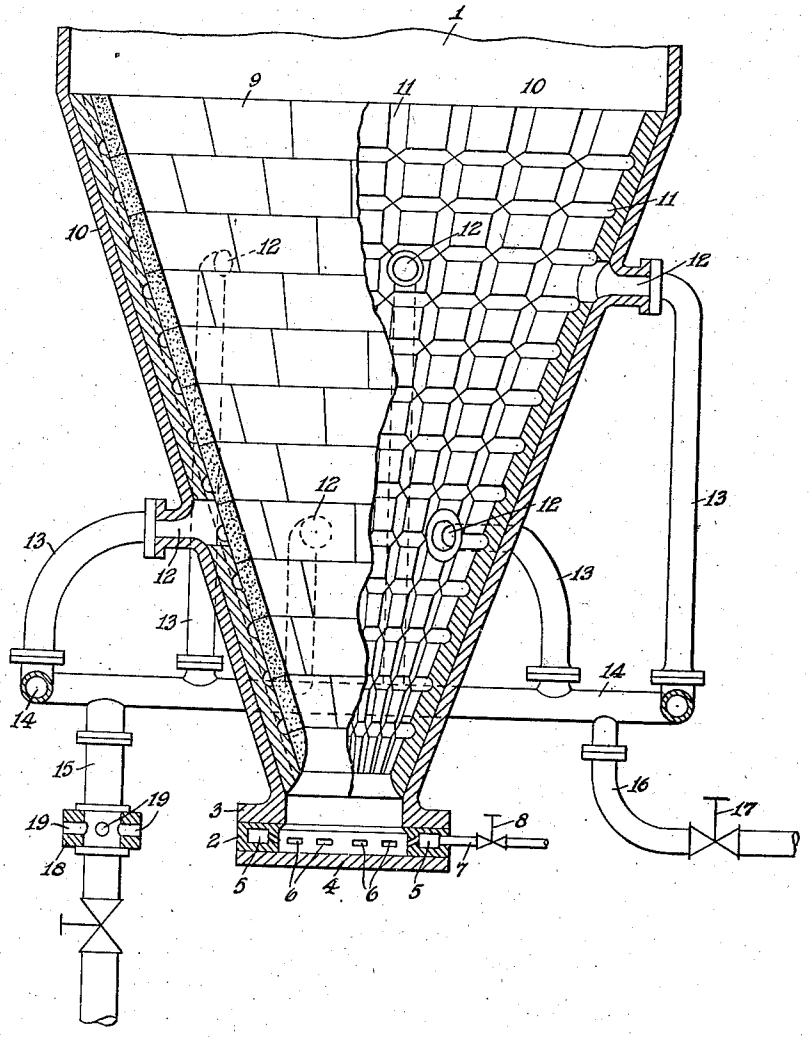
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APPARATUS FOR THE SACCHARIFICATION OF CELLULOSE

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APPARATUS FOR THE SACCHARIFICATION OF CELLULOSE

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5 Claims. (Cl. 127—1)

This invention relates to an apparatus for the saccharification of cellulose with dilute acids and is a division of my copending application Serial No. 741,374, filed August 25, 1934.

Patent No. 1,990,097 inter alia relates to a process of saccharification of cellulose wherein cellulose material is maintained at an elevated temperature and pressure and dilute acids intermittently flowed therethrough at predetermined intervals of time to promote the saccharification reaction and remove the sugar formed. During the intervals between the passage of the acids, the cellulose material is in a wet, moist or semi-dry state and the saccharification reaction continues.

Decomposition of sugar formed in the aforementioned process is a function of the length of time during which the formed sugar remains exposed to the action of the acid and heat (time of exposure). In the process aforementioned, the sugar may be removed very rapidly, but this requires large quantities of dilute acid whereby the process is rendered uneconomical.

In the process described previously, the sugar is obtained in the form of a 4% solution and in a quantity up to 80% of the theoretical yield of sugar. The theoretical sugar yield of 100 kgs. of wood amounts to approximately 60 kgs. in round figures, but in practice the yield in approximately 48 kgs. Aside from a small remainder of undecomposed cellulose, the difference between theoretical and practical yields (approximately 12 kgs.) is lost through the processes of decomposition. Approximately 6 kgs. are lost through the decomposition of the sugar within the particles of the initial material and the remainder lost on the way through the percolator.

The figures given in the paragraph immediately preceding are not generally applicable and depend among other things on the kind of wood and the size of the particles. The high degree of decomposition within the particles finds its explanation in that sugar with the old process required a comparatively long time to diffuse from the particle and reach the flowing stream.

The instant process contemplates shortening the time of exposure in such a manner whereby it is unnecessary to increase the quantity of liquid and also the rapid and careful removal of the sugar.

The shortening of the time of exposure, according to the principles of the instant process, is secured by repeated and periodic increase and/or decrease of temperature and/or pressure with or without changes in the acid concentration, and also the choice of a favorable proportion between the time of exposure and the total time of reaction.

The nature and object of the instant invention will become more apparent from the following description, appended claims and accompanying drawing, wherein the figure illustrates a view, partly in section, of the lower portion of a percolator constituting the instant invention.

In carrying out my process, cellulose material is introduced in the percolator and preferably compressed therein. While the cellulose material is being heated under pressure, batches of dilute acid solution are intermittently introduced at the top of the percolator and flowed into and out of the cellulose material to promote the saccharification reaction and remove the sugar formed. In the intervals between the passages of acid, the cellulose material is in a wet, moist or semi-dry state, but the saccharification reaction continues. This procedure is more fully explained in Patent No. 1,990,097.

As above explained, in the process described in Patent No. 1,990,097, the cellulose material is periodically at rest in the half-moist state and surrounded by vapors. In the pores of the cellulose particles there is naturally present more or less moisture which is retained by capillary forces.

I have found that a simple but very effective way of shortening the time of exposure of the sugar and increasing the sugar yield consists in the use of comparatively cold batches of dilute acid. If a batch of liquid flows at a very low temperature, for example 10 to 20 degrees below the temperature of the cellulose material, through the percolator, there occurs within the range of the batch of liquid, and especially below the batch of liquid, a decrease of temperature and pressure which causes steam to develop in the capillaries of the particles. The steam expels the liquid from the particles. Thus, the sugar present in the particles is very quickly removed from the interior thereof and reaches the batch of liquid which transports it from the reaction vessel. The batch of liquid, being under a higher pressure, can then again enter the capillaries.

I have also found that the cold batches of the liquid afford the additional advantages of flowing in a comparatively compact manner and with comparatively great speed. The flow of the batch of liquid through the percolator in accordance with the instant invention may be accelerated by applying pressure thereto from above, for example, with steam having a higher temperature

than the cellulose material and, therefore, also a higher pressure than that previously present in the percolator. The increase in pressure caused by the steam when it is introduced, as just explained, may be approximately 2 atmospheres. In this way, it is possible to force the batch of liquid through a layer of compressed cellulose material having a height of 10 m. in a few minutes.

The use of cold batches of liquid and the added pressure by means of steam, above described, tend to impart to the upper part of the percolator a temperature higher than the temperature in the lower portion of the percolator. Such an operation would result in the reaction in the upper part of the percolator taking place at a greater rate than in the lower part, with the consequence that an unequal disintegration impairing the total result would be secured. In order to eliminate this defect, the invention also contemplates blowing in steam from below during the period of rest. The steam may be introduced in the lower portion of the percolator in any convenient manner, such as, for example, through a suitable steam range on the lower cone of the percolator or through the filter itself.

The introduction of steam from below is preferably continued while the batch runs into the upper part of the percolator. As a result, at first the batch of liquor cannot enter into the material since this is prevented by the steam meeting the batch of liquor. As soon as the whole batch of liquor is introduced into the percolator, the introduction of steam from below is stopped and steam is introduced from above, as previously described, whereby the acid is caused to flow through the percolator.

In addition to the change of temperature and pressure, as above explained, a change in acid concentration may be also used to advantage.

If aqueous solutions of acid are used and the acid concentration thereof is considerably lower in the lower part of the percolator than in the upper part of the percolator, the weak acid functions to remove the sugar, while the stronger acid solution largely adheres to the material and causes the saccharification reaction to continue during the rest period. The weak acid solution portion of the following batch then meets the material that has absorbed the stronger acid of the preceding batch and the higher concentration potential created thereby between the liquid in the particles, and the liquid outside the particles is favorable to the removal of the sugar from the particles.

Another advantage which the use of batches of unequal acidity offers when employed in the manner just mentioned is the saving of acid, since the sugar is removed with that part of the batch which is poor in or free from acid. Therefore, the speed of the reaction is essentially determined by the acid concentration of the batch which largely adheres to the material.

The employment of different acid concentrations may be utilized in such a manner that the acid content of the batches is increased. In other words, each succeeding batch may have an acid concentration higher than that of the preceding batch. A preferred embodiment contemplates the use of comparatively low concentration acids at the beginning with correspondingly higher temperatures.

The two expedients, i. e. the use of batches having an unequal concentration and the use of batches with increasing acid content, may be used simultaneously. Instead of one batch, the acid content of which is higher in the upper part than in the lower, there may, of course, be used two batches, the first of which is poor in or free from acid, while the second is rich in acid, the two batches following each other at short intervals. The gradual increase of acid concentration replaces entirely or partially the increase of temperature of the process described in Patent No. 1,990,097.

I have found that by raising the factors in the reaction, i. e. temperature and acid concentration, the sugar solution formed is approximately of a constant concentration or gradually becomes lower. For instance, it is expedient at the beginning of the process to keep the sugar concentration around 7% and gradually decrease it to 3% in the course of the process. Then suddenly, at the end of the process, the concentration drops to nothing, which indicates that the reaction is at an end.

The batches of aqueous acid solution used in accordance with the previously-described process must be in a certain proportion to the size of the fill. I have found it expedient to use at the beginning an especially large batch, the size of which also depends on the moisture of the material. In the case of the first batch, a ton of dry wood substance should meet about 1 cbm. of liquid. The batch may be larger or smaller, depending upon whether the material is dry or moist. The subsequent batch may be two-thirds of the first batch. In the course of the process, the batch may be made continuously smaller, approximately in the same proportion as the fill of the percolator is diminished in volume by reason of the saccharification of the cellulose and the removal of the sugar. The interval between the smaller batches towards the end of the process may also become shorter.

In the instant process, applicant contemplates performing subsequent hydrolysis to form glucose by retarding the cooling.

The sugar solutions prepared in accordance with the present process have the peculiarity that subsequent hydrolysis is very effective in the first fractions, declines in the middle fractions, and is less successful in the final fractions. The subsequent hydrolysis is, therefore, carried out with advantage, especially with the first fractions. Subsequent hydrolysis may be effected in the lower part of the percolator by leaving the batch therein for a short time, say 30 minutes, at about 155° C. in the presence of 0.4% sulphuric acid.

Another procedure consists in withdrawing the liquid that is to be subsequently hydrolyzed from the percolator, cooling it to 100° C., and keeping it in this state in ordinary containers, for example, wooden vats. It is found that about the first tenth of the sugar obtained by means of decomposition requires, at 90° C.–100° C. and with a 0.4% concentration of sulphuric acid, a subsequent hydrolysis of about 24 hours, while in the case of the second and third tenths, there suffices a period that is shorter by several hours for each case. Obviously, the periods of the subsequent hydrolysis depend greatly on the temperature and also on the acid concentration. They are approximately in simple inverse proportion to the concentration of acid.

The subsequent hydrolysis may also take place in separate containers under pressure at temperatures above 100° C., for instance, by heating for

1 hour at 145° C. with an acid concentration of 0.4% sulphuric acid.

As previously explained, the subsequent hydrolysis is particularly advantageous with the first half of the sugar formed in the process. No important gain is secured by hydrolysis of the second half of the formed liquid. It may even be harmful. Consequently, the subsequent hydrolysis is confined to the first half of the sugar obtained.

After completion of the subsequent hydrolysis, neutralization, filtration and the cooling of the wort may take place immediately. The cooling of the wort may be carried out in the known manner by, for example, means of heat exchange with fresh water for the percolation.

I have found by my investigations that the harmful decomposition of the sugar is a function of the ratio between the time of exposure of the sugar and the total reaction of the cellulose. By practical operation, I have determined that absolute time of exposure of the sugar cannot be shortened at will. It has a natural maximum. Thus, the speed with which the sugar is removed from the particles and the percolator has natural limits. Experience has shown that by using judicious conditions of reaction (low temperature and acid) to permit the process to go on at so slow a rate that the time of reaction is more than 12 hours. The ratio between the time of exposure and the total reaction time is thereby made more favorable and the yield higher.

Another procedure of shortening the time of exposure of the sugar within the particles consists in the use of short-fiber material. When wood waste, such as shavings, etc., is not used, and logs, sticks and the like are cut up, it is expedient to cut the wood across the grain into small pieces, with short fibers of a length of a few millimeters only, the pieces having a diameter of 1 to 2 cm. When introduced into the percolator, these plates assume a horizontal position, the fibers being more or less vertical and in a position to be easily penetrated by the liquid flowing through the percolator. This shortens the intra-particular time of exposure.

Referring now to the drawing, wherein like reference numerals designate like parts, the reference numeral **1** designates a percolator of the common type having at the upper portion thereof means to supply the cellulose material and means to supply the dilute acids, as well as means to introduce steam. Since these features are well known, they have not been illustrated in the drawing. In the lower part of the percolator, there is provided means to introduce steam into the lower portion of the percolator. In the form illustrated, this means consists of an annular member **2**, mounted in any convenient manner between the end of the percolator **3** and the closure **4**. The ring **2** is provided with a continuous channel **5** which communicates with orifices **6** to form a nozzle-like structure directing any steam supplied thereto to the interior of the percolator. The steam is supplied from a suitable source of supply (not shown) by the feed pipe **7** and is controlled by the valve **8**.

It is also expedient to use for the purpose of steaming, besides the device previously mentioned, the filter arranged in the cone of the percolator, because this offers at the same time the advantage of cleaning the filter. The filter consists of acid-proof, porous filter stones made of granulated material, stone, resin or the like. The stones or bricks are made so that the smallest openings are on the inside. These filter stones lie on a supporting layer **10** which serves as a lining for the percolator. To prevent the filter stones from lying solidly on the lining **10**, it is advantageous to provide grooves or channels **11** therein as shown, for example, in the drawing. If desired, the lining may be made smooth, and the filter stones provided with raised parts, or a network may be arranged between them. This offers the advantage of easy passage for the steam which enters between the lining **10** and the filter stones **9**. It also offers the additional advantage that the acid passing through the filter stones easily finds its way to the outlet.

Underneath the filter stones, there are provided openings **12** to which pipes **13** are connected leading to an annular pipe **14**. The pipe **14** is connected to an outlet pipe **15**. A pipe **16** is also connected to the annular pipe structure **14**. The pipe **16** is provided with a stop valve **17** by means of which steam may be introduced into the pipe **16**, and hence into the pipe **14**, thence into the pipe **13**, through the outlet **12**, and into the interior of the cone. If desired, several outlets **12** may be provided. The drawing shows one illustrative embodiment utilizing a plurality of such outlets.

In the outlet connection **15**, there is provided an observation device **18** whereby one can ascertain whether acid liquid flows from the percolator and also to determine when the steam should be introduced. This observation device may, if desired, be also or alternatively provided in the pipes **13**. As the sugar wort tends to obscure the observation glasses and make them non-transparent, it is preferred to provide glass insertions **19** projecting into the interior of the pipe **15** which are continually washed by the wort flowing through said pipe in a manner to keep the glasses clean.

In actual practice, when the person watching at the observation point **18** has convinced himself that no more acid wort or no more batches flow from the percolator, either the valve **8** may be opened or, if both devices are provided, the valves **8** and **17**. The steam entering at **8** will then flow from the foot of the percolator and pass out of the nozzle **6** into the interior of the percolator while the steam coming from the pipe **16** passes through the annular pipe **14**, the pipes **13**, the openings **12**, and through the filter stones **9**, distributing itself through the channels **11** over the entire surface of the cone, penetrating the filter stones, and cleaning them at the same time during its passage to the interior of the percolator.

The process can be applied to all kinds of cellulose material, thus to wood, peat, etc. It may also be carried out with any desired acids and mixtures of acids as long as the acid solution has a sufficient concentration of hydrogen ions.

An illustrative example of the carrying out of the process with the arrangement that has been shown and with a percolator having a content of 20,000 liters is hereafter set forth:

Arrangement

Size of percolator: 20,000 liters content.

The percolator is of the type described in Patent No. 1,990,097, modified as herein described.

Used

5,000 kgs. of dry wood substance.

50 cbm. of water, 400 kgs. of 50% sulphuric acid (50 cbm. 0.4% sulphuric acid).

Test conditions

Running-in time is 5 minutes, running-off time and pause in working are each 20 minutes. Total reaction time, 14 hours. There are used 20 batches of 5 cbm. to 1.8 cbm., gradually decreasing. Temperature rising from 150° C. to 190° C.

Yields

In relation to dry wood substance: 53% of reducing sugar or 43% of fermentable sugar or 27 liters of spirits from 100 kgs. of dry wood substance.

Concentration of worts

5.3% of reducing sugar,
4.3% of fermentable sugar.

Since it is obvious that various changes and modifications may be made in the above description without departing from the nature or spirit thereof, this invention is not restricted thereto except as set forth in the appended claims.

I claim:

1. In a percolator for the saccharification of cellulose, a support in the lower portion thereof provided with inter-connecting grooves or channels, a layer of filter stones on said support, and means cooperatively connected with said grooves to withdraw the wort or introduce steam as desired.

2. In a percolator for the saccharification of cellulose, a support in the lower portion thereof provided with inter-connecting grooves or channels, a layer of filter stones on said support, said grooves being connected to a plurality of openings, means to withdraw the wort from the percolator through said openings, means to introduce steam into the percolator through said openings, and means permitting the use of the wort-withdrawal means or the steam-supply means with said openings as desired.

3. In a percolator for the saccharification of cellulose, a closure at the lower end thereof, an annular member disposed between said lower end and said closure, said annular member having a continuous channel connected to a plurality of orifices communicating with the interior of the percolator, and means to supply steam to the said annular member whereby it is introduced into the interior of the percolator.

4. In a percolator for the saccharification of cellulose, a closure at the lower end thereof, an annular member disposed between said lower end and said closure, said annular member having a continuous channel connected to a plurality of orifices communicating with the interior of the percolator, means to supply steam to the said annular member whereby it is introduced into the interior of the percolator, and a support in the lower portion thereof provided with interconnecting grooves or channels, a layer of filter stones on said support, and means cooperatively connected with said grooves to withdraw the wort or introduce steam as desired.

5. In a percolator for the saccharification of cellulose, a closure at the lower end thereof, an annular member disposed between said lower end and said closure, said annular member having a continuous channel connected to a plurality of orifices communicating with the interior of the percolator, means to supply steam to the said annular member whereby it is introduced into the interior of the percolator, a support in the lower portion thereof provided with inter-connecting grooves or channels, a layer of filter stones on said support, said grooves being connected to a plurality of openings, means to withdraw the wort from the percolator through said openings, means to introduce steam into the percolator through said openings, and means permitting the use of the wort-withdrawal means or the steam supply means with said openings as desired.

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