May 7, 1968 RYUZO NODA 3,382,088

METHOD OF MANUFACTURING SELF-COPYING SHEET

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FIG. 1

FLOW SHEET OF THE PROCESS

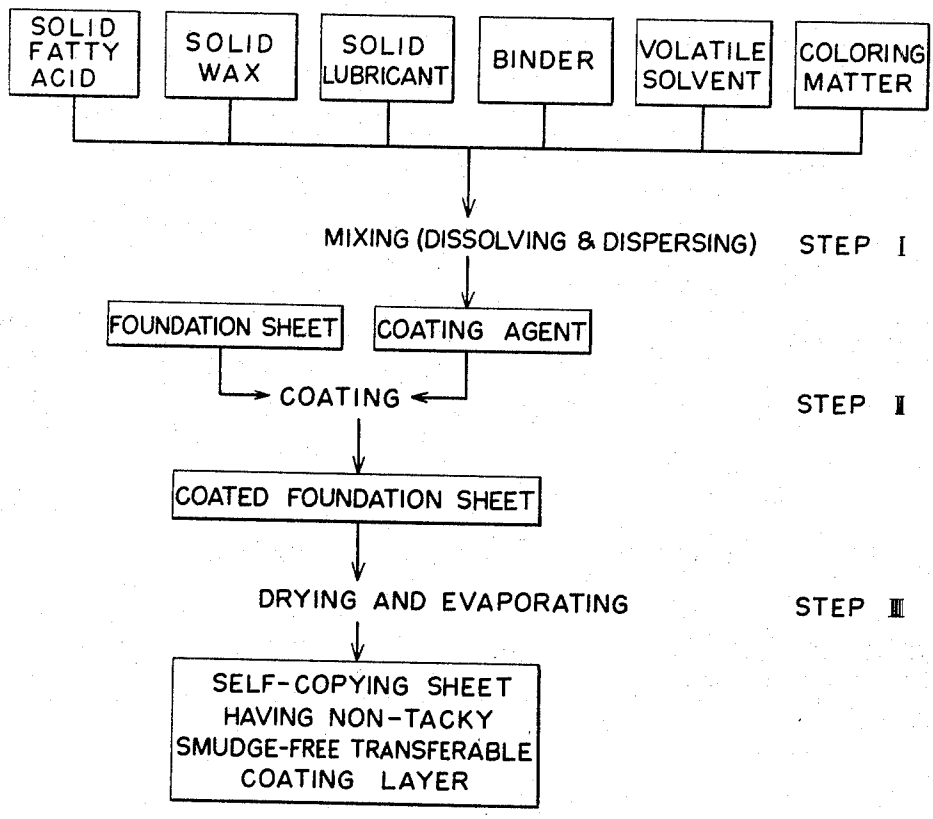
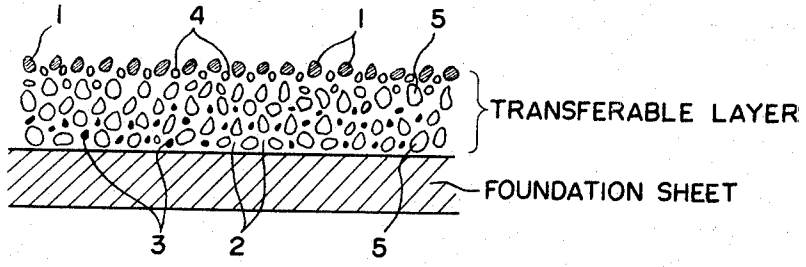


FIG. 2



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METHOD OF MANUFACTURING SELF-COPYING SHEET

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Continuation-in-part of application Ser. No. 257,836, Feb. 12, 1963. This application Apr. 28, 1965, Ser. No. 451,439

7 Claims. (Cl. 117—36.1)

This application is a continuation in part of my application 257,836, filed Feb. 12, 1963, and now abandoned by filing this application.

This invention relates to a novel self-copying sheet, for example, of paper with a transferable layer coated on one side thereof and a method of manufacturing the same. It is an object of the invention to provide a self-copying sheet which eliminates various defects in or disadvantages inherent to conventional manifolding papers including carbonized forms, spot and strip carbons.

According to the invention, a transferable layer is coated on one side of a foundation sheet. And when a plurality of such sheets is placed one over another and applying the pressure of writing or typewriting on the top surface of the piled sheets, such writing or typewriting portions are recorded on the top surface and simultaneously the corresponding pressed portions of each transferable layer are removed to transfer to a sheet positioned therebelow. Thus, according to the inventive self-copying sheet, there is no need of carbon paper or the like to be inserted between sheets.

Carbonized forms prepared by prior processes soil the user's hands on ordinary handling, and the transferred inscriptions or images tend to smudge when touched. Furthermore, even the slightest scratch and rub applied on the surface of these forms often produce unintentional dirty marks on subsequent copying sheets positioned underneath, so that the user is required to use them with utmost care. These defects or disadvantages are due to the tackiness or stickiness of the carbon ink coated as the transferable layer on the reverse of the forms. The carbon ink coating becomes tacky or sticky and dirty-to-touch because liquid oils or fats are employed in the color vehicle to allow the ink coating to transfer to a copy sheet positioned therebelow under the pressure of writing. When such liquid oils or fats, which exhibit sticky or fluidity characteristics, are removed from the carbon ink, tacky or stickiness and consequently dirty-to-touch defects can be diminished, however, transferability is extremely decreased.

The present invention eliminates all the above defects or disadvantages and presents a non-tacky and clean-to-touch transferable layer without using liquid oils or fats and simultaneously improves the transferability to a great extent. While conventional carbonized forms are coated with hot melted ink at temperatures of from 70° C. to 80° C., just as high temperature as when pigments or dyes are mixed or dispersed in waxes and oils to prepare the ink, this invention produces the transferable layer by coating one side of the foundation sheet with a mixture or dispersion of ingredients at normal temperature and drying such mixture or dispersion by air, likewise at normal temperature. According to the invention, the coating agent forming the transferable layer consists of solid fatty acid or solid fatty acid and solid wax, solid lubricant, binder, volatile solvent and coloring matter.

Solid fatty acid used in this invention is finely crystallized at the top surface of the transferable layer so that the top surface of the layer exhibits uniform and discontinuous layer. Such solid fatty acids are, for example, stearic acid, palmitic acid, myristic acid, behenic acid

and so on, which are easy to be crystallized with the aid of volatile solvent.

Solid wax in this invention assists to control the size of crystallized particles of solid fatty acid and effects on the hardness of the layer and such solid waxes are, for example, paraffin wax, carnauba wax, montan wax, beeswax and microcrystalline wax.

Solid lubricant used in this invention assists to disperse coloring matter in the coating agent and tends to give smooth layer. Such solid lubricants are for example, refined talc, prepared calcium carbonate and so on.

Binder in this invention is dissolved in the volatile solvent to bind solid fatty acid, solid wax, solid lubricant and coloring matter and further assists to bind the layer with the sheet. Such binders are, for example, ethylcellulose of low viscosity, nitrocellulose of low viscosity, chlorinated rubber and so on.

Volatile solvent is used in this invention for forming the coating agent and volatile solvent dissolving or dispersing solid fatty acid, solid wax and binder is mixed together with coloring matter and solid lubricant. When volatile solvent is vaporized during drying step, it assists in the crystallization of solid fatty acid at the top surface of the layer. Such volatile solvents are, for example, toluol, methanol and so on.

Coloring matters used in this invention are such as, for example, carbon black, iron blue (pigment) and victoria blue, rhodamine (dyestuff).

The present invention will be more readily understood from the following description when read in conjunction with the accompanying drawing, in which:

FIGURE 1 is a flow sheet showing an illustrative process for preparing the novel self-copying sheet having a non-tacky, smudgeless, transferable coating according to the instant invention; and

FIGURE 2 shows a cross-section of one embodiment of the self-copying sheet made according to this invention.

In FIGURE 1, a binder, a solid fatty acid, a solid wax, a solid lubricant, a coloring matter and a volatile solvent are mixed, for example in a ball mill, until the solid components are dissolved and uniformly dispersed in suspension in the solvent, whereupon one side of the foundation sheet is coated at normal temperature with the coating agent so obtained. The coated foundation sheet is dried at normal temperature to crystallize the solid fatty acid and to evaporate the solvent, thereby obtaining the self-copying sheet having a non-tacky smudgefree, transferable coating. In the section thereof shown in FIGURE 2 the constitution of the transferable layer is clearly shown.

In accordance with the present invention non-tacky solid fatty acid or said solid fatty acid and solid wax are used as the color vehicle instead of liquid oils or fats and a solid lubricant is added thereto so as to decrease the coefficient of friction on the surface of the layer, thus protecting the layer from being damaged by scratch or rub. However the carbon ink transferable layer prepared by use of non-tacky solid fatty acid or solid fatty acid and solid wax as the color vehicle has the big disadvantage in that it loses the transferability. Although this invention uses non-tacky solid fatty acid or non-tacky solid fatty acid and solid wax as the color vehicle, the solid fatty acid is crystallized at the top surface of the transferable layer thus making the layer non-continuous. As a result the layer becomes more liable to break and transfer under the pressure of writing, but, at the same time, the resistance to mechanical strength, particularly the resistance to scratch and rub, will be substantially weakened. In order to overcome this defect a solid lubricant is added to the ingredients of the transferable layer. Such addition of solid lubricant decreases the coefficient of friction on the surface of the layer, while permitting transferability

of the layer under the pressure of pen, pencil, stylus, type bar, etc. and yet prevents the layer from being damaged or soiled by scratch or rub.

Furthermore, the inscriptions or images transferred from the layer become sharp, non smudging and clean-to-touch.

Recently some self copying papers, commonly known as "carbonless paper" or "manifolding paper," have appeared on the market but the present invention is essentially different from them. The prior art self copying papers comprise a transfer sheet whose back is coated with a chemical transferable layer and an adjacent copy which has a receptive chemical layer or acceptor on the surface. To form inscriptions or images, the two chemical coatings must be contacted, thus, a set consisting of transfer sheet and receptive sheet is required. The present invention eliminates the need of such receptive coating, and no chemical interaction is involved. The transferable layer coated only on the back of a sheet produces sharp, non smudging, clean-to-touch inscriptions under the pressure of writing. Therefore, as compared with prior art self copying papers, the manufacturing method is much simpler and economical. As lubricant in this invention, refined talc is most suitable. The quantity of talc to be incorporated in the mixed ingredients of the layer should be 5 to 50% by weight and the particle size of the talc of from 0.5 to 50 microns. It is noted, however, that there is no specific limitations on the quantities of other ingredients other than talc and those quantities may be easily selected by the persons in the art referring to the examples shown below. In the following examples, parts are designated by weight.

Example I

	Parts
Ethylcellulose of low viscosity	1
Stearic acid	5
Paraffin wax	5
Iron blue	1
Toluol	80
Refined talc	30

Example II

	Parts
Ethylcellulose of low viscosity	2
Stearic acid	10
Iron blue	1
Toluol	80
Refined talc	30

Example III

	Parts
Chlorinated rubber	1
Palmitic acid	8
Carnauba wax	2
Prepared calcium carbonate	20
Carbon black	1
Toluol	70

The above ingredients are ground and mixed uniformly in a ball mill until solid ingredients are dispersed in suspension, the uniformly mixed ingredients presenting a suitable condition for coating at normal temperature. Then the mixture is coated evenly over one side of web or roll paper A (shown in FIG. 2) by coating machine equipped with suitable drying apparatus, by this latter apparatus the coating is dried and the volatile agents therein are evaporated by, for example, air of normal temperature. Then the solid fatty acid such as stearic acid is crystallized as shown in FIG. 2 as crystalline particle **1** at the top surface of the coating with the solid wax **4** such as paraffin wax scattered therebetween, while the binder **2** such as ethylcellulose of low viscosity, the solid lubricant **5** such as refined talc and coloring matter **3** are dispersed in the transferable layer with the result that the satisfactory self copying sheet with a non-tacky transferable layer is obtained. A thickness of 5 to 7 microns is suitable for such layer.

In accordance with the invention, solid fatty acid such as, for example, stearic acid is crystallized at the top surface of the transferable layer so that the top surface of the layer exhibits uniform and discontinuous layer, thereby, when applying pressure of writing or typewriting, only the pressed portions of the transferable layer are easily transfered to the sheet positioned therebelow. As aforementioned, in the conventional copying sheet, use is made of sticky or liquid oils or fats with the result that the obtained images or inscriptions lack clarity due to their tacky or sticky characteristics of the transferable layer. On the contrary, in accordance with the invention, which avoids to use such sticky or liquid oils or fats, adhesion between the transferable layer and the foundation sheet and the bonding force among ingredients constituting transferable layer are weak, and consequently, only the pressed portions of the layer will be easily removed from the foundation sheet and clearly transfered to the sheet positioned therebelow. It is hereby noted that when the pressure of writing or typewriting is applied to the transferable layer, crystallines of solid fatty acid are destroyed by the pressure and thus solid fatty acid recovers its inherent adhesion to the sheet. The advantages of the use of solid lubricant are as aforementioned in detail, and such addition of solid lubricant increases the uniformity of the transferable layer and decreases the coefficient of friction on the surface of the layer and furthermore, decreases smudging of the transferred inscriptions or images since coloring matter, which forms such inscriptions or images, is protected by solid lubricant which decreases the coefficient of friction from being damaged by scratch or rub.

Thus the self copying sheet prepared by this method is clean-to-touch itself and provides smudge-free, sharp inscriptions and yet the method is much simpler and economical as compared with prevailing manifolding or carbonless paper because only one side of paper is coated and no chemical interaction is involved according to the present invention.

What is claimed is:

1. A method of preparing a self-copying sheet having a non-tacky, clean-to-touch, and non-smudging transferable layer coated on one side thereof, which comprises mixing a solid fatty acid selected from the group consisting of stearic acid, palmitic acid, myristic acid and behenic acid, a solid wax, a solid lubricant selected from the group consisting of refined talc and prepared calcium carbonate, a coloring matter selected from the group consisting of pigment and dyestuff, a binder of low viscosity of the group consisting of ethylcellulose, nitrocellulose and chlorinated rubber and a volatile solvent of the group consisting of toluol and methanol to form a dispersion solution, coating the resulting solution on one side of the foundation sheet at normal temperature to crystallize the solid fatty acid and evaporate the solvent.

2. A method according to claim **1**, in which the binder is of low viscosity and of the group consisting of ethylcellulose, nitro-cellulose and chlorinated rubber and the volatile solvent is of the group consisting of toluol and methanol, and the solid wax is of the group consisting of paraffin wax, carnauba wax, montan wax, beeswax and microcrystalline wax.

3. A method of preparing a self-copying sheet having a non-tacky, clean-to-touch, and non-smudging transferable layer coated on one side thereof, which comprises mixing stearic acid, refined talc, iron blue as coloring matter, ethylcellulose of low viscosity as a binder and toluol as a volatile solvent to form a dispersion solution, the solution is composed by weight of ten parts of the stearic acid, thirty parts of refined talc, two parts of ethylcellulose of low viscosity, one part of iron blue, eighty parts of toluol, coating the resulting solution on one side of the foundation sheet at normal temperature

to crysatllize the solid fatty acid and evaporate the solvent.

4. The method according to claim **1** in which the solid fatty acid is stearic acid, the solid wax is paraffin wax, the solid lubricant is refined talc, the binder is ethylcellulose of low viscosity, the coloring matter is iron blue, the volatile solvent is toluol, and the solution is composed by weight of five parts of stearic acid, five parts of paraffin wax, thirty parts of refined talc, one part of ethylcellulose of low viscosity, one part of iron blue and eighty parts of toluol.

5. The method according to claim **1** in which the solid fatty acid is palmitic acid, the solid wax is carnauba wax, the solid lubricant is prepared calcium carbonate, the binder is chlorinated rubber, the coloring mater is carbon black and the volatile solvent is toluol and the solution is composed by weight of eight parts of palmitic acid, two parts of carnauba wax, twenty parts of prepared calcium carbonate, one part of chlorinated rubber, one part of carbon black and seventy parts of toluol.

6. A self-copying sheet with a non-tacky, clean-to-touch transferable layer in which one side of the sheet is coated with the transferable layer consisting of solid fatty acid selected from the group consisting of stearic acid, palmitic acid, myristic acid and behenic acid, a solid wax,

a solid lubricant selected from the group consisting of refined talc and prepared calcium carbonate, a coloring matter selected from the group consisting of pigment and dyestuff, a binder of low viscosity and of the group consisting of ethylcellulose, nitro-cellulose and chlorinated rubber and a volatile solvent of the group consisting of toluol and methanol, the solid fatty acid being dispersed substantially in the top surface of the layer in the crystal form with the solid wax scattered therebetween, the binder, the solid lubricant and the coloring matter are dispersed in the layer underneath the top surface thereof.

7. A self-copying sheet according to claim **6**, in which the solid wax is of the group consisting of paraffin wax, carnauba wax, montan wax, beeswax and microcrystalline wax.

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