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3,427,971 HECTOGRAPHIC UNIT INCLUDING A MASTER SHEET HAVING A PIGMENTED PROTEINA-CEOUS CELLULAR COATING Anthony J. Steber, Appleton, and Robert E. Weber, Neenah, Wis., assignors to Kimberly-Clark Corporation, Neenah, Wis., a cornoration of Delaware

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ABSTRACT OF THE DISCLOSURE

A hectographic master unit including a master sheet having a pigmented proteinaceous cellular coating on the master sheet. The master sheet contacts the usual transfer sheet with the cellular coating contacting the oilwax coating composition of the transfer sheet. No interleaving sheet is required between the transfer sheet and master as the cellular coating of the master inhibits premature transfer in shipment, storage and the like. 20

This invention relates to paper products for use in producing hectographic copies.

In the usual hectograph procedure a master sheet is $\mathbf{25}$ employed to make a plurality of copies, of printed matter for example. The master sheet is itself prepared from a transfer sheet. The transfer sheet usually carries a transferable coating of an oil-wax composition in which a dye 30 is incorporated. Dye-stuffs such as Victoria Blue or Crystal Violet may serve the purpose. Pressure on the face of the blank master, while the master is in contact with the oil-wax surface of the transfer sheet, causes printing in reverse to be transferred to the back of the master. A right-reading copy is then made from the master as is 35 well-known to the art by moistening a blank copy sheet with alcohol or other suitable duplicating solvent and contacting the master with the copy sheet. Alternatively, the oil-wax composition may contain a dye-forming com-40 ponent which, in contact with a solvent wet copy sheet containing a dye coupler, results in the marking of the copy. Such a procedure is broadly termed an alcohol transfer process. Usually a considerable plurality of copies may be made from the master sheet. 45

Marketing of the master and transfer sheet blanks as a unit poses a problem. If the transfer sheet contacts the master, the transfer of the oil-wax coating by migration or otherwise occurs prematurely. Accordingly, it is the practice to supply the master transfer blanks as a unit but with an interleaving tissue therebetween. Such inhibits premature transfer in shipment, storage and the like but requires removal of the tissue for use of the master and transfer sheets and involves waste of the interleaving tissue. Additionally, in some operations the master and transfer sheet are stored as a unit after a partial printing and this requires not only removal but reinsertion of the interleaving tissue.

It is an important object of this invention to provide a novel master sheet which is capable of resisting unwanted transfer to it from the transfer sheet of oil-wax by migration or otherwise when in simple stacked contact but which readily accepts the oil-wax component under writing (pencil, pen) pressure, typing pressure or other machine pressure; further, the novel master sheet accepts 65 and retains the oil-wax by marking pressure to the extent that at least as many, and usually more, copies may be obtained from the master than with masters sold in commerce. Stacks of the master-transfer sheets (81/2" x 11") have been stored in contact under elevated temperature conditions (120° F.) under a 50-pound load for a period 70 of two weeks without evidence of transfer.

It is a particular purpose of this invention to provide a master sheet, transfer sheet combination useful in a solvent transfer process and which does not require the inclusion of an interleaving tissue as a protective element during storage, shipping and the like.

To achieve the elimination of the interleaving sheet from the hectographic unit (master plus transfer sheet) the master is provided on one side with a coating containing a pigmented binder film of cellular structure. This film, for optimum effectiveness, we have found, should contain protein, a paper coating pigment and a relatively critical volume of cells throughout the coating both as to cell size and as to cell concentration. These cells are produced by the volatilization of oil from an oil-in-water emulsion following water evaporation.

The protein serves as a binder to retain the pigment particles together and to the sheet; more importantly, the protein has been found to resist migration of the components of the oil-wax. The protein is apparently not wetted by the oil-wax transfer material when the transfer and master sheets are in simple stacked contact; migration of the oil-wax component is then inhibited. The quantity of protein in the coating should be limited to that which is effective as binder and as migration inhibitor. Excess of protein will result in lesser effective transfer of the oilwax under pressure and increase coating cost. Thus, the protein need be present only in sufficient amount to serve as binder and oil migration inhibitor. The usual proteins which serve as binders in paper coating compositions are useful soy, casein, animal glue and the like.

The pigment in the coating aids the transfer of the oilwax to the master. Apparently it provides some "tooth" in the coating, permitting adhesion of the wax. An excess of pigment, however, tends to increase oil-wax migration, probably because it interrupts the binder film unduly.

The cellular structure cooperates with the protein content in inhibiting migration from the transfer sheet. This effect of the cellular structure is more pronounced as aging of the sheets in contact increases. Protein itself apparently is sufficiently resistant to transfer of the oil-wax to be immediately useful for this purpose; but with time significant migration develops in the absence of a cellular structure. The combination of the protein with the cellular structure, however, provides a product wherein no significant migration occurs after long periods of contact. From the commercial viewpoint this is important as some units may be stacked or stored for long periods before use.

We have found that on an absolute volume basis with protein serving as the only binder, there should be present in the binder for each absolute volume of the protein between 0.25 to 0.84 volume of pigment and 1 to 6 volumes of cells. By absolute volume is meant the reciprocal of the absolute density (in contrast to bulk density) of the material and predicated in the case of the cells upon oil density. The inclusion of starch or other water dispersible binders as a partial substitute for the protein binder reduces the effectiveness of the coating as to preventing migration; however, substantially the same proportions of binder (starch+protein) to pigment and cell volume will hold and may be readily adjusted by one skilled in the art to obtain the desired result. The cells themselves are derived from an oil-in-water emulsion in which the oil droplets are of approximately 0.5 to 2.5 microns in diameter; the cells have approximately the same dimensions as the oil droplets from which they are derived. The oil should have a vapor pressure less than that of water and may be selected from any of a wide variety of oils such as the hydrocarbons, kerosense, Stoddard's solvent, No. 1 and No. 2 fuel oils and the like.

We have also found that, while the pigment may be any of several paper coating pigments, some are preferable to others. Kaolin clay, for example, is more effective than calcium carbonate in inhibiting binder migration, though the carbonate is very useful. Also, starch is useful as a binder constituent but is of very little significance in preventing oil-wax component migration; for this latter purpose a protein content of the binder of at least 50% by weight is indispensable for a satisfactory product. Such protein is preferably soy though casein and animal glue serve the purpose. A too extensive cellular structure is effective to prevent migration; but such leads to a softness in the coating and results in smudging when transfer to the master is effected. A too small cellular content leads to unwanted migration upon aging of the hectograph unit.

In effect, we have found that there is a balance between the quantity of paper coating pigment, the cellular structure and the protein component, and that the effiisciency of the coating for its specific purpose as a hectograph master varies somewhat with the component quantities.

The base sheet to which the coating is applied may be any conventional base sheet employed as a master in 20 hectographic processes. We have found a base web containing 50% groundwood pulp and 50% kraft pulp and having a basis weight of about 28 pounds ($25'' \times 38'' \times$ 500 sheets) to be suitable. The base may, however, be any sheet commonly useful for hectographic purposes and 25 sheets having a basis weight of 60 pounds ($25'' \times 38'' \times$ 500 sheets) have been tested and found useful.

The invention will be more fully understood from the following detailed description and accompanying drawings wherein:

FIG. 1 illustrates a prior art master-transfer unit including the interleaving tissue;

FIG. 2 illustrates a master-transfer unit of this invention; and

FIG. 3 is a sectional view of the master sheet generally 35 illustrating the coating on the underside of the master.

Referring to the drawings for convenience of explanation, the prior art product improved by this invention is illustrated in FIG. 1. The numeral 1 designates the usual master sheet having commonly a conventional coating of 40 binder and pigment on each side thereof. The purpose of such coating is to improve the printability of copies and to prolong the life of the master.

A transfer sheet 2 is detachably joined with the master 1 at 3 to form an integral hectographic unit. The master 4 and transfer sheet may be provided separately. The transfer sheet 2 carries the usual dark colored oil-wax coating 4 and may contain either a dye or a colorless dye forming component as already noted above.

Interposed between the master and the transfer sheet 50 is a tissue sheet 5. This sheet is simply a slip sheet and is removed when the unit is employed. This sheet is itself commonly glazed on at least one side to minimize migration from the transfer sheet to the master.

As illustrated in FIGS. 2 and 3, the product of this in- 55 vention comprises the master 6 having a top side 6a and a bottom side 6b. A transfer sheet similar to that at 2 in FIG. 1 is integral with the master and is designated at 7, the oil-wax coating being indicated at 8.

The master 6 carries on its underside at 6b the oil-re- 60 sistant coating 10, containing the protein, pigment and having the cellular structure. The cells of the cellular structure are indicated at 11.

Many combinations of the variable factors set out above are possible to produce a suitable hectographic 65 master sheet and hectographic unit. Examples of particularly useful combinations are described below:

EXAMPLE 1

	Parts by weight	Parts by vol.	,
Soy protein (low viscosity)	200	144	• 1
Papermaker's clay	200	77	
Ammonia aqueous 28%	ca. 28	30	
Water	850	850	
Oil (Stoddard's solvent S.G0.78)	390	500	

NOTE.--Cell volume=3.5 per unit volume of protein.

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In this example, as in those which follow, the protein, pigment, ammonia and water are first slurried together and cooked for about 15 minutes at 90° C. The ammonia assists in dispersing the protein. To the thin pigmented coating material at 90° C. the oil is slowly added. Emulsification takes place under these conditions readily and light mixing provides oil droplets in the emulsion of 0.5 to 2.5 micron size. Such composition contains about 24.4% solids, exhibits a pH of 8.6 and a viscosity (Brookfield #6 spindle at 100 r.p.m.) at 50° C. of about 3500 centipoises. This composition is stable and is readily applied to conventional base sheets by usual coating procedures including blade, roll and by adjusting viscosity suitably with water addition the composition may be printed. When blade coated on a base sheet and dried in an air blast at about 300° F. (air temperature), the coating was very effective in resisting migration and very receptive to wanted transfer.

EXAMPLE 2

The formulation in this instance contained a lesser quantity of oil and was as follows:

	Parts by weight	Parts by vol.
Sov protein (low viscosity)	200	144
Papermaker's clay	200	77
Ammonia (aqueous)	28	30
Water	800	800
Oil (Stoddard's solvent)	156	200

NOTE.--Cell volume=1.30 per unit volume of protein.

The procedure was otherwise as in Example 1. The percent solids in this instance was 36.5; pH 8.6; and viscosity 3600 Brookfield. The coated paper product in this instance was excellent in image transfer but about 10% less copies of the same quality were produced than in the case of the sheet of Example 1.

EXAMPLE 3

The formulation in this instance was as in Example 1 except that a larger quantity of oil was employed and the water was increased slightly to attain a useful viscosity. The formulation:

144 77 30 900 800

NOTE.—Cell volume=5.58 per unit volume of protein.

The solids content was 21.5%, the pH 8.6 and the viscosity about 5000 Brookfield. In this instance the greater cell volume tended to affect the sharpness of the wanted oil-wax transfer to reduce quality slightly over that of Example 1 but migration prevention was excellent and about the same number of copies were producible from the master.

EXAMPLES 4, 5 AND 6

Examples 1, 2 and 3 were repeated but substituting for the papermaker's clay an equal weight of calcium carbonate. The results were generally in accord with those containing the clay but with some slight tendency to oilwax migration over a long period of storage.

In general, the elimination of the oil from the formulation of Example 1 resulted in a coating which was effective to inhibit migration but resulted in poor oil-wax transfer under marking pressure. Elimination of both oil and pigment also provided a product effective as to inhibiting migration but resulted in extremely poor transfer under marking pressure. Elimination of pigment alone from the formulation as in Example 1 similarly resulted in resistance to migration but also gave poor transfer. It is apparent that the protein is not wetted significantly 75 by the oil-wax and that the inclusion of the oil (cell

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producer) and pigment overcomes, at least under marking pressures of a pen, pencil, typewriter or the like, the poor transfer characteristic of the protein. Elimination of the protein by substitution in toto of another binder such as starch or latex does not result in a product, we have found, which is acceptable to prevent oil-wax migration.

The oil-wax composition may be, insofar as we have been able to determine, any of those commonly used in providing transfer in hectograph units; it is such materials to which the invention is directed for preventing 10 premature transfer. Compositions containing beeswax, castor oil and combinations of waxes and oils fall into this category and are well known in the art. The wax itself may contain oil and oil addition is not then necessary.

The three dimensional cellular structure of the coating 15 does not collapse significantly under marking pressure, and the coated master does not change opacity. While the cells contribute to opacity, the important contribution in hectographic masters is their capacity for resisting migration, particularly upon product aging; thus opacity contribution by the cells is a bonus permitting the use of somewhat thinner coatings and thereby providing some additional flexibility in a master of a given paper basis weight. Such papers are commonly of themselves white or very light in color, and the coating is also suitably so as 25 in the specific examples set forth above.

By "normal" contact of the master and transfer sheet as described herein is meant face-to-face contact under pressures occurring in shipment storage and the like in contrast to marking pressures which are non-uniformly 30 applied as concentrated forces to the hectograph unit.

It will be understood that this invention is susceptible to modification in order to adapt to different usages and conditions and, accordingly, it is desired to comprehend such modifications within the invention as may fall within 35 the scope of the appended claims. What is claimed is:

1. A hectograph master unit comprising a master sheet and a transfer sheet detachably joined along one edge, said transfer sheet having a transferable hectographic oilwax coating composition on a face thereof, said master sheet having on a face fronting the oil-wax coating composition a proteinaceous binder coating which is cellular, pigment containing, and resistant to migration of the oilwax component thereto under simple contact pressure but which is adapted under marking pressure to receive and retain the oil-wax component of the transfer sheet, said proteinaceous binder coating having at least 50% by weight of protein and not more than 50% by weight of water dispersible binder, there being for each volume of binder between about 0.25 to 0.84 volumes of pigment and also for each volume of binder between about 1 to 6 volumes of cells.

2. A hectograph master unit as claimed in claim 1 and in which protein is the only binder component.

3. A hectograph master unit as claimed in claim 1 and in which the binder contains at least 50% by weight of protein and starch as a binder to the extent of not more than 50% by weight.

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