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2,969,302

## METHOD OF MAKING PAPER

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No Drawing. Filed Feb. 14, 1957, Ser. No. 640,079

5 Claims. (Cl. 162—164)

This invention, in general, relates to the manufacture of paper and, more particularly, to improvements in retention of fillers and fines in said manufacture.

Paper is manufactured for the most part from wood pulp. A small amount of high grade paper is manufactured from rag pulp. There are four different kinds of wood pulp: mechanical pulp, sulfite pulp, sulfate or kraft pulp and soda pulp. The first is prepared by purely mechanical means, the other three by chemical means. The mechanical pulp contains substantially all of the wood except the bark and that lost during storage and transportation. Chemical pulps, however, are essentially pure cellulose, the unwanted and unstable lignin and other noncellulosic components of the wood having been dissolved away by the treatment. Because of this, chemical pulps are much superior to mechanical pulp for fine paper making. However, because of the special processing required, they are too expensive for the cheaper grades of papers such as newsprint.

If the pulp fibers were the only constituents of a paper sheet, the usefulness of the paper would be very restricted because the sheet would be soft, have a yellowish color and could not be written or printed upon with ink successfully. If the sheet were thin, it would be transparent to matter printed upon the opposite side. It is necessary, then, to add other substances, such as sizing, coloring agents, and fillers, to the cellulosic fibers to produce paper suited to its many uses.

Virtually all papers except the absorbent types and filter paper must have a finely ground filler added to them, the purpose of which is to occupy the spaces between the fibers—thus giving a smooth surface, a more brilliant whiteness, improved printability and improved opacity. The fillers are inorganic substances and may be either naturally occurring materials such as talc, agalite, pearl filler, barytes and certain clays such as china clay or artificial fillers such as suitably precipitated calcium carbonate, crown filler (pearl hardening), blanc fixe and titanium dioxide pigments. Sizing is added to the paper other than absorbent papers and filter paper to impart resistance to penetration by liquids. Common sizing agents added to the pulp before it is formed into a sheet are a wax emulsion or a soap made by saponification of rosin with alkali. The sizes are precipitated with alum.

Pulp stock is prepared for formation into paper by two general processes, beating and refining. There is no sharp distinction between the two operations. Mills use either one or the other alone or both together. The most generally used type of beater is that known as the Hollander. Beating the fibers makes the paper stronger, more uniform, more dense, more opaque and less porous. It is in the beater that fillers, coloring agents and sizing may be added. The beaters are batch machines, and for this reason some processes for making lower grades of paper have done away with the beaters entirely and use only refiners. The standard practice in making the

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finer grades of paper, however, is to follow the beaters with the refiners, the latter being continuous machines. The standard refiner is known as a Jordan engine.

While the usual practice is to add filler, sizing and color to the beaters, they may be added in the Jordan or in both. In some instances, all or part of the sizing is applied to the formed, dried sheet—using animal glues, starches or gelatin as the sizing. The order in which the materials are added to the beaters may vary with different mills. Generally, however, the filler is first added to the blended pulp, and, after sufficient beating, the sizing and coloring are added.

The machines used for the actual formation of the paper sheet are of two general types, the Fourdrinier machine and the cylinder machine. The basic principles of operation are essentially the same for both machines. The sheet is formed on a travelling bronze screen or cylinder, dewatered under rollers, dried by heated rollers and finished by calender rolls. In the Fourdrinier machine the stock of the foregoing operations is sent to the headbox from which it flows onto a moving, endless bronze wire screen. The pulp fibers remain on the screen while a greater portion of the water, containing unretained fiber fines and unretained filler, drains through. As the screen moves along it has a sidewise shaking motion which serves to orient some of the fibers and give better felting action and more strength to the sheet. While still on the screen the paper passes over suction boxes to remove water and under a dandy roll which smooths the top of the sheet. In the cylinder machine there are several parallel vats into which similar or dissimilar dilute paper stocks are charged. A wire covered rotating cylinder dips into each vat. The paper stock is deposited on the turning screen as the water inside the cylinder is removed. As the cylinder revolves further, the paper stock reaches a point where the wet layer comes in contact with and adheres to the moving felt. This felt and paper after removal of some water come into contact with the top of the next cylinder and pick up another layer of wet paper. Thus a composite wet sheet or board is built up and passed through press rolls and onto the drying and smoothing rolls.

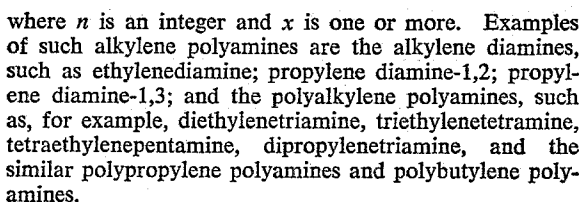
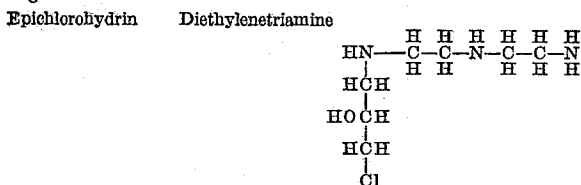
A certain amount of pulp fibers are of sufficiently small dimension that they are not caught in the fibrous matting on the sheet-forming screens, and these fibers pass through with the water. It is with improved retention of these fines that this invention is primarily concerned. Another improvement in paper manufacture also accomplished by the present invention is an increase in filler retention.

It is an object of the present invention, therefore, to provide means for improving the retention of fiber fines in the fiber matting of paper. A further object is to provide processes for manufacture of paper wherein an improved retention aid is employed for increasing the amount of fillers retained in the paper sheet during the manufacture thereof. Other objects will appear hereinafter.

Briefly, the paper manufacturing processes of the present invention comprise the addition of the condensation products of alkylene polyamines and polyfunctional halo-hydrins to paper stock at any convenient point. The dosage range of the active ingredient may be varied between about 0.05 pound to one pound per ton of produced paper.

The reaction products of the alkylene polyamines and polyfunctional halo-hydrins are hydrophilic polymers. The hydrophilic alkylene polyamine polyfunctional halo-hydrin polymers are reaction products of alkylene polyamines with polyfunctional 2-4 carbon halo-hydrins such as dihalohydrins, e.g., glycerol alpha-dichlorohydrin, di-

The alkylene polyamines which are reacted with the poly-functional halohydrins for the purpose of the invention are well known compounds having the general formula


$$(1) \quad \text{CH}_2\text{CHCH}_2\text{Cl} + \text{H}_2\text{NCH}_2\text{CH}_2\overset{\text{H}}{\text{N}}\text{CH}_2\text{CH}_2\text{NH}_2 \longrightarrow$$


The hydrophilic condensation polymers employed for the purpose of the invention are of a relatively high molecular weight which is believed to be in excess of 1000 and in most cases greater than 2000, but because of the difficulty of determining molecular weight, the most satisfactory way of ascertaining the proper amount of condensation is by viscosity measurement. The products which have been found to be especially suitable for the practice of the invention have a minimum viscosity in an aqueous alkaline pH solution containing 20% by weight of the condensation polymer at a temperature of 75° F. of about 7 centipoises. The upper limit of the viscosity is anything short of gel formation and may be, for example, up to 150 to 200 centipoises. However, the preferred range of viscosity is about 14 to 90 centipoises. The viscosity determinations were made by using a 20% polymer solution having a pH of about 12.6.

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The relative proportions of polyamine and polyfunctional halohydrin employed in making polyamines for the purpose of the invention can be varied depending upon the particular type of polyamine and polyfunctional halohydrin and the reaction condition. In general, it is preferable that the molar ratio of the polyfunctional halohydrin to polyamine be in excess of 1:1 and less than 2:1. Thus, in the preparation of a condensation polymer solution from epichlorohydrin and tetraethylenepentamine, good results have been obtained at a molar ratio of 1.4:1 to 1.94:1. The reaction temperature is preferably in the range of about 40–60° C.

### EXAMPLE A

| Ingredients:                             | Parts by weight |
|------------------------------------------|-----------------|
| Tetraethylenepentamine                   | 10.3            |
| Epichlorohydrin                          | 9.7             |
| Water (added prior to reaction)          | 25.3            |
| Water (added after reaction is complete) | 54.7            |

The tetraethylenepentamine was dissolved in a volume of water equal to 25.3% of the final batch weight. While the solution was being stirred the epichlorohydrin was added slowly over a 1.5 hour period. During this addition the temperature of the reaction was maintained between 45° C. and 50° C. with cooling. The reaction mixture was allowed to stand for an additional ½ hour with stirring at the same temperature. It was then diluted with the remainder of the water and cooled to room temperature (about 19° C.). The resultant solution contained about 20% by weight of active polymer and had a pH of 7.6.

### EXAMPLE B

| 45 | Ingredients:                                                | Parts by weight |
|----|-------------------------------------------------------------|-----------------|
|    | Softened water (added prior to reaction) _ _ _              | 17.85           |
|    | Tetraethylenepentamine _ _ _ _ _                            | 11.22           |
|    | Epichlorohydrin _ _ _ _ _                                   | 8.77            |
|    | 50% NaOH in water _ _ _ _ _                                 | 7.59            |
| 50 | Softened water (added after reaction is complete) _ _ _ _ _ | 54.47           |

The 17.85 parts of softened water was added to a stainless steel jacketed reaction vessel. The tetraethylenepentamine was then added to the vessel with agitation. The epichlorohydrin was then added to the vessel gradually at a rate such as to produce the desired temperature. The temperature was controlled by a cooling jacket. After all of the epichlorohydrin had been added, the solution of NaOH in water was added rapidly while cooling the reaction vessel. The rest of the softened water was then added. At the time of the addition of the tetraethylenepentamine to the water, the temperature of the mixture was raised to 105° F. to 110° F. The epichlorohydrin was then added at such a rate as to bring the temperature up to and keep it at 130° F. to 135° F. This addition time was approximately 1½ hours. After all of the epichlorohydrin had been added and the temperature had just started to drop, the sodium hydroxide solution was added in a period of about 5 minutes. The temperature rose to a peak and then dropped. After the temperature had reached a peak and had dropped to about 110–115° F. the water in the cooling jacket was cut off and the rest of the softened water was added to the vessel as rapidly as possible.

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The pH of the finished 20% polymer solution was 12.6.

In this example the sodium hydroxide solution was added in order to force the reaction, stabilize the product and reduce corrosion. Various contaminants such as succinic acid, isopropanol, ethanolamine and citric acid cause the product to gel and therefore should be avoided.

A water solution of the foregoing condensates may be introduced into the paper stock by addition into the white water at the fan pump suction or at the mix box just prior to stock addition. The paper stock may contain about 0.5-10%, generally about 1-5%, cellulosic fibers by weight. If it is desired to dilute the polycondensate solution prior to addition, unchlorinated water may be used for the dilution. The invention is applicable in the manufacture of papers wherein a smoother surface, a more brilliant whiteness, improved printability and/or improved opacity are desired—these properties in the paper being imparted by the addition of a filler. Types of paper and paperboard manufacture to which the instant invention is applicable are the manufacture of newsprint, specialty

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maintain basis weight specifications in this run due to increased retention of fines.

### EXAMPLE II

In a printing paper run on a Fourdrinier machine operating at about 36 tons per day and at about 400 feet per minute, a dosage of three pounds per ton of a 20% water solution of the polycondensation product of epichlorohydrin and tetraethylene pentamine was fed to the white water in the mix box just prior to stock addition. After shutting off glue feed, no trouble in the operation was experienced throughout the run and all readings on the test stayed within the standards. The suction at the couch went up about 1¼" and the wax pick increased over that during the operation of the run with glue just prior to the test run.

The test readings during the run and just prior thereto are reported in the following table. The chemical components per ton of produced paper were: clay, 700 pounds; alum, 45 pounds; size, 45 pounds; starch, 15 pounds; and silicate, 15 pounds.

Table 2

| Time              | Basis     | Density | Mullen | Bulk  | Fold |      | Wax Pick | Opacity   |
|-------------------|-----------|---------|--------|-------|------|------|----------|-----------|
|                   |           |         |        |       | C.D. | M.D. |          |           |
| Standard.....     | 44.4/46.4 | 16/24   | 14/21  | 38    | 20   | 20   | 13+      | 89/91     |
| Glue—Average..... | 44/45.6   | 22-33   | 22     | 38-39 | 35   | 30   | 13+      | 88.7/89.1 |
| 50 min.....       | 45.9      | 39      | 22.2   | 39.3  | 28   | 32   | 13+      | 89.6      |
| 85 min.....       | 45.9      | 31      |        | 39.5  |      |      |          | 89.7      |
| 125 min.....      | 44.8      | 25      | 23.6   | 38.3  |      |      | 16       | 89.4      |
| 165 min.....      | 46.0      | 25      |        | 39.3  | 33   | 35   |          | 89.3      |
| 215 min.....      | 45.7      | 29      | 24.0   | 39.1  |      |      | 14       | 89.3      |
| 255 min.....      | 45.8      | 28      |        | 39.2  |      |      |          |           |
| 300 min.....      | 45.8      | 27      | 23.8   | 39.2  | 32   | 35   | 16       | 89.4      |

papers, book paper, cover paper, writing paper, wrapping paper, container boards, folding box board and Bristol board.

The invention will be further understood from the following examples.

### EXAMPLE I

In a mimeograph paper run on a Fourdrinier machine, a 20% solution of a condensation product of epichlorohydrin and tetraethylenepentamine was fed into the white water at the mix box just prior to stock addition. Feed ratio was approximately one and one half to two pounds per ton of produced paper. Glue was being used in the commercial run, and after the feed of the condensation product, glue feed was stopped and the condensation product solution feed was begun. The machine speed was 350 f.p.m. The chemical components of the paper stock per ton of produced paper were as follows: titanium dioxide, 75 pounds; clay, 320 pounds; alum, 45 pounds; gum beater starch, 100 pounds; and size, 45 pounds.

The characteristics of the paper and the pH at various time intervals during the run are reported in the following table:

Table 1

| Time              | Smoothness | Ash   | Basis Wt. | Density | Bulk | Wax Pick | Opacity | Brightness | pH      |
|-------------------|------------|-------|-----------|---------|------|----------|---------|------------|---------|
| Standard.....     | 10         | 10    | 44-47.5   | 16-20   | 50   | 9-11     | 93      | 72-74      |         |
| Glue—average..... | 10-11      | 10-11 | 46        | 24-30   | 48   | 7        | 93-94   | 75         | 4.6/5.2 |
| 30 min.....       | 10         | 9.6   | 43.7      | 26      | 50   | 9        | 91.3    | 73.5       | 5.1     |
| 65 min.....       | 12         | 10.3  | 47.5      | 28      |      |          | 95.0    | 75.5       | 4.3     |
| 100 min.....      | 14         |       | 47.7      | 32      | 50.6 | 9        | 94.5    | 75.0       | 4.4     |
| 140 min.....      | 14         | 10.2  | 46.0      | 32      | 50.5 | 9+       | 94.6    | 75.0       | 4.5     |

These results indicate improved smoothness, greater density and increased wax pick due to better loading by fines and filler. Two furnish reductions were necessary to

The brightness of the paper as measured at the 50 minute to 300 minute intervals was 74. The standard brightness is 72-74 and the average brightness of paper made with glue is 73.5.

The ash content of the paper at 50 minute to 300 minute intervals varied from 11.7 to 12.4. The ash content of paper made with glue averages around 12.2 and the standard varies from 12 to 19.

The couch suction in each instance at 50 to 300 minute time intervals was 10.5" as compared with average couch suction in paper made with glue of 9.25".

In this example, increases in mullen, wax pick and opacity can all be attributed to increased fiber retention. The increased couch vacuum indicates better loading and a drier sheet but operation at the increased vacuum also tends to hold finished sheets ash down.

The test run was continued for about an additional 18 hours during which the readings were very close to those reported. The pH ran very close to 4.5 throughout the entire run.

### EXAMPLE III

In the manufacture of newsprint on a Fourdrinier

machine operating at 990 f.p.m., a dosage of one pound per ton of produced paper of a 10% water solution prepared by diluting a 20% solution of the polycondensation

product of epichlorohydrin and tetraethylenepentamine was fed to the white water at the fan pump suction. Clay feed at the beginning of the trial run was 144 pounds per ton. After one hour running time there was no change in the couch suction. General observations made during the initial portion of the run showed no great decrease in ash, opacity and brightness—all of prime importance.

After two hours and 45 minutes the polycondensate solution feed was increased to two pounds per ton. The immediate effect was a  $\frac{1}{2}$ " increase at the couch. After  $4\frac{1}{4}$  hours the feed of the polycondensate was changed from the 10% solution to a 20% solution at the same feed rate. Observations after the change to the 20% solution are reported in the following table.

Table 3

| Time         | G.p.m. Clay | Mullen | Bright-ness | Smoothness |     | Opacity | Ash  |
|--------------|-------------|--------|-------------|------------|-----|---------|------|
|              |             |        |             | F          | W   |         |      |
| 30 min.....  | 20          | 10     | 55.7        | 90         | 97  | 89.3    | 3.32 |
| 90 min.....  | 22.8        | 10.5   | 55.7        | 100        | 109 | 90.0    | 4.05 |
| 150 min..... | 22.8        | 9.8    | 56.0        | 106        | 115 | 88.5    | 4.93 |

The retention of clay was increased to a degree greater than the clay increases in the furnish.

the 20% polycondensate solution, 60% of the clay was retained in this grade newsprint. Retention of clay in the sheet remained high during periods of decreased clay feed to the system. Printability tests made on the newsprint indicated that this grade prints better than most grades (both black and gray) with a minimum of show-through.

## EXAMPLE IV

On a Fourdrinier machine operating at about 45 tons per day and at about 400 feet per minute, a dosage of two pounds per ton of a water solution of the polycondensation product of epichlorohydrin and tetraethylene pentamine was fed to the suction of the fan pump, at the rate of 2 pounds per ton. Machine operation was maintained with no difficulty, and couch vacuum increased almost from  $9\frac{1}{2}$ " to  $10\frac{1}{2}$ ". The greatest effect of the retention aid was an increase in sheet weight, and it was necessary to cut back the flow of stock to the machine to maintain a 45 lb. sheet. Upon conclusion of the test and the discontinuation of the retention aid, the couch vacuum immediately returned to  $9\frac{1}{2}$ " and basis weight dropped 1 pound.

The test readings prior to, during and after the run are reported in the following table. The chemical components per ton of produced paper were: alum 84 lbs., size 18 lbs., clay 700 lbs.

Table 5

| Time           | Mullen | Size | Density | Ash | Vacuum | Opacity             | Bright-ness | Weight |
|----------------|--------|------|---------|-----|--------|---------------------|-------------|--------|
| Standard.....  | 12     | 6"   | 40      | 20½ |        | As High as Possible |             | 45     |
| Prior Avg..... | 16½    | 3"   | 14      | 19  | 9½"    | 86½                 | 77          | 45     |
| 10:30 a.m..... | 17     | 3    | 30      |     |        | 86                  |             | 45     |
| 11:05 a.m..... | 17½    | 3    | 22      |     | 9½     |                     | 79          | 45     |
| 11:40 a.m..... | 17½    | 3    | 22      | 19  | 10½    |                     |             | 45½    |
| 12:10 p.m..... | 18     | 12   | 29      |     |        | 87                  |             | 45½    |
| 12:40 p.m..... | 16½    | 24   | 18      |     | 10½    |                     | 79          | 45½    |
| 1:15 p.m.....  | 17     | 18   | 26      |     |        | 88½                 |             | 45     |
| 1:40 p.m.....  | 16½    | 10   | 20      |     | 10½    |                     |             | 45½    |
| 2:15 p.m.....  | 17     | 8    | 18      | 19  |        | 86                  | 78½         | 45½    |
| 2:45 p.m.....  | 17½    | 7    | 18      |     | 10½    | 86                  |             | 45½    |
| 3:15 p.m.....  | 17½    | 6    | 17      |     |        | 87                  |             | 45½    |
| 3:45 p.m.....  | 17½    | 7    | 19      | 19  | 10½    |                     | 78½         | 45½    |
| 4:15 p.m.....  | 18     | 5    | 24      |     | 9½     |                     |             | 44½    |
| 4:45 p.m.....  | 17     | 5    | 27      |     |        | 86                  |             | 45     |
| 5:15 p.m.....  | 17     | 6    | 27      |     | 9½     |                     | 78          | 45     |
| 5:45 p.m.....  | 17½    | 6    | 34      |     |        |                     |             | 45     |
| 6:20 p.m.....  | 17     | 6    | 27      |     | 9½     |                     |             | 45     |

<sup>1</sup> Retention aid addition begun and  $1\frac{1}{2}$ " stock cut off.

<sup>2</sup> Retention aid addition stopped and basis weight dropped.

Later the feed of the 20% polycondensate solution was cut to one pound per ton. Observations made during the latter part of the run at a one pound per ton feed are reported in the following table:

Table 4

| Time           | G.p.m. Clay | Mullen | Bright-ness | Smoothness |     | Opacity | Ash  |
|----------------|-------------|--------|-------------|------------|-----|---------|------|
|                |             |        |             | F          | W   |         |      |
| 9:00 p.m.....  | 22.8        | 9.8    | 56.4        | 109        | 117 | 89.3    | 5.73 |
| 11:05 p.m..... | 22.8        |        | 56.7        | 103        | 111 | 89.3    | 5.89 |
| 3:00 a.m.....  | 20.6        | 9.2    | 56.0        | 109        | 121 | 89.0    | 6.09 |
| 4:45 a.m.....  | 20.6        |        | 56.7        | 100        | 119 | 89.9    | 6.07 |
| 7:05 a.m.....  | 20.6        |        | 56.5        | 97         | 110 | 89.0    | 5.83 |
| 8:15 a.m.....  | 22.8        |        | 56.0        | 85         | 88  | 90.0    | 5.83 |
| 9:05 a.m.....  | 20.0        | 9.7    | 56.5        | 110        | 124 | 90.0    | 5.91 |
| 10:20 a.m..... | 16          |        | 55.8        | 75         | 76  | 89.5    | 5.03 |
| 11:30 a.m..... | 16          | 9.3    | 55.3        | 118        | 128 | 88.5    | 4.57 |
| 12:25 p.m..... | 16          |        | 56.3        | 117        | 121 | 89.5    | 4.57 |

All tests, including ash, remained within the desired range. Feed of the polycondensate was stopped at 1:45 p.m. Generally, the trial proceeded more smoothly with better results when the feed was changed to the undiluted chemical as compared to the use of a diluted solution with water containing a chlorine residual. Retention tests following this trial showed that at a one pound feed of

The above data indicates increases in basis weight due to improved fines retention.

The tests reported in the foregoing tables were used to determine various characteristics of the paper. Results reported under "Mullen" are determined from a test for determining the bursting strength of paper. It is a comparative test and is expressed as the ratio of strength of the paper to weight of the sample. Opacity is a comparative test usually measured by a photometer. Brightness is the measure of the reflectivity of a sheet of pulp or paper for blue light measured under standardized conditions on an instrument designed and calibrated specifically for the purpose. Smoothness is the property of the paper surface determined by its variations from an ideal surface such as a plain or cylindrical surface. Bulk is the thickness of a pile of a specific number of sheets under a specified pressure. Wax pick determines the bonding strength of the paper. Bonding strength is a measure of the resistance of the paper to removal of a coating or fibers from the sheet.

In certain of the foregoing tables, a "standard" was set forth for the various characteristics of the produced papers. The standards are in each case the desired values set by the paper manufacturer on whose equipment the tests were run.

The invention is hereby claimed as follows:

1. A process for improving filler and fiber fines retention in the manufacture of cellulosic paper which comprises adding to paper stock containing an inorganic filler from 0.05 to one pound per ton of produced paper of a hydrophilic alkylene polyamine-polyfunctional halohydrin polymer; and thereafter forming a paper sheet from said stock, said stock being substantially free from other synthetic linear polymers. 5

2. A process for improving filler and fiber fines retention in the manufacture of cellulose paper which comprises adding to paper stock containing an inorganic filler from 0.05 to one pound per ton of produced paper of a polyalkylene polyamine-dihalohydrin polymer; and thereafter forming a paper sheet from said stock, said stock being substantially free from other synthetic linear polymers. 10 15

3. A process for improving filler and fiber fines retention in the manufacture of cellulosic paper which comprises adding to paper stock containing an inorganic filler from 0.05 to one pound per ton of produced paper of a polyethylene polyamine-epichlorohydrin polymer; and thereafter forming a paper sheet from said stock, said stock being substantially free from other synthetic linear polymers. 20 25

4. A process for improving filler and fiber fines retention in the manufacture of cellulosic paper which comprises adding to paper stock containing an inorganic filler from 0.05 to one pound per ton of produced paper of a polyethylene polyamine-dihalohydrin polymer; and thereafter forming a paper sheet from said stock, said stock being substantially free from other synthetic linear polymers.

5. A process for improving filler and fiber fines retention in the manufacture of newsprint which comprises adding to paper stock containing an inorganic filler, cellulosic fibers, clay, and 0.05 to one pound per ton of produced paper of a hydrophilic alkylene polyamine-polyfunctional halohydrin polymer; and thereafter forming a newsprint sheet from said stock, said stock being substantially free from other synthetic linear polymers.

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**UNITED STATES PATENT OFFICE**  
**CERTIFICATION OF CORRECTION**

Patent No. 2,969,302

January 24, 1961

Jerome Green

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 5, line 52, for "ocdensation" read  
-- condensation --; columns 7 and 8, Table 5, under the  
heading "Ash", and opposite "Standard", for "20½" read  
-- 20% --; column 8, line 14, for "o f2" read -- of 2 --;  
column 9, line 11, for "cellulose" read -- cellulosic --.

Signed and sealed this 22nd day of August 1961.

(SEAL)

Attest:

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Commissioner of Patents