many of the cellulose fibres are themselves sub-divided and it is practically impossible in the production of mechanical pulp to preserve much of the strength of the original plant filaments. Mechanical pulp is produced only from wood.

For the manufacture of paper the cellulose fibres should, as far as possible, preserve their original length and quality. It is true that in the beating engine certain long fibres, such as cotton fibres, are cut to a shorter length but the cellulose fibres which in nature are short already, such as those in straws and stalks and wood, should reach the beater, as far as possible, in their original length.

Speaking broadly, the problem in the present invention lies in utilising a wide variety of fibrous vegetable materials (in each locality the cheapest available), in cutting out or cutting down costly operations or costly materials, and yet producing a semipulp which can be sent to the beating engine with the confidence that it will produce marketable paper.

Fibrous vegetable materials are composed broadly speaking, of cellulosic fibres which are embedded or encrusted in non-cellulosic material including lignins, waxes, resins, pectic and mucic acids, albuminoid matter, starchy matter, sugars (like pentosanes) and inorganic matter.

It is already known that the encrusting materials may be removed from fibrous vegetable matter by treatment with alkali and with chlorine. The alkali interacts with certain encrusting substances of an acid character or saponifiable character, such as waxes and resins and the pectic and mucic acids, whilst chlorine reacts with other encrustants, particularly with the lignins to form chloro-lignins and hydrochloric acid, all of which can then be leached out.

According to the present invention, a process for the manufacture from fibrous vegetable matter of semipulp suitable for paper-making and cardboard, comprises reacting on the vegetable matter successively with alkali and chlorine gas and is characterized in that the quantities of the reagents employed are no more than sufficient to remove a part only of the non-cellulosic constituents and in that the alkali reaction is carried out with the application of heat at a temperature between 75° C. and 100° C. and the chlorine reaction is carried out without the application of heat.

It is found that the more objectionable noncellulosic constituents can be attacked by a more dilute reagent than that required to attack the other constituents. Thus, in carrying out the process referred to above, the dilution of the reagents may be so selected as to attack mainly the more objectionable non-cellulosic constituents while leaving the greater part of the other constituents in the semipulp.

The process may also be carried out by providing only just sufficient quantity of the reagent as will remove a predetermined proportion of the non-cellulosic constituent, while the strength of the reagent may be such as to attack any or all of the non-cellulosic constituents. Furthermore, the process may be carried out by so controlling the time over which the reaction takes place that a predetermined quantity of the non-cellulosic constituent is rendered capable of removal while the strength of the reagent may be such as to attack any or all of the non-cellulosic constituents.

It has been found that the temperature under which the reactions are carried out also affects the physical characteristics of the resulting fibre in that high temperature conditions tend to damage the fibre, the fibre being increasingly damaged as the temperature rises above 100° C. A still more important consideration is that uniformity of resolution is favoured by carrying out the reactions at low temperature. The reactions with alkali and with chlorine are exothermic and high temperatures are liable to be built up and accordingly it is a feature of the invention that the treatment with alkali and/or with chlorine is not only restricted as regards the extent of removal of non-cellulosic constituents but also is carried out under conditions which ensure that the temperatures built up during the reactions are sufficiently low to diminish damage to the fibre.

A still further feature of the invention consists in that the process comprises a preliminary stage in which the material is treated with a dilute alkali solution, a second stage in which the material is treated with chlorine and a third stage comprising a treatment with a very dilute alkali solution. Since these reactions are exothermic, the temperatures built up are dependent, inter alia, on the velocity at which reactions take place and on the rata at which the evolved heat is dissipated through the material and through the walls of the reaction vessel and accordingly it may be desirable to cool the reaction mass during the chlorination and final alkali treatment.

In a process for treating fibrous vegetable materials with caustic soda and with chlorine, the cost of the chemicals is a very important item. In the present invention caustic soda may be used in a dilute aqueous solution (a form of alkali readily obtainable by electrolysis of common salt solution). A cheap alkali such as soda ash may also be employed. The gaseous chlorine may be used in a moist state diluted with the gases of the air, a form of chlorine which is likewise very readily produced by electrolysis of salt solution. This invention therefore includes a process for the manufacture from fibrous vegetable matter of semi-pulp suitable for paper-marking in which an aqueous solution of common salt is electrolysed to produce a dilute alkali solution and moist gaseous chlorine and in which the moist gaseous chlorine diluted with air is used for the second stage of the process and the caustic soda solution (at the required concentration) is used for the first and third stages of the process.

The process in its preferred form comprises three stages, viz:

1. A treatment of the fibrous vegetable matter with a solution of caustic soda of 1% or less for the removal of a substantial proportion of the acid and saponifiable encrusting constituents;

2. A treatment of the resulting material with moist chlorine gas diluted with air, which treatment is controlled in the manner set forth above;

3. A treatment of the resulting material with very dilute solution of caustic soda of .2% or less.

As already indicated, the amount of caustic soda used during the first or third treatment and/or the amount of chlorine used during the second treatment can be so controlled that these chemical reagents are completely used up in attacking a predetermined amount of the non-cellulosic constituents. It is an important practical feature of this invention that the vegetable matter receives its chemical treatments in a continuous manner (as distinct from batch treatment), the reactions being carried out successively in separate reaction chambers.

A further feature of the invention consists in that the vegetable material is caused to descend under gravity through reaction towers and that the reagents are introduced at zones near the upper ends of the towers. With this arrangement, the vegetable material becomes progressively consolidated as it descends through the tower and this tends to force the reagents to flow in countercurrent in spite of the fact that in the case of chlorine the gas is heavier than air. By reason of the downward movement of the vegetable matter, a certain amount of the reagents may also be drawn downwardly. In the case of the chlorine, by suitably selecting the point of its introduction into the tower and by controlling its rate of feed and the rate of feed of the vegetable material, the chlorine is prevented from reaching either end of the tower before being completely used up in the reaction. Thus there can be no leakage of chlorine into the atmosphere nor are any sealing means required at the ends of the tower.

The velocity at which the reaction takes place in the alkali treatment is dependent to some extent on the strength of the alkali solution employed.

For treating straw, should it be required to remove the greater part of the non-ligneous and non-cellulosic material and to remove about half the ligneous non-cellulosic constituents, the strength of the alkali solution in the preliminary stage is of a concentration up to about 1%. When such a solution is employed, the relative proportions of the reacting substances are selected to be 1 part by weight of vegetable material to 3 parts of the solution. The reaction is carried out in an open vessel without pressure for a period of from 1–4 hours according to the strength of the solution. The temperature of the reaction which is exothermic may vary from 75° C. to 100° C. This treatment would result in a reaction with the greater part of the non-ligneous and non-cellulosic constituents.

The chlorine reaction is also exothermic. Preferably, the chlorine gas is diluted with air and it may be moistened. The reaction mass may also be moistened. The chlorine is used in quantities of about 4 to 5 parts by weight to 100 parts of the straw, and this will result in the reaction with about one half of the ligneous non-cellulosic constituents. About half of the chlorine reacts to form chloro-lignin whilst the remainder of the chlorine is converted into hydrochloric acid and it is advisable that the time during which the fibres are in contact with the hydrochloric acid, resulting from the chlorination, should be reduced to a minimum since the cellulosic fibres are liable to be attacked thereby.

An important feature of the invention consists in that the chlorination treatment and the final alkali treatment are conducted in the cold. The dilution of the chlorine with air has a dual effect. It reduces the reaction velocity and thus the rate at which heat is evolved, and it also serves to absorb and conduct away some of the heat. The moistening of the gas and the reaction mass serves by absorption to reduce concentration within the fibre of the hydrochloric acid which is there formed and thereby minimises its deleterious effect on the fibre. As soon as chlorination is complete the reaction mass is immersed in cold water. The proportion of air used with the chlorine is dependent on the nature of the vegetable material being treated and it may vary from .5 to 1 part by volume of air to one part by volume of chlorine, whilst the moisture content of the chlorine may be up to its saturation point and the moisture content of the mass may be two to five times the weight of the mass itself.

The proportion of chlorine-air mixture to the vegetable material may vary with the nature of the material being treated, and may vary from 4 to 5 parts by weight of chlorine to 100 parts of vegetable material. The length of chlorination treatment may vary from half-an-hour to three hours by varying the rate of traverse of the material through the apparatus. The length of the chlorination treatment may vary from half-an-hour to three hours.

The compounds formed by these two reactions may not be entirely removed by washing and the third stage, namely, the treatment with a very dilute alkali solution, effects the removal from the material of those compounds affected by the first two treatments but still remaining on the fibres. The strength of the solution employed in this final stage may be about 1%. The quantity of the solution employed is from 5 to 20 parts by weight of solution to 1 part by weight of the vegetable material and the reaction is carried out in the cold.

The following is a description of one form of apparatus for carrying out the process described above, reference being made to the accompanying drawings, in which—

Figure 1^a and Figure 1^b taken together represent a side elevation of the plant showing diagrammatically the general arrangement,

Figure 2 is a vertical section on the line 2—2 of Figure 1^a showing the alkali towers in elevation,

Figure 3 is a section through a tower on the line 3—3 of Figure 1^a,

Figure 4 is an enlarged view of one of the mixing devices of Figure 2,

Figure 5 is an enlarged view of the mechanism for tilting the hopper nozzle as shown in Figure 4.

Figure 6 is a side elevation of the device shown in Figure 5.

Figure 7 is a plan view of the extractor for the reaction towers.

Figure 8 is an elevation of the arrangement shown in Figure 7.

Figure 9 is a cross section on the line 9—9 of Figure 8.

Assuming stalks or straw of plants, such as are referred to at the commencement of the specification, are to be treated, these are cut into lengths of a few inches and are loaded into hoppers 10 arranged at the top of the apparatus. The apparatus about to be described is designed for producing about 15 tons of cellulose per day. Each hopper is arranged to deliver the material to an apparatus 11 which mixes the stalks with an alkali solution in the required proportions and delivers the mixture to the upper ends of a pair of vertically-arranged reaction towers 12. Each mixing apparatus thus serves two towers, and comprises a long horizontally-disposed chamber 13 open at the top, the lower wall 14 of which is inclined downwardly from each end to the centre to form a V. A screw-conveyor 15 is horizontally-disposed along the length of the chamber and has two sets of helical blades arranged end to end, which blades are oppositely pitched to one another, and are driven by a motor 22. Disposed above the chamber and at the centre of the screw-conveyor is a V-shaped deflector 16. The hopper is arranged to deliver the material on to the deflector through a flexible nozzle 17 which may be swung from one side to the other of the V-shaped deflector thereby adjusting the relative quantities of the material being fed to the two towers. The movement of the nozzle may be effected by a hand wheel 18 fixed to a worm shaft 19 in engagement with a rack 20 secured to the nozzle. The worm shaft is mounted in a bracket 21 secured to the chamber 15. Caustic soda solution is supplied to the chambers through a conduit 23 and a suitable device for maintaining a constant level of the solution in the V-shaped recess in the bottom of the chamber but which level does not reach the blades of the conveyor. As already indicated, the strength of the solution may be 1% or less. Each end of the chamber is open and the screw-conveyor forces the material mixed with the alkali out through said open ends from which mixture drops into the open end of the reaction tower 12.

The reaction towers may be about 30 feet high and in cross section are flattened oval-shaped (as will be seen from Figure 3), the smaller dimension being about 15 inches and the larger dimension 8 feet. The conveyor 13 is so arranged as to effect a mixture of straw and solution in proportions of one of straw to three of the solution. Steam is introduced through the walls of the tower by a number of pipes 24 in a zone substantially above the horizontal centre line, e. g., one-quarter of the length from the top. The steam generating plant 25 and main delivery pipe 26 are shown to the left of Figure 1. The mass of material gradually descends under gravity and becomes compacted as it descends, and at the bottom forms a seal of interlaced fibres which prevent too rapid an escape of the liquid. The time of descent may vary from one to four hours according to the nature of the material and the strength of the caustic solution. During this time the alkali is almost completely used up to form spent liquor of a dark colour. The tower is provided with inspection windows 9 and sockets 8 for thermometers at different heights.

The mass of interlaced fibres is removed from the bottom of the tower by an extractor 27, the speed of extraction being adjustable; thus, the time of treatment may be varied.

As will be seen from Figures 7 to 9 the extractor comprises two parallel shafts 28 each of which carries a number of star-shaped members 29. The two shafts are rotated through gearing 30 in opposite directions so that the arms of the stars draw the material downward and first move towards one another and then away from one another. The mountings for shafts 28 and the gearing 30 may be so arranged that the distance apart between the shafts may be varied. The gearing may be driven by an electric motor (not shown) the speed of which may be controlled thereby controlling the rate of withdrawal of the material from the bottom of the towers.

The fibres are separated from the spent liquor by a screening device 31 through which the material is fed by a screw conveyor 32. The screening device is formed in two parts 33, 34 arranged end to end along the length of its conveyor and the part at the outlet end 34 receives a supply of wash water through a pipe 35 which wash liquor is delivered by an outlet pipe 36 to a tank 37. The washed and partly-treated fibres are delivered by the conveyor through an outlet conduit 38 to a screw-press 39 where the remainder of the spent liquor is squeezed out of them and delivered to a tank 40 through a pipe 41. The consolidated mass is then passed through a conduit 42 to an opener 43 having spiked rollers or combs where it is converted into a floccular state. It is then raised by air blowers 44 through suitable conduits 45 to a receiving chamber 46 arranged above the upper end of a set of chlorinating towers 47. The upper ends of the chlorinating towers are open and the conduits 45 are directed on to a deflecting plate 48 in the collecting chamber 46 whereby the floccular material is caused to fall into the towers.

These towers may also be about 30 feet in height although those shown in the drawing are shorter. They are of a flattened oval shape in cross-section, the general dimensions being the same as those of the alkali towers. Chlorine is introduced into the fibrous mass at a zone about 10 feet from the top of each tower. As already indicated the top of each tower is open but the 10 foot column of fibrous material above the zone is sufficient completely to absorb the chlorine and prevent leakage, and the sealing action is enhanced by the fact that the material is descending. The chlorine is introduced into the zone by pipes 49 which extend from a header pipe 50 through the walls of the tower and stop flush with the inside face, and also by means of a set of pipes 51 which extend downwardly from a header pipe 52. From practical considerations the width between the side walls of the tower cannot be made much less than 15 inches and since chlorine cannot readily penetrate the fibrous material to that extent, the additional vertical delivery pipes are provided. Thus a comparatively small penetration is required without the necessity of applying substantial pressure to the chlorine. Chlorine with the required degree of moisture may be prepared in an electrolytic plant 53 from which it passes through a delivery conduit 54 to a blower 55 driven by a motor 56. The inlet or suction side of the blower communicates with the atmosphere through a valve 57 and air pipe 58. By adjusting the valve 57 a desired quantity of air may be mixed with the chlorine. The chlorine air mixture is delivered by a pipe 59, to the aforesaid header pipes 49 and 52 through suitable valves 53 and 54. The fibrous material gradually sinks through the tower and is removed from the bottom of the tower by an extractor 55, similar to the aforesaid extractor 27 the speed of extraction being adjustable as in the case of the alkali tower. As in the alkali tower inspection windows 9 and sockets for thermometers are provided.

The material is then delivered by the extractor into a tank 60 having rotating stirrers 61 therein through which tank cold water is circulated. The material passes thence along a trough 62 to a rotating conical sieve 63, the axis of which is horizontal and the angle of inclination of the walls of which is such as to feed the material from the smaller end of the sieve to the larger end. The diluted hydrochloric acid percolates out through the sieve walls into a tank 64 and the fibrous material thus partially freed of acid is passed through screw-presses 65 where it is consolidated. The fibrous material is then passed through a mixing vat 66 containing a weak solution of caustic soda, e. g., a soda solution of .2% concentration or less. The material is treated in this vat for a fraction of an hour and this results in the removal of the chloro-lignin compounds and/or other non-cellulose soluble matter. Also, any remaining hydrochloric acid or chlorine combines with the caustic soda. Thereafter the fibres are passed through the usual washing and sorting apparatus where knots are removed.

As already indicated the towers both for the alkali and chlorine treatments are provided with thermometers so that the predetermined temperatures of the reactions may be maintained, for example, by controlling the rate of extraction of the material from the bottom of the towers. Furthermore, means are provided for periodically testing the air and moisture content of the chlorine mixture.

The starting material may be considered to contain roughly 50% cellulosic constituents and 50% non-cellulosic constituents. The three-stage treatment, namely, the first alkali treatment and the chlorine treatment and the final stage, removes the required amount of the readily removable constituents and at the same time leaves behind on the fibres a substantial part of the more desirable non-cellulosic constituents, especially the ligneous constituents. If the control of the chemical treatment is such that only half the non-cellulosic material is removed, the total yield of solids in the pulp would be about 75% of the original weight of vegetable matter. By operating in this manner it will be appreciated that not only is the time of treatment very considerably reduced but also the consumption of chemicals is very much less than where the treatment is carried out to remove the whole of the encrusting materials from the cellulosic fibres. The semipulp which is obtained by the process herein described if desired is capable of being satisfactorily bleached, satisfactorily treated in the beating engine and may be easily felted to form a good grade of paper. The process, therefore, may be very cheaply operated to produce high-grade semipulp.

I claim:

A continuous process for the manufacture of semipulp from straw which consists in mixing the straw with an alkali solution of 1% strength and in proportions of 1 part by weight of straw to 3 parts by weight of the solution, traversing the mixture through a reaction tower raised to a temperature between 75° C. and 100° C. at such a rate that the material remains in the tower from 1 to 4 hours, transferring the material to a vessel where it is washed with water, dewatering the material and introducing it with chlorine gas diluted with air in proportions of .5 to 1 part by volume of air to 1 part by volume of chlorine and in quantities such that there are 4 to 5 parts by weight of chlorine introduced to 100 parts by weight of straw, traversing the material at such a rate that it remains in the reaction vessel from half an hour to three hours, immersing the material, immediately after leaving the last said vessel, in a cold water bath, transferring the material from said bath to another vessel and treating it with an alkali solution of a strength of .1% and in quantities from 5 to 20 parts by weight of solution to 1 part by weight of the vegetable material.

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