Patented June 29, 1943

2,323,022

UNITED STATES PATENT OFFICE

2,323,022

PROCESS FOR SACCHARIFYING CELLULOSE

Demetrio Ferrari and Mario Torresi, Rome, Italy; vested in the Alien Property Custodian

No Drawing. Application February 18, 1941, Serial No. 379,480. In Italy March 2, 1940

1 Claim. (Cl. 127—37)

In order to obtain sugary substances from cellulose and all the other substances containing cellulose, various processes and methods have been applied, all predicated on the hydrolizing power exercised by mineral acids on such cellulosic substances.

One of the most important known processes, which have been realised on an industrial scale, works with diluted sulphuric acid at a high temperature and pressures, it requires complicated preparatory dispositions and yields sugars at around the 60% of the theoretical output. Another of these processes attacks the cellulosic substances with superconcentrated hydrochloric acid (40–42%), obtaining high yields, but, on the other hand, it requires very expensive installations and is difficult to carry out. There are further processes which work by firstly attacking the starting material in a warm condition with diluted sulphuric acid, and then, in a second cycle, with concentrated sulphuric acid, giving satisfactory results.

In the process according to the present invention we solve the problem of saccharifying cellulosic substances with a consumption of sulphuric acid inferior to that till now necessary for the realisation of the above mentioned processes which employ diluted and concentrated sulphuric acid and proceed to the attack of the cellulosic material in two separate and consecutive cycles. At the same time the process of our invention reduces the consumption of steam considerably, although obtaining yields in sugar if not better at least identical to those of the processes above mentioned which employ mineral acids. In fact, the total consumption of sulphuric acid has been reduced by employing sulphuric acid at 60° Bé. and simultaneously reducing the quantity of steam necessary for completing the hydrolysis of about 30% with respect to already known processes; thus remarkably simplifying the necessary preparatory working disposition and at the same time lowering the cost of each installation unit.

A further fundamental characteristic of the process according to our invention lies in the fact that the cellulosic material must be mechanically crushed and triturated before the acid attack.

Until now it had been retained impossible to attack cellulosic materials crushed and triturated beyond a certain limit, owing to the impossibility of removing the heat which is developed by the attack with sulphuric acid and which, with its reaction speed, may endanger the normal development of the process.

After numerous experiments we have ascertained that it is possible to proceed to the attack of the material crushed and disintegrated to the consistency of wood meal, provided such a trituration be accomplished by an extremely slow simultaneous sprinkling of sulphuric acid, the operation being preferably accomplished in closed containers. This attack with sulphuric acid of the ligneous material produces a very high saccharification of the same without all the alarming inconveniences exhibited by other processes.

In a preferred embodiment of the process according to the present invention one hundred parts in weight of ligneous material are mechanically crushed and comminuted in a closed container and brought to the consistency of wood meal of the order of granulometric size employed to stamp plastic substances, the mechanical crushing being accompanied by a very slow and fine sprinkling of sulphuric acid at 60° Bé. This operation takes, according to the ligneous starting material used, from two to three hours, from 50 to 60 parts of sulphuric acid at 60° Bé. being employed in all. After this operation the paste obtained is poured into closed or open maturation vats, and left to rest for a more or less long period, then it is kneaded at a great pressure in a closed container. During this second kneading at a high pressure cold water is added up to a quantity equal to that of the acid employed.

The dense liquid thus obtained, after a treatment with steam, having the object of completing the hydrolysis and removing all that may be removed by distillation in a steam current (furfurol and the like), is diluted by means of about four parts of filtered and neutralised water. The filtrate thus obtained presents a contents in sugars reaching even the 95% of the theoretical output.

The following explicative example will aid a better understanding of the invention.

100 kg. of straw are preventively mechanically crushed and then triturated until the straw is reduced to a powder having a grain of the same size as that of the wood meal used to be mixed with synthetic resins for stamping. This is the fundamental condition to obtain the desired results.

The powder thus obtained is conveyed to another machine where, by means of mechanical pressure (grinding or similar) or by means of the vacuum, it is kneaded for half an hour with a quantity of sulphuric acid at 60° Bé. corresponding to 15 kg. During this operation the greatest care is required by the mechanical part,

as both grinding and vacuum have the function of allowing the penetration of the sulphuric acid right into the very inside of the minute particles of straw powder.

It is to be remarked that the sulphuric acid should not be mixed with the straw powder rapidly, but very slowly, in order to avoid a partial destruction of the material under process due to the increase of temperature produced by the quick reaction.

In this operation better results have been obtained by conveying the material into a special apparatus provided with an internal stirring device, and wherein a considerable vacuum may be attained, rather than by grinding, as this apparatus allows firstly the removal of even the slightest traces of air and dampness inside the ligneous particles, and at the same time a complete imbibition of the sulphuric acid inside the material.

After this operation the acid material is diluted with four hundredweight of water and conveyed to a pressure reservoir where it is subjected to a first cooking for a quarter of an hour under a pressure of from three to four atmospheres. During this cooking in a steam current the furfurol and the acetic acid are extracted.

At the end of this operation the pressure reservoirs are discharged and all their contents is conveyed to hydroextractors which separate the solid part from the liquids already containing from 20 to 22 kg. of carbon hydrates.

The solid part discharged from the hydroextractors and which has been reduced to about 60 kg. (calculating with reference to the dry material) is then put, after preventive desiccation (the maximum dampness remaining should be from 5 to 8%) into maturation chests where it remains from 8 to 10 hours at about 100° C.

When this operation is over the material is again conveyed to be ground under a very high pressure and simultaneously 40–45 kg. more sulphuric acid are sprinkled over the same concentrated at 60° Bé. (this operation should last about two hours).

The black paste obtained in the above mentioned operation, diluted in twice its weight in water, is again conveyed to the pressure reservoir, where it is subjected to a further cooking at 3–4 atmospheres for a quarter of an hour.

At the end of the cooking the creamy liquid in the pressure reservoir is filtered again so that a solid residue (lignin) and liquids containing further 12–14 kg. of carbon hydrates are obtained.

Conclusively, in the treatment of a hundredweight of straw with contents of about 38% cellulose and 15% dampness, the following results have been attained.

Semi-industrial installation		Installation of experimental laboratory which may treat 100 kg. of material daily	
	Kg.		*Kg.*
Straw (cellulose 38%, dampness 15%).	100	Straw (cellulose 38%, dampness 15%).	100
Sulphuric acid at 60° Bé.	55–60	Sulphuric acid at 60° Bé.	55–60
Carbon hydrates (with reference to the dry substance).	37–38	Carbon hydrates (with reference to the dry substance).	34–36
Furfurol (with reference to the dry substance).	1,800–2	Furfurol (with reference to the dry substance).	1,600–1,800
Acetic acid (with reference to the dry substance).	0,800–1	Acetic acid (with reference to the dry substance).	0,700–0,800
Lignin	40–45	Lignin	40–45

It is clear that alterations may be introduced in practice to the process disclosed above without infringing the limits of protection or the gist of the present invention as defined by the following claim.

What we claim is:

A process for saccharifying cellulosic material comprising the steps of comminuting the cellulosic material mechanically to the consistency of wood meal, agitating said wood meal for about half an hour while very slowly adding thereto about 15% by weight of sulphuric acid at 60° Bé., diluting the resulting mixture with 400% by weight of water, cooking the diluted mixture for a quarter of an hour under a pressure of from three to four atmospheres, extracting the furfurol and acetic acid, separating the sugar-containing liquid from the solid parts of the mixture, drying the solid residuum and retaining it for from 8 to 10 hours in a maturation chest at a temperature of about 100° C., grinding the maturated material under very high pressure for about two hours while gradually admixing thereto between about 66.6 and 75% by weight of sulphuric acid at 60° Bé., diluting the black paste obtained with twice its weight of water, cooking the mixture for a quarter of an hour under a pressure of from 3–4 atmospheres, and filtering the resulting creamy liquid to separate the sugar-containing liquid from the solid lignin residuum.

DEMETRIO FERRARI.
MARIO TORRESI.