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2. European patent number or publication
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0649436



3. Full name and address of the or of each
applicant for or proprietor of the
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Patents ADP number (if you know it)

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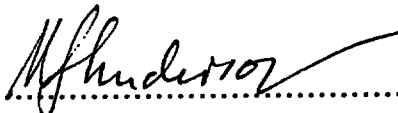
MR. DAVID ALCOCK
0171-831 7929

In the matter of European Patent
No. 0649436
granted on European Patent
Application No. 94913441.5
in the name of LENZING AKTIENGESELLSCHAFT

DECLARATION

I, Noel George Anderson, BSc, PhD, FRSC, CChem, MITI, FIL, BDÜ, of 73 Hawthorne Avenue, Eastcote, Ruislip, Middlesex, HA4 8SR, hereby certify that to the best of my knowledge and belief the following is a true translation made by me, and for which I accept full responsibility, of European Patent No. 0649436 granted on European Patent Application No. 94913441.5
in the name of LENZING AKTIENGESELLSCHAFT

Signed this 13th day of March 1996

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The present invention relates to a stable moulding material and spinning material containing cellulose and an aqueous tertiary amine oxide. The present invention furthermore relates to the use of specific stabilizers for the stabilization of cellulose dissolved in a tertiary amine oxide.

US-PS 2,179,181 discloses that tertiary amine oxides are capable of dissolving cellulose and that cellulosic fibres can be obtained by precipitation from these solutions. A process for preparing such solutions is disclosed, for example, in EP-A-0 356 419. According to this publication, a suspension of cellulose is first prepared in an aqueous tertiary amine oxide. The amine oxide contains up to 40% by mass of water. The aqueous cellulose suspension is heated and the water is distilled off under reduced pressure until the cellulose dissolves.

During the dissolution and also during the storage of the solution at elevated temperature, a degradation of the cellulose and of the solvent occurs. The degradation products may discolour the moulded bodies, for example fibres and films, obtained after precipitation to an appreciable extent and are recovered during the recovery of chemicals only at appreciable expense. The latter is necessary, however, to avoid polluting the environment.

EP-A-0 047 929 recommends the use of propyl gallate (PG) as stabilizer. However, it has been found that the stabilizing action of this compound persists only for a relatively short time, with the result that a heated cellulose solution has to be processed rapidly. In addition, PG is expensive and also its solubility in polar solvents, such as water, leaves something to be desired.

The object of the invention is to provide a stabilizer

which does not have the above disadvantages and which can stabilize the dissolved cellulose against degradation for a longer period of time than PG.

- 5 The object mentioned is achieved, according to the invention, in that a mixture of glucosides of gallic acid and of m-digallic acid is used to stabilize the cellulose against degradation.
- 10 The invention furthermore relates to a stable moulding material or spinning material containing cellulose, a tertiary amine oxide, especially N-methylmorpholine N-oxide, optionally water and a stabilizer, wherein a mixture of glucosides of gallic acid and of m-digallic
- 15 acid is provided as the stabilizer.

A preferred embodiment of the stable moulding material or spinning material according to the invention contains tannic acid as the stabilizer.

- 20 According to pharmacopoeias, a mixture of esters of D(+)-glucose with gallic acid and m-digallic acid is described as tannic acid [CAS 1401-55-4]. Preferred are substances in which all the alcoholic OH groups of the glucose are
- 25 esterified with gallic acid and m-digallic acid, the m-digallic acid best forming a proportion of 40% to 90% of the sum of gallic acid and m-digallic acid.

- 30 Tannic acid is also contained in the so-called tannins which, according to the invention can also be used as stabilizers. A further preferred embodiment of the stable moulding material and spinning material according to the invention consequently contains tannin as stabilizer.

- 35 Tannins are products which are used for tanning on an industrial scale. A distinction is made between vege-

table and synthetic substances, the vegetable products being obtained extractively from natural substances such as woods and plants.

5 According to the invention, it has been found that both vegetable tannins and tannic acid itself have good stabilizing properties for cellulose in tertiary amine oxides. It was found that, under the conventional conditions of solution preparation, the degradation of
10 cellulose is prevented and a reduced decomposition of the tertiary amine oxide is observed.

It has been found that very good results can be achieved with the stabilizers used according to the invention even
15 in a concentration of a maximum of 0.05% by mass (based on the cellulose). This concentration is below that which is recommended for propyl gallate in the prior art. A particularly preferred embodiment of the moulding material or spinning material according to the invention
20 consequently contains the stabilizer in a concentration of a maximum of 0.05% by weight, based on the weight of cellulose.

The invention is explained in still greater detail by the
25 example below.

Example

An aqueous solution containing 70% by mass of NMMO was
30 used for solution preparation. Cellulose was added to said NMMO solution and water was evaporated off in a laboratory kneader at a pressure of 250 mbar and a temperature of approximately 95°C and the cellulose was dissolved. The amounts were chosen so that the solution
35 obtained contained 10% by mass of cellulose, 15% water and 75% NMMO.

The viscosity of the solution was measured 30 minutes and 750 minutes after the dissolution of the cellulose by means of a Bohlin rheometer (measurement temperature: 120°C). The results obtained are reported in the table below as "comparison".

The above procedure was then repeated twice, but PG (concentration: 0.05%, based on cellulose). or tannic acid (concentration: 0.05%, based on cellulose; tannic acid supplied by Aldrich Chemicals) was added as stabilizer. The results obtained are also reported in the table below.

Table

Stabilizer	Viscosity (Pa·s)	
	after 30 min	after 750 min
Comparison (without stabilizer)	753	18
PG	985	383
Tannic acid	985	755

From the table it can be seen that, if PG is used as stabilizer, the viscosity of the solution has decreased to an appreciably lower value than if tannic acid is used. This means that tannic acid stabilizes the cellulose better against degradation than PG during a time of 750 min (= 12 hours 30 minutes).

Similarly good results were also obtained with other mixtures of glucosides of gallic acid and of m-digallic acid.



CLAIMS

1. Stable moulding material or spinning material containing cellulose, a tertiary amine oxide, especially
5 N-methylmorpholine N-oxide, a stabilizer and optionally water, characterized in that a mixture of the glucosides of gallic acid and of m-digallic acid is provided as the stabilizer.
- 10 2. Stable moulding material or spinning material in accordance with Claim 1, characterized in that tannic acid is provided as the stabilizer.
- 15 3. Stable moulding material or spinning material in accordance with Claim 1, characterized in that tannin is provided as the stabilizer.
- 20 4. Stable moulding material or spinning material in accordance with one of the Claims 1 to 3, characterized in that the stabilizer is present in a concentration of a maximum of 0.05% by weight, based on the weight of cellulose.
- 25 5. Use of a mixture of glucosides of gallic acid and m-digallic acid for the stabilization of a moulding material or a spinning material containing cellulose and a tertiary amine oxide.