G. H. TOMLINSON.

PROCESS OF PRODUCING FERMENTABLE SUGARS.

APPLICATION FILED JUNE 13, 1917.

1,358,898. **Patented Nov. 16, 1920.**

Fig. 2.

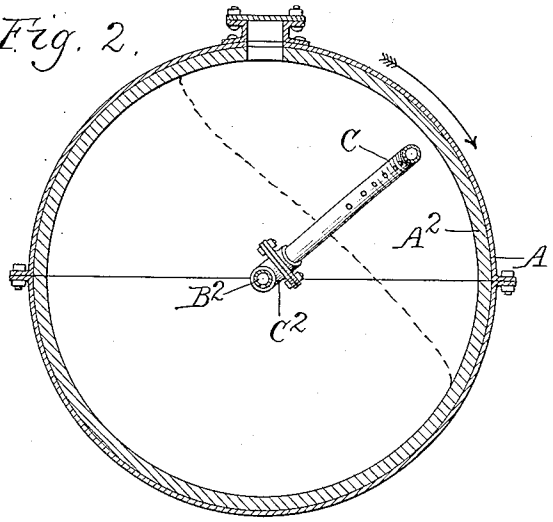
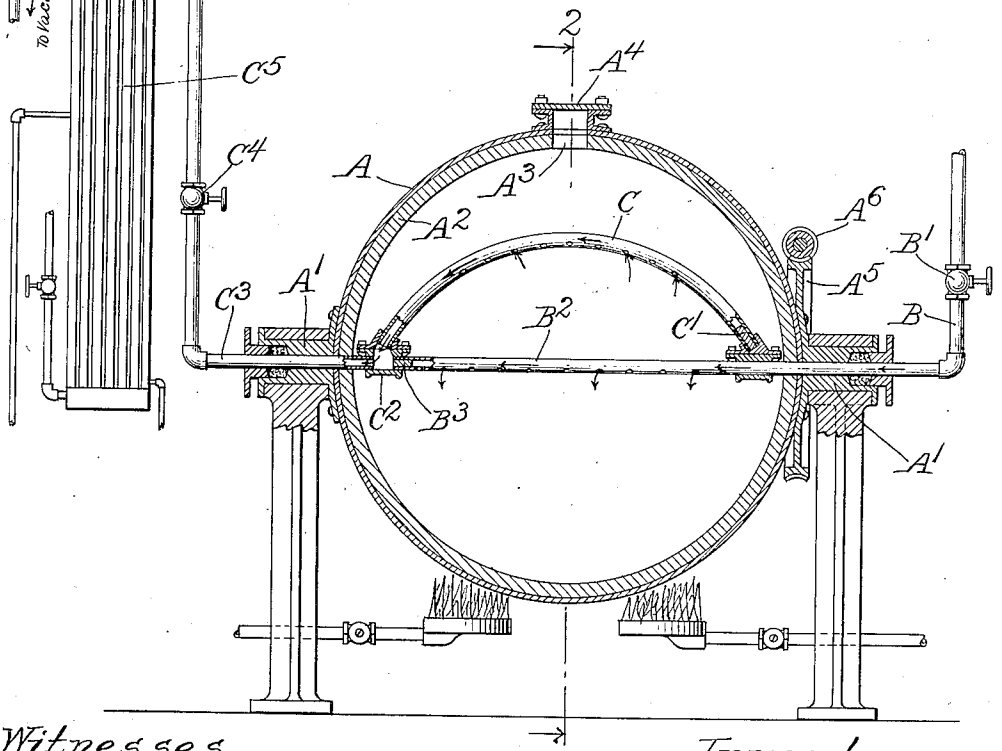


Fig. 1.



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UNITED STATES PATENT OFFICE.

GEORGE H. TOMLINSON, OF NIAGARA FALLS, ONTARIO, CANADA.

PROCESS OF PRODUCING FERMENTABLE SUGARS.

1,358,898. Specification of Letters Patent. Patented Nov. 16, 1920.

Application filed June 13, 1917. Serial No. 174,483.

To all whom it may concern:

Be it known that I, GEORGE H. TOMLINSON, a subject of the King of Great Britain, residing at Niagara Falls, in the county of Welland and Province of Ontario, Dominion of Canada, have invented a certain new and useful Improvement in Processes of Producing Fermentable Sugars, of which the following is a specification.

My invention relates to improvements in the process of producing fermentable sugars. It has for one object to provide a new and improved process of producing such fermentable sugars from wood waste as a step in the process of producing ethyl or grain alcohol from wood waste.

Another object of my invention is to provide a process for producing fermentable sugars on a commercial scale and to avoid the formation during the treatment of the wood waste of foreign substances which will interfere with the fermentation of the fermentable sugars or result in a deteriorated product.

This invention, therefore, generally relates to the production of fermentable sugar from sawdust, wood waste, or other cellulosic raw materials by the action of heat, in the presence of suitable hydrolyzing agents, by which sugars can be produced on a large scale and in a form readily permitting of their conversion into ethyl alcohol by the action of yeasts.

In the various processes heretofore proposed, it has been found, that whereas excellent yields have in many cases been obtained in laboratory or small scale working, that in such attempts as have been made to translate these into commercial practice, the yields obtained have been much lower, usually not exceeding fifty per cent. of that anticipated.

I have discovered, that in working large quantities of comminuted wood, such as saw dust (as for example several tons at a time) that as a result of the mass involved, so called secondary reactions occur, which are not met with when dealing with small quantities and therefore to successfully handle large quantities in a commercial way different conditions must be considered.

That is to say the mere fact that a conversion process is perfectly successful when carried out in a test tube or on a small scale laboratory basis is no real indication of the merit of the process when carried out on a large scale on a commercial basis and it frequently happens that processes which seem perfectly satisfactory in the laboratory are on account of the greater time and heat required or the decreased rapidity of the action on account of the large mass utterly unsuccessful.

The chemical re-actions which occur, when wood is strongly heated with dilute mineral acids, are extremely complicated and poorly defined, but the following general steps may be traced.

1st. Th. formation of soluble substances which may be termed polysaccharids. The formation of these is extremely rapid under a steam pressure of 100 pounds, and with an acid concentration of a $\frac{1}{2}$ per cent. or more. With common species of wood it reaches a maximum of 25 to 30 per cent., in a very few minutes but if cooled at once very little fermentable sugar is found.

2nd. By continuing the heating these intermediate products break down into dextrose in progressive amounts, depending upon the temperature. If the heating is continued at high temperatures decomposition into pyroligneous products such as tar, acetic acid, etc., also proceeds and the higher the temperature the more rapidly does this occur. Such decomposition products not only result in the loss of dextrose, but also through their presence retard the action of yeasts on the resulting product.

The conversion of the intermediate products or polysaccharids into dextrose proceeds at temperatures even below that of the boiling point of water but the rate of re-action at such a temperature is slow and it is therefore desirable to seek a temperature zone, in which the re-action will proceed at a rate compatible with the necessities imposed by commercial considerations, and at which decomposition products are formed only in such quantities as may be tolerated. The product so formed while consisting mainly of dextrose, will also contain a great variety of organic compounds in less amounts, many of which have not been identified; and some of which seem to be of an unstable or changing character.

In the processes so far proposed the material is treated usually under pressure for various periods of time, with varying quantities of acid, and after "blowing off," that is reducing the pressure to atmospheric, the reactions are considered complete. Such is

more or less the case in small scale working when the mass is small and its cooling consequently rapid from natural conditions. When handling large masses, however, as for example several tons at a charge such cooling does not occur quickly since the mass will, if allowed to stand retain a temperature approximating 212° Fahrenheit for many hours, and in the usual methods of working part or all will retain such a temperature for a considerable interval of time, during which secondary re-action may occur, which cannot be controlled on account of the unstable character of some of the products already formed. This condition results in great lack of uniformity in results where alcohol is the end product desired.

The above explanation is inserted to illustrate the complicated nature of the re-actions which are involved in carrying on the conversion into dextrose on the commercial scale. My invention consists in cooking the comminuted wood under pressure with a dilute mineral acid, as for example sulfuric or sulfurous acid, in such amount and at such pressures as may seem best fitted to the commercial considerations imposed, and then quickly reducing the pressure by exhausting to the atmosphere, then creating a vacuum within the digester until the temperature is so far reduced that such secondary re-actions cease. In this way the temperature may be quickly and uniformly reduced to 100° F. or lower if desired.

By following this procedure in addition to obviating any such secondary re-actions as have been described certain other advantages accrue which are of inestimable value.

1st. As has been shown no matter how carefully the cooking is carried out under commercial conditions, appreciable quantities of pyroligneous products are formed, which if not removed have the effect of inhibiting fermentation to a greater or less extent, depending upon the quantity present. By continuing the ebullition under vacuum, as described such products can be almost entirely removed, since they are of a volatile character, or have the property of distilling over with steam. This results in a product which is more readily fermentable and permits of carrying the reaction to greater completion than otherwise permissible.

2nd. The amount of water in the resultant mass is reduced through further evaporation which occurs, and the concentration of the sugars in the product is thus increased.

3rd. The material is left cool and in a condition which permits of its being more readily handled than is otherwise possible. Heretofore the handling of the digested product through subsequent steps in a hot state has involved unusual difficulties in conveyer construction which are now removed.

An apparatus for carrying out my process is illustrated more or less diagrammatically in the accompanying drawing wherein—

Figure 1 is a digester and associated parts part in elevation and part in section.

Fig. 2 is a section along line 2—2 of Fig. 1.

A is a preferably spherical metallic digester rotatably mounted upon the trunnion bearings A^1 and having an acid resisting lining A^2 and a filling opening A^3 closed by a suitable steam tight door A^4. A^5 is a worm wheel mounted rigidly upon the digester and A^6 is a worm in mesh with the wheel adapted to rotate the digester about its trunnion bearings.

B is a steam supply pipe fed with steam under pressure from any suitable source not here shown controlled by a valve B^1. This steam supply pipe discharges inwardly through trunnion bearings into the digester where it communicates with a perforated steam discharge pipe B^2 which pipe is closed at its inner end as at B^3.

C is a blow off perforated pipe, upwardly curved above the pipe B^2, its free end being mounted on the pipe B^2 and being closed at C^1. C^2 is an elbow mechanically connected to the end of the pipe C and communicates with a pipe C^3 discharging outwardly through one of the trunnion bearings. This pipe C^3 controlled by a valve C^4 discharges into a condenser C^5 which may be controlled and operated in any usual manner not specifically illustrated here because not part of my invention.

Referring more specifically to Fig. 2 the level of the sawdust or wood pulp in the digester is shown in dotted lines and the direction of rotation is shown by an arrow. It will thus be noted that the pipe C is at all times above the level of the material in the digester whereas the pipe B^2 is below it. Thus the pressure may be blown out through the perforations in the pipe C without any difficulty or any cloging of the perforations therein.

It is understood, of course, that my showing is exceedingly diagrammatic. Many forms of digesters, condensers controlling and operating means might be used but the form which I have illustrated is a satisfactory one and will do to illustrate any suitable means for carrying out my process.

I have mentioned the desirability of exhausting from the condenser quickly. This, of course, means that pressure should be reduced without needless waste of time and the time taken might consume from 5 to 30 minutes without causing any appreciable difference in result.

It will be understood that if the reaction is carried to practical completion at from 75 to 330° F. thus producing a sufficient

amount of pyroligneous products, these products might be sufficiently removed to permit of excellent results by carrying on the distilling operation under vacuum or at least under reduced pressure for a long enough period to remove substantially all of such products. It will be understood, of course, that the volatile impurities may be distilled without maintaining reduced pressure in the condenser. Such a procedure might take the form of blowing off steam under more or less pressure. This would have the disadvantage that the temperature would not be reduced, at least not as much as if an actual vacuum were used, such a process might nevertheless be perfectly satisfactory.

The use and operation of my invention is as follows:—

The digester is first filled with sawdust or other cellulosic material preferably in a finely comminuted or broken up condition. This material is introduced through the man hole in the usual way and may be accompanied by an acid if desired. The man hole is then closed and the digester is rotated about its axis. The steam is then admitted by manipulating the control valve until the pressure and temperature is raised to that point at which a rapid conversion of the ligno-cellulose takes place. The acid, if desired, may, of course, be introduced with the steam or in any other suitable way and the temperature may be raised by means other than the introduction of steam if desired. The essential thing is to raise the temperature by any suitable method as suggested, for instance, if not steam by fire as indicated in the drawings until the proper temperature for the reaction is reached.

I have found that a desirable way of operating is to raise the temperature quickly to approximately 300° F., hold it at this temperature for fifteen minutes and then to reduce the temperature by reducing the pressure to about 250° F. and maintaining this temperature for from thirty minutes to an hour though it will be obvious that with different raw materials both time and the temperature may frequently need to be radically changed, the change depending upon the concentration and kind of acid used and the kind and condition of the raw material treated.

When this cooking or treating process has gone on long enough the steam supply or other heating means is discontinued and the pressure on the material is reduced by opening the discharge connection in the condenser. This results in further reducing the temperature and pressure as the condenser operates and the mass of material in the digester may thus be thoroughly cooled.

Evidently if steam is not introduced, the material must be moistened by other means as for instance the introduction of water with the material. This is necessary because the chemical action will not take place except in a moist atmosphere. Whether steam is introduced to heat the mass or it is heated by other means, steam will be present and the operation of the condenser will cause a reduction of pressure and temperature. This is obviously the case as the steam is in a confined space and when withdrawn or allowed to escape, both pressure and temperature used for the mass must drop.

It will be understood then that the preferable method, both on account of satisfactory operation and for other reasons, is to operate by heating the digester and contents with heat introduced from a boiler or other suitable source.

The operation of the condenser suddenly decreasing the pressure and temperature causes a sudden radical decrease in temperature throughout the entire contents of the digester and this temperature reduction is, of course, expedited by the continuous stirring which takes place in the digester as it rotates about its axis. The reduction in pressure below atmospheric causes the volatile foreign substances to be distilled off and under some circumstances it may even be desirable to introduce more steam and pass the steam through the digester to assist in this distilling off of the volatile products which would otherwise prevent the formation of fermentable sugars.

In the specification and claims I have pointed out more or less generally time intervals during which parts of the process can or should go on. It is obvious of course that I am merely suggesting approximate times and that so long as the actual time does not vary too widely from the time suggested the process will go on with desirable results, and the periods stated are to be regarded in the nature of practical limits within which good results will occur.

I claim:—

1. The process of producing fermentable sugars from cellulosic material which consists in heating it in a closed vessel in contact with a hydrolyzing agent until fermentable sugars are formed and then quickly reducing the pressure in the vessel below the atmospheric pressure and distilling off the volatile products of the reaction.

2. The process of producing fermentable sugars from cellulosic material which consists in heating it in a closed vessel in contact with a hydrolyzing agent until fermentable sugars are formed and then quickly reducing the pressure in the vessel below the atmospheric pressure and distilling off the volatile products of the reaction and continuing this process until such products are all of them substantially removed from the mass.

3. The process of producing fermentable sugars of ligno-cellulose which consists in introducing the material in a closed vessel in contact with a hydrolyzing agent, raising the temperature quickly to approximately 300° F., holding this temperature for fifteen minutes, then reducing it to about 250° F., holding this temperature for from thirty minutes to an hour and then suddenly reducing the pressure and temperature to a point below that at which reaction takes place and subjecting the material in treatment to a vacuum.

4. The process of producing fermentable sugars of ligno-cellulose which consists in introducing the material in a closed vessel in contact with a hydrolyzing agent, raising the temperature quickly to approximately 300° F., holding this temperature for fifteen minutes, then reducing it to about 250° F., holding this temperature for from thirty minutes to an hour and then suddenly reducing the pressure and temperature to a point below that at which reaction takes place and distilling off the volatile products of the reaction and subjecting the material in treatment to a vacuum.

5. A process which consists in treating cellulose materials with a dilute hydrolyzing agent in the presence of steam under pressure in a closed vessel at temperatures between 212° Fahrenheit and 320° Fahrenheit, then releasing the pressure, and thereafter placing the mass under a vacuum to substantially reduce the temperature to prevent further reaction.

6. A process which consists in treating ligno cellulose or other cellulose materials with a hydrolyzing agent such as dilute sulfuric or sulfurous acid in the presence of steam under pressure in a closed vessel at temperatures between 212° Fahrenheit and 320° Fahrenheit, then releasing the pressure, and thereafter placing the mass under a vacuum to reduce the temperature.

7. A process of converting cellulose or ligno cellulose materials into fermentable sugars, which consists in placing a quantity of such material in a digester in the presence of a dilute hydrolyzing agent, introducing steam into the vessel and holding it at temperatures between 212° Fahrenheit and 320° Fahrenheit until the desired reaction is complete, and then reducing the temperature by means of a vacuum to prevent further reaction.

8. A process of converting ligno cellulose or other cellulose materials into fermentable sugars, which consists in placing a quantity of such material in a digester in the presence of dilute sulfuric or sulfurous acid, introducing steam into the vessel and holding it at temperatures between 212° Fahrenheit and 320° Fahrenheit until the desired reactions are complete, then releasing the pressure, and thereafter placing the mass under a vacuum to reduce the temperature and retard further reaction.

9. A process of converting ligno cellulose or other cellulosic materials into fermentable sugars, which consists in placing a quantity of such material in a digester together with approximately 1% to 2% of a hydrolyzing agent, such as sulfuric or sulfurous acid, heating the mass at temperatures between 212 degrees and 320 degrees Fahrenheit until the desired reactions are complete, then releasing the pressure, and thereafter placing the mass under a vacuum to reduce the temperature of the mass below that at which further reactions will proceed.

10. The process of producing fermentable sugars from cellulose materials which consists in heating the material under pressure at temperatures above the boiling point in a closed vessel in contact with a dilute hydrolyzing agent until the reaction is complete and then reducing the pressure below atmospheric and the temperature below the point at which reaction will take place.

In testimony whereof, I affix my signature in the presence of two witnesses this 28th day of May, 1917.

GEO. H. TOMLINSON.

Witnesses:
R. E. CHAPMAN,
A. KUNKENGER.