The present invention relates to improvements in the flotation process of clarifying regenerating baths used in the preparation of cellulose articles of artificial origin. More specifically it has to do with carrying out of the process in the presence of a betaine in the regenerating bath.

The primary object of this invention is to provide a composition of regenerating medium which will allow an economical and efficient application of the flotation process and which will be entirely free from deleterious effects upon the extrusion step. The application of this invention to the viscoso process will be described in detail in the following specification, but it will be understood that the scope of the invention is intended to embrace any process of regenerating cellulose solutions in which the regenerating medium, in general a liquid bath, becomes fouled with impurities or by-products inherent to the extrusion step, such as, for example, the cuprammonium process.

In accordance with the applicants' invention the process of purifying the regenerating bath by flotation, which has been the subject of several United States Letters Patents, including Nos. 2,105,684 and 2,153,449, is carried out in the presence of an acid stable frothing agent, which compound is characterized by the following general formula:

\[
\begin{align*}
R_1 & \rightarrow \neg \neg \neg \neg \\
R_2 & \rightarrow \neg \neg \neg \neg \\
R_3 & \rightarrow \neg \neg \neg \neg \\
R_4 & \rightarrow X
\end{align*}
\]

wherein \( N \) is a nitrogen atom, \( R_1 \) is an alkyl group of from 3 to 20 carbon atoms, \( R_2, R_3 \) and \( R_4 \) are alkyl groups at least one of which must contain from 8 to 20 carbon atoms in the event that \( R_2 \) contains less than 5 carbon atoms, and \( X \) represents the residue of a carboxyl or sulfonic acid group. Such compounds may be classed generally as betaines.

By residue of a carboxyl is meant the configuration

\[
\begin{align*}
\text{O} & \rightarrow \neg \neg \neg \neg \\
\text{O} & \rightarrow \neg \neg \neg \neg \\
\text{O} & \rightarrow \neg \neg \neg \neg 
\end{align*}
\]

resulting from the loss of the ionizable \( H^+ \) from the carboxyl group in consequence of the neutralization of the latter upon the nitrogen atom. Likewise, by residue of a sulfonic acid group is meant the configuration

\[
\begin{align*}
\text{O} & \rightarrow \neg \neg \neg \neg \\
\text{O} & \rightarrow \neg \neg \neg \neg \\
\text{O} & \rightarrow \neg \neg \neg \neg 
\end{align*}
\]

resulting from the loss of the ionizable \( H^+ \) from the sulfonic acid group in consequence of the neutralization of the latter upon the nitrogen atom.

Applicants have found that the presence of minute quantities of compounds of the above structure makes possible the production of a copious froth by the use of flotation machinery of conventional design even when dealing with regenerating baths of the usual high electrolyte concentrations particularly those containing acid. This result is not obtainable by the use of the usual flotation reagents in the presence of high electrolyte and acid concentrations. Moreover it has been found that such substances are excellent collectors of the sulfur and other impurities usually found in precipitating solutions so that the flotation process is very efficient without the further addition of substances which might be deleterious to the extrusion step.

A further advantage exhibited by the compounds of the structure set forth above is their freedom from forming precipitates with the heavy metals which are usually present in regenerating baths, for example, zinc and lead. An important source of loss of compounds used in minute quantities is thus prevented.

A still further advantage of such compounds is their lack of pronounced ionogenic tendencies.

The importance of this property will be shown below: The efficiency of the flotation method as a whole for the clarification of rayon regenerating baths depends in no small part upon the prevention of accumulation of solid impurities at their point of inception, i.e., at, and even in the spinnneret orifices themselves. Since the formation of solid impurities undoubtedly takes place at or near the boundary between the strongly alkaline spinning solution and the highly acid regenerating bath, it is obvious that a surface active agent substantially neutral in character will have the best likelihood of being present in the critical zone of precipitation of such impurities. As a result of the presence of compounds of the structure disclosed above the precipitated solid particles of impurities are prevented from agglomerating or adhering to the spinnneret surfaces and are rendered hydrophobic in character, so as to be readily removed by the flotation process. In other words the special virtue of these compounds is that they are strongly polar but with little or no tendency to flocculate.

In this connection it should be pointed out that many of the surface active agents heretofore employed for attaining the above mentioned ends,
as exemplified by the cation active compounds, have a marked affinity for cellulose, which property results in a high rate of exhaustion by the continuous production of a fresh cellulose surface as the extrusion process is carried out over a period of time.

The invention will be better understood by citing the following specific examples:

I. (a) In a commercial operation a regenerating bath containing approximately 10% sulfuric acid and 20% sodium sulfate by weight was treated continuously with an amount of C-cetyl trimethyl betaine that would give approximately 0.00005% of the bath being circulated from the storage and heating tanks over the spinning frames and through a flotation machine of conventional design before returning to the storage tanks. It is estimated that under such conditions a concentration of the C-cetyl trimethyl betaine was maintained at a level of approximately 50% above the ratio at which it was added owing to the incomplete removal of the frothing agent in the flotation step. Under such conditions the amount of suspended impurities in the spinning bath was reduced from 18 P.P.M. to 5 P.P.M. by a single pass through the flotation machine, the concentration of suspended impurities being determined by turbidimetric records which previously had been calibrated gravimetrically.

(b) In a continuation of the operation described above the addition of the C-cetyl trimethyl betaine was doubled with the following results: The average suspended matter was reduced from 14 P.P.M. to 2 P.P.M. during a single pass through the flotation machine.

(c) In a further continuation of the operation the addition of the C-cetyl trimethyl betaine was trebled over that used in I (a), i.e. brought to approximately 0.0001%, with the following results: The suspended impurities were reduced from 13 P.P.M. to 2 P.P.M. in a single pass. However, the amount of froth was excessive under these conditions resulting in difficult manipulation of the flotation machine and the loss of considerable quantity of the spinning bath. It is concluded from the above tests that the optimum amount of this frothing agent in a bath of approximately the composition cited would be near to that mentioned in Example I (b).

II. In a similar manner the compound C-myristyl trimethyl betaine was tested at varying rates from 0.00003% to 0.00009%. The results in this case were substantially the same except that the optimum concentration was found to lie slightly higher than that of the C-cetyl betaine above.

It will be understood that the above examples were given for illustration only and the applicants do not wish to be limited to the scope of the examples cited.

We claim:

1. In the froth flotation process of clarifying acid regenerating baths for repeated use in the preparation of cellulose articles from viscose, which baths are fouled by impurities incident to the regeneration step, the step of floating said impurities out of the bath in a froth formed in the presence of a betaine of the following general formula:

\[ R_N - R_1 \]

wherein \( N \) is a nitrogen atom, \( R_1 \) is an alkyl group of from 3 to 20 carbon atoms, \( R_2, R_3 \) and \( R_4 \) are alkyl groups at least one of which must contain from 8 to 20 carbon atoms in the event that \( R_1 \) contains less than 5 carbon atoms, and \( X \) represents the residue of a sulfonic acid group.

2. In the froth flotation process of clarifying acid regenerating baths for repeated use in the preparation of cellulose articles from viscose, which baths are fouled by impurities incident to the regeneration step, the step of floating said impurities out of the bath in a froth formed in the presence of a betaine of the following general formula:

\[ R_N - R_1 - R_2 - N - R_3 \]

wherein \( N \) is a nitrogen atom, \( R_1 \) is an alkyl group of from 3 to 20 carbon atoms, \( R_2, R_3 \) and \( R_4 \) are alkyl groups at least one of which must contain from 8 to 20 carbon atoms in the event that \( R_1 \) contains less than 5 carbon atoms, and \( X \) represents the residue of a carboxyl group.

3. The process as described in claim 2 in which the betaine is C-cetyl trimethyl betaine.

4. The process as described in claim 3 in which the C-cetyl trimethyl betaine is present in a concentration of from 0.00003% to 0.0001%.

5. The process as described in claim 2 in which the betaine is C-myristyl trimethyl betaine.

6. The process as described in claim 5 in which the C-myristyl trimethyl betaine is present in a concentration of from 0.00003% to 0.00008%.

JOSEPH L. COSTA
WILLIAM HARLOW KAHLER.

REFERENCES CITED
The following references are of record in the file of this patent:

**UNITED STATES PATENTS**

<table>
<thead>
<tr>
<th>Number</th>
<th>Name</th>
<th>Date</th>
</tr>
</thead>
<tbody>
<tr>
<td>2,074,380</td>
<td>Piett</td>
<td>Mar. 23, 1937</td>
</tr>
<tr>
<td>2,105,654</td>
<td>Costa</td>
<td>Jan. 18, 1938</td>
</tr>
<tr>
<td>2,145,527</td>
<td>Polak</td>
<td>Jan. 31, 1939</td>
</tr>
<tr>
<td>2,153,449</td>
<td>Booth</td>
<td>Apr. 4, 1939</td>
</tr>
<tr>
<td>2,221,485</td>
<td>Kirby</td>
<td>Nov. 12, 1940</td>
</tr>
<tr>
<td>2,274,551</td>
<td>Kenyon et al.</td>
<td>Feb. 24, 1942</td>
</tr>
<tr>
<td>2,274,658</td>
<td>Booth</td>
<td>Mar. 3, 1942</td>
</tr>
<tr>
<td>2,310,208</td>
<td>Bley</td>
<td>Feb. 9, 1943</td>
</tr>
<tr>
<td>2,337,118</td>
<td>Lantz</td>
<td>Dec. 21, 1943</td>
</tr>
</tbody>
</table>