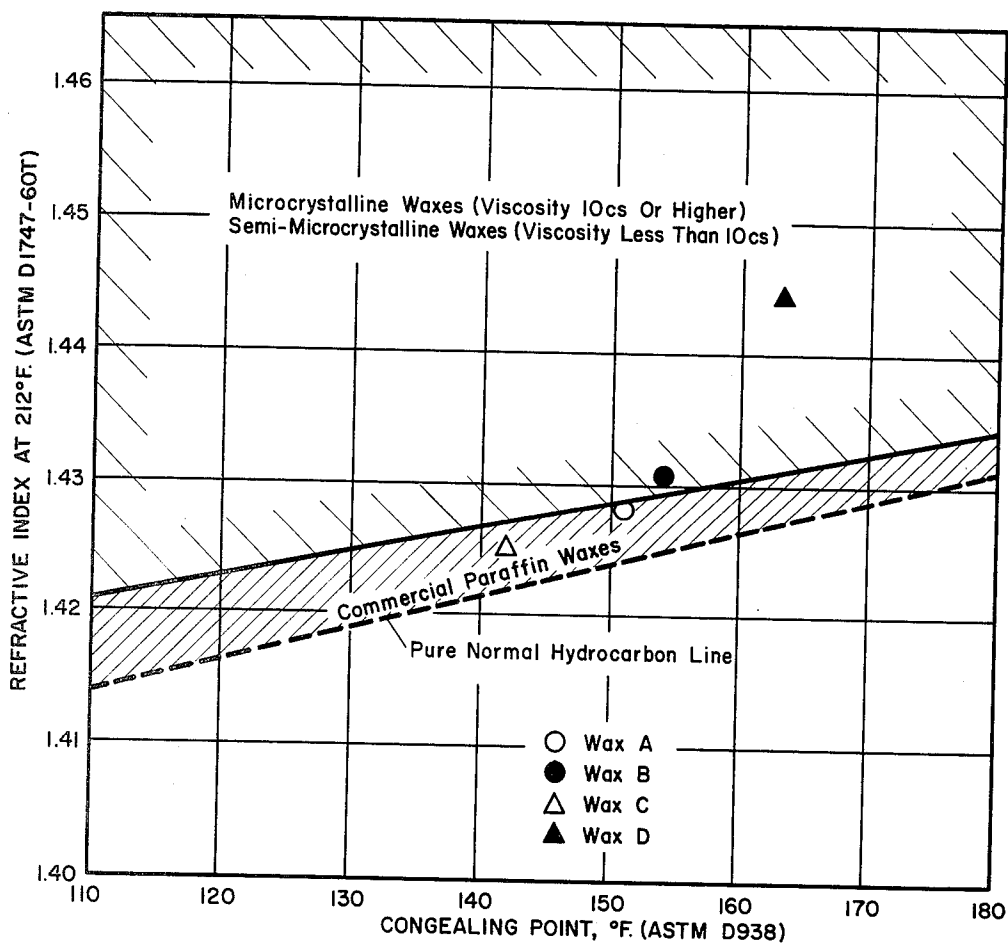


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COMPOSITIONS CONTAINING PARAFFIN WAX, SEMI-MICROCRYSTALLINE
WAX, ETHYLENE-VINYL ACETATE COPOLYMER AND POLYETHYLENE
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COMPOSITIONS CONTAINING PARAFFIN WAX, SEMI-MICROCRYSTALLINE WAX, ETHYLENE-VINYL ACETATE COPOLYMER AND POLY-ETHYLENE

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The present invention is concerned with an improved wax-plastic composition which is particularly adapted for the coating of fibrous substances such as paper and the like. In accordance with the present invention, an unexpectedly improved wax-plastic composition is secured by the blending of a specific paraffin wax, a semi-microcrystalline and/or a micro wax and a copolymer of ethylene and vinyl acetate having certain critical physical properties. Particularly desirable wax compositions comprise the use of polyethylene in conjunction with the above formulations. The resulting compositions yield coatings which exhibit superior quality in a number of respects, e.g., moisture barrier, oil resistance, scuff resistance, gloss and sealing strength. These are the performance qualities widely sought for package coatings for a number of end uses.

In the refining of hydrocarbon oils such as petroleum oils, it is known to segregate paraffin waxes from so-called paraffin distillates, waxy lubes and the like. The segregation of these waxes is secured by a number of processes. For example, it is known to chill the selected wax-containing fraction in order to secure crystallization of the wax and to remove the wax crystals from the oil by filtering, centrifuging and the like. It is also known to use various dewaxing solvents such as liquid normally gaseous hydrocarbons, such as propane, as well as other solvents, such as methylethyl ketone and the like. It is also known to utilize in dewaxing operations solvent mixtures wherein one solvent comprises a wax precipitating solvent while the other comprises a solvent having a high solubility for oil. A solvent mixture of this character, for example, comprises 60% by volume of toluene and 40% volume of methylethyl ketone. In utilizing a mixture of this character, it has been the practice to add the mixture in toto or incrementally to the waxy distillate as it is being chilled. In dewaxing operations, it is also known to use various filter aids and other agents in order to render the dewaxing and filtering operations more efficient.

The wax segregated from the hydrocarbon oil, termed "slack wax," usually contains from about 10% to 40% of oil. The slack wax is refined either by conventional sweating or by solvent crystallization to produce "crude scale wax" in a manner to reduce the oil content to less than about 5% by weight. The slack wax may be distilled to obtain the desired boiling range prior to sweating, if desired. This "crude scale wax" generally has an oil content of about 1% to 3% by weight. In order to remove this oil from the scale wax to produce a refined wax having an oil content below about 0.5%, usually below about 0.3%, various procedures have been proposed and employed.

It is also known in the art to separate a petroleum

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wax which may be identified as a semi-microcrystalline wax from distillate lubricating oil fractions. The process for this preparation differs from the process for paraffin wax manufacture in the conditions of crystallization.

Paraffins are prepared under conditions of relatively higher temperatures in a solvent crystallization process to yield a greater amount of straight chain hydrocarbon fractions. Semi-microcrystalline waxes are prepared at relatively lower temperatures, which cause crystallization of more of the so-called non-normal hydrocarbons, which include the iso-paraffinic and cyclic hydrocarbon structures.

It is also known in the art to segregate microcrystalline waxes from residual oils. These microcrystalline waxes are of a relatively high melting point and of different crystalline structure than the paraffin waxes described in the foregoing paragraphs. The microcrystalline wax is obtained from a so-called petrolatum which may be prepared from any of the paraffinic or mixed base crude oils. The undistilled residue may be treated with sulfuric acid and neutralized to remove the tarry matter and unsaturated hydrocarbons. The undistilled residue also may be deasphalted. The treated stock, containing a fairly high percentage of wax, as evidenced by a very high pour point, may be dewaxed by blending with a dewaxing solvent, such as propane, methylethyl ketone-benzol, or petroleum naphtha, chilling, and filtering or centrifuging to separate the petrolatum stream from the oil solution.

This deoiling operation produces the so-called petrolatum, which contains microcrystalline wax together with about 10 to 30% oil. The wax may be again put in solution with more solvent or naphtha and chilled, and filtered or recentrifuged to further reduce the oil content. The wax which separates in these operations is referred to as crude microcrystalline wax. This wax, separated in the second deoiling process, after stripping to remove solvent is fairly dry and of a low oil content. This wax should not be confused with petroleum jellies which contain large amounts of oil. The crude microcrystalline wax may be again treated to remove color and odor constituents. In one method of treatment, the crude wax is again put into solution with naphtha and filtered through clay or an equivalent material in order to improve its color. The clay filtered solution is distilled to remove the naphtha, the residue being a refined microcrystalline wax having a melting point within the range of about 122° F. to 180° F. Additionally, color may be improved by hydrofining the microwax. The source of the crude oil, the conditions of crystallization and the oil content of the refined microcrystalline product will affect the melting point of the final wax product. The refined microcrystalline wax, sometimes called amorphous wax, is of very small crystal structure.

For purposes of this invention a system for classifying waxes based upon certain physical constants of the waxes is used. By this system, its congealing point in °F., is measured by ASTM D938, its refractive index at 212° F., is measured by ASTM D1747-60T, and its viscosity at 210° F., in centistokes, is measured by ASTM D445. The data on congealing point and refractive index are then plotted and compared with the standard reference plot of pure normal hydrocarbons which is reproduced

in the drawing, which is the only figure in this case. In this plot, the position of the normal hydrocarbons line is expressed by the equation:

$n_D^{212} = .0002514 t + 1.3861$ where t = congealing point in ° F. and n_D^{212} = refractive index at 212° F. The solid line, which separates commercial paraffin waxes from semimicrocrystalline and microcrystalline waxes, is expressed by the equation:

$n_D^{212} = .0001943 t + 1.3994$ where t = congealing point in ° F. and n_D^{212} = refractive index at 212° F. For purposes of this invention, waxes which have a congealing point and refractive index such that their coordinates fall in the area between the two lines, are identified as paraffin waxes. Waxes having coordinates which fall on or above the paraffin wax line are identified as either semi-microcrystalline or microcrystalline, depending upon their viscosity. A wax with coordinates above the solid line and having a viscosity of less than 10 cs. is identified as a semi-microcrystalline wax, while a wax with coordinates above the solid line and having a viscosity of 10 cs. or greater is identified herein as a microcrystalline wax.

For the purposes of the present invention the following precise definitions are used:

Paraffin wax.—A petroleum wax consisting principally of normal alkanes, having an observed refractive index at 212° F. lower than that given by the expression $n_D^{212} = 0.0001943 t + 1.3994$ but higher than that given by the expression $n_D^{212} = 0.0002514 t + 1.3861$, where t is the observed congealing point, ° F.

Semi-microcrystalline wax.—A petroleum wax containing substantial proportions of hydrocarbons other than normal alkanes, having an observed refractive index at 212° F. equal to or higher than that given by the expression $n_D^{212} = 0.0001943 t + 1.3994$, where t is the observed congealing point, ° F. and having a viscosity at 210° F. below 10 centistokes.

Microcrystalline wax.—A petroleum wax containing substantial proportions of hydrocarbons other than normal alkanes, having an observed refractive index at 212° F. equal to or higher than that given by the expression $n_D^{212} = 0.0001943 t + 1.3994$ where t is the observed con-

formance. For instance, U.S. 2,756,217 teaches the addition of butadiene-styrene copolymer to wax coating compositions. Also U.S. 2,877,196 discloses a wax coating composition containing a copolymer of ethylene-vinyl acetate in conjunction with either a paraffin or microcrystalline wax.

It has now been found that wax compositions containing an unusually high melting paraffin wax of 150° F. (Wax A), a semi-microcrystalline wax melting at 155° F. (Wax B), polyethylene and an ethylene-vinyl acetate copolymer having certain critical physical properties will have unexpectedly superior flexibility characteristics while maintaining the excellent hardness properties expected of a brittle, high melting paraffin wax blend. This makes our composition of broader value in a wide variety of end use applications than previously obtainable. In particular, it has been discovered that adding a copolymer of ethylene-vinyl acetate having the following characteristics:

Ethylene-vinyl acetate copolymer "X"

(1) Inherent viscosity at 30° C. (0.25% by wt. in toluene) -----	0.85
(2) Comonomer ratio, wt. percent ethylene/vinyl acetate -----	72/28
(3) Melt index (ASTM D 1238-57T) -----	15
(4) Tensile strength, p.s.i. -----	2000
(5) Elongation at break, percent -----	750
(6) Density, g./cc. at 30° C. -----	0.95
(7) Refractive index, n_D^{25} -----	1.482
(8) Softening point, ring and ball, ° F. -----	276

to a wax composition containing a wax such as Wax A will yield a coating which will have unexpectedly superior flexibility properties when compared to coatings containing lower melting and more flexible paraffin waxes combined with the same ethylene-vinyl acetate copolymer. Furthermore, this enhancement of the coating's flexibility properties does not arise when a very similar but lower molecular weight ethylene-vinyl acetate copolymer is used in conjunction with Wax A discussed above. The properties of the waxes described in this paper are summarized in Table I.

Table I

Composition, Wt. Percent	Wax A	Wax B	Wax C	Wax D	Wax F
140° F. Paraffin Wax -----			100		
150° F. Paraffin Wax -----	100				60
155° F. Semi-Micro Wax -----		100			40
175° F. Micro Wax -----				100	
Kinematic Viscosity at 210° F., cs. -----	5.6	7.04	4.3	21.1	
R.I. at 212° F. -----	1.4282	1.4309	~1.425	1.4447	
Congeaing Point, ° F. -----	151	154	~142	163	
Characterization ^a -----	Paraffin	Semi-Micro	Paraffin	Micro	
ASTM Melting Point, ° F. -----	150		140	175	
Petrolatum Melting Point, ° F. -----		155			
Tensile Strength, p.s.i.: -----					
At 73° F. -----	262	315	301		275
At 40° F. -----	383	311			352
Elongation, in. X 10 ³ : -----					
At 73° F. -----	25	31	20		21
At 40° F. -----	23	19			20
Plastic Flow, in. X 10 ³ : -----					
At 73° F. -----	9	13	9		9
At 40° F. -----	7	6			6
Scuff, mgs./100 in. ² , Coating on Sulfite Paper -----	30.0	22.8	35.0		23.4

^a As described by the chart in Figure 1.

^b 2,000 gram load, smooth side of abrasive paper.

gealing point, ° F. and having a viscosity at 210° F. of 10 centistokes or higher.

Petroleum waxes produced as described above have been extensively used in the coating art, particularly where liquid proofness and moisture proofness are desired. Furthermore, addition of various polymers and copolymers to paraffin and microcrystalline wax compositions is also known as a means of enhancing certain performance features of the wax, for certain end-use applications, such as the wax's over-all coating per-

A preferred composition range for the present invention is 60-95% of Wax A and 5-40% of copolymer X in the two component embodiment. When Wax B and polyethylene are also added the preferred composition range is 42-55% of Wax A, 28-37% of Wax B, 6.5-25% of copolymer X and 1.5-5% of polyethylene. Specific desirable compositions are described in the examples given later in this paper.

Table II compares the coating properties of Wax C with that of Wax A when blended with a constant amount

of copolymer X as identified above. It is obvious from these results that the basically harder Wax A yields blends which are superior to Wax C in the hardness and toughness properties of scuff and tensile strength. At the same time, blends based on Wax A show unexpectedly superior flexibility properties, as measured by the elongation and plastic flow tests. Table II also includes blends of the above paraffin waxes with Wax D and a low molecular weight polyethylene (M.W. approx. 1,500). These additives, as is known in the art, produce coating compositions having enhanced physical properties. However, the singular feature of Wax A yielding blends superior in both hardness and flexibility properties compared to that of the Wax C remains unaltered. Unexpectedly it has additionally been found that Wax B can be successfully used in the above blends without the loss of any desirable properties, while enhancing certain critical properties of the coating. Furthermore, the addition of polyethylene in minor amounts to these coating compositions will also increase the values of the desirable physical properties of the blend. A preferred polyethylene form is one that would have a molecular weight in the range of 1,000 to 20,000, preferably 1,500. Use of such low molecular weight polyethylene yields the desired coating properties without increasing the viscosity of the blend, which tends to be high due to the presence of the ethylene-vinyl acetate copolymer.

Each of the physical properties of the coating compositions given in Table II is important in one or more of the end-use applications. The methods by which these properties are determined are given below:

(a) *Scuff*.—The method used to determine the scuffing tendency of the various wax-copolymer blends uses a modification of the Marathon scuff tester, using 3-M Tri-M-ite abrasive-polishing paper as the scuffing agent. By this method, sulphite paper coated with common paraffin waxes gives a scuff tendency of about 30–40 mgs. of coating scuffed off per 100 in.² of surface when loaded with a 2000 gram weight using the polishing side of the paper as the scuff medium. The scuffing tendency of wax coated paper is an important property for some end-use applications where a hard, difficult-to-scratch surface is desired (such as for milk cartons, frozen food cartons, etc.).

(b) *Tensile strength*.—This property is determined by stretching a dumb-bell mold of wax (at 40° F.) until it fails on an Instron Tensile Tester. When tested, the mold is placed in the jaws of the Tensile Tester so that the initial jaw separation is 0.8" and the cross section of the neck of the mold is 0.5" x 0.5" in size. Due to the rather high viscosity encountered with blends having relatively large concentration of the ethylene-vinyl acetate copolymer, it is necessary, in order to get reproducible results, to preheat the molds to the temperature of the molten wax (220–250° F.). If this is not done, the test wax will shrink appreciably during the cooling and will give erroneous results on the Instron Tensile Tester. The tensile strength is a measure of the maximum force per unit area that the wax can withstand before failure. An average value for ordinary paraffin waxes is 300 p.s.i.

(c) *Elongation and plastic flow*.—These properties are also determined by subjecting a wax mold of the desired blend to stretching in an Instron Tensile Tester until the wax fails. The sample mold is dumb-bell shaped and has the dimensions described in the tensile strength test. The properties of elongation and plastic flow are a measure of the toughness and the flexibility of the wax. This is important for such applications as milk carton coatings, frozen food coatings, bread wraps, etc., where the wax coating must withstand bending or dropping without breaking or cracking at temperatures near or below the freezing point of water. Elongation is the total stretch in inches of the sample from the initial stress point to the breaking point. Plastic flow is the inelastic distortion and is measured from the point of inelastic

stretching to the breaking point. Average values for normal paraffin waxes based on molds having a 0.8" effective test length are: elongation= 30×10^{-3} in. which is equivalent to about 4% longitudinal elongation and plastic flow= 5×10^{-3} in. which is equivalent to about 1% of longitudinal elongation.

Table II

COATING PROPERTIES

[Ethylene—vinyl acetate copolymer X=25% +]

Scuff, mgs./100 in.² (2,000 g. load):

75% Wax C=10.0

75% Wax A=3.4

65% Wax C+10% Wax D=4.0

65% Wax A+10% Wax D=2.6

Tensile strength, p.s.i. (Instron @ 40° F.):

75% Wax C=672

75% Wax A=711

65% Wax C+10% Wax D=732

65% Wax A+10% Wax D=753

Elongation, in. $\times 10^{-3}$ (Instron @ 40° F.):

75% Wax C=66

75% Wax A=97

65% Wax C+10% Wax D=86

65% Wax A+10% Wax D=121

60% Wax C+10% Wax D+5% lmw.¹ polyethylene=10760% Wax A+10% Wax D+5% lmw.¹ polyethylene=121Plastic flow, in. $\times 10^{-3}$ (Instron @ 40° F.):

75% Wax C=32

75% Wax A=63

65% Wax C+10% Wax D=53

65% Wax A+10% Wax D=92

60% Wax C+10% Wax D+5% lmw.¹ polyethylene=7860% Wax A+10% Wax D+5% lmw.¹ polyethylene=89¹ Low molecular weight.

Table III illustrates the critical nature of the choice of the particular ethylene-vinyl acetate copolymer. It compares wax blends derived from the ethylene-vinyl acetate copolymer X whose composition has been previously described in this paper with those derived from an ethylene-vinyl acetate copolymer Y having the same chemical properties but different physical properties. The latter copolymer has the following characteristics:

Ethylene-vinyl acetate copolymer Y

(1) Inherent viscosity at 30° C. (0.25% by wt. in toluene)	0.78
(2) Comonomer ratio, wt. percent ethylene/vinyl acetate	72/28
(3) Melt index (ASTM D 1238–57T)	25
(4) Tensile strength, p.s.i.	1000
(5) Elongation at break, percent	700
(6) Density, g./cc. at 30° C.	0.95
(7) Refractive index, n_D^{25}	1.482
(8) Softening point, ring and ball, ° F.	255

The copolymers X and Y are each blended with Wax E which is a mixture of Wax B and C, and Wax F, a mixture of Wax A and B (as shown in Table I). The results show quite clearly that unexpected higher flexibility values for the blends containing Wax A compared with similar blends containing Wax C occur when Wax A is combined with copolymer X but not when the same waxes are combined with copolymer Y. Also, the table shows that the addition of Wax B improves the properties of both Wax A and Wax C in the cases where copolymer X is used. This improvement is not found when Wax B is added to Wax A and Wax C in conjunction with copolymer Y.

Table III

COATING PROPERTIES			
Ethylene-Vinyl Acetate Copolymer X		Ethylene-Vinyl Acetate Copolymer Y	
Wax F Exhibits Superior Properties		Wax F Shows Little or No Improvement	
Scuff mgs./100 in. ² (2,000 g. load)			
75% Wax C+25% Copolymer X	10.0		
75% Wax A+25% Copolymer X	3.4	75% Wax A+25% Copolymer Y	4.6
75% Wax E+25% Copolymer X	3.3	75% Wax E+25% Copolymer Y	5.0
75% Wax F+25% Copolymer X	<1.6	75% Wax F+25% Copolymer Y	3.3
Tensile Strength, p.s.i. (Instron at 40° F.)			
75% Wax C+25% Copolymer X	672		
75% Wax A+25% Copolymer X	711	75% Wax A+25% Copolymer Y	711
75% Wax E+25% Copolymer X	779	75% Wax E+25% Copolymer Y	676
75% Wax F+25% Copolymer X	816	75% Wax F+25% Copolymer Y	673
Elongation, in. $\times 10^{-3}$ (Instron at 40° F.)			
75% Wax C+25% Copolymer X	66		
75% Wax A+25% Copolymer X	97	75% Wax A+25% Copolymer Y	82
75% Wax E+25% Copolymer X	84	75% Wax E+25% Copolymer Y	86
75% Wax F+25% Copolymer X	108	75% Wax F+25% Copolymer Y	86
Plastic Flow, in. $\times 10^{-3}$ (Instron at 40° F.)			
75% Wax C+25% Copolymer X	32		
75% Wax A+25% Copolymer X	63		
75% Wax E+25% Copolymer X	54	75% Wax E+25% Copolymer Y	54
75% Wax F+25% Copolymer X	77	75% Wax F+25% Copolymer Y	56

Table IV discloses that the enhancing effect of copolymer X on the flexibility characteristics of wax blends containing Wax A are in evidence over a rather wide range of copolymer X concentrations. Further, Wax A maintains its superior hardness properties as shown by its consistently lower scuff values.

resin, e.g. M.W.=1,500, to 250° F. with good agitation (a "Lightnin" mixer or similar equipment should be employed). Copolymer X (ethylene-vinyl acetate copolymers having the requisite physical properties defined for copolymer X are commercially available) is then added with continuous stirring until a total of about 6.5 parts

Table IV
INSTRON FLEXIBILITY AT 40° F.

	Tensile Strength, p.s.i.	Elongation, in. $\times 10^{-3}$	Sealing Strength, gms./in.	Scuff, ¹ mgs./100 in. ²
Typical Paraffin Wax	252	17	10	35.0
With 5% Copolymer X:				
Wax C	502	30	22	28.9
Wax A	568	30	25	24.8
With 15% Copolymer X:				
Wax C	687	50	105 FT ²	14.2
Wax A	668	66	160 FT	5.9-8.4
With 25% Copolymer X:				
Wax C	672	66	260 FT	10.0
Wax A	711	97	300 FT	3.4
Wax A+10% Wax B	753	121	400 Paper Tear ³	2.6
Wax A+25% Wax B ⁴	816	108	do.	<1.6
With 40% Copolymer X:				
Wax C	860	184	500 Paper Tear	<1
Wax A	773	550	do.	<1

¹ Using 2,000 gm. load and Tri-M-ite polishing paper.

² Fiber tear observed during the strength test, as partial picking of the fibers from the paper.

³ Paper tear is observed during the strength test, when the wax seal is so strong that the paper substrate tears apart, rather than the wax seal.

⁴ Also containing 5% low molecular weight polyethylene.

Compositions containing blends of Wax A and copolymer X are especially suitable for use as coatings on milk cartons, frozen food and other cold storage containers, nested containers, and flexible packages, and as hot melt adhesives. Further possible uses include direct coatings for food such as meats and cheeses, coatings for metallic surfaces as in cans, binders or nonskid backings for rugs and fabrics, coatings for corrugated cartons and other laminated structures, low cost dielectric coatings, and molding compounds.

Example I

An excellent coating composition is prepared by heating about 55 parts of Wax A, about 37 parts of Wax B, and about 1.5 parts of a low molecular weight polyethylene

of the copolymer resin has been introduced. The copolymer should be added slowly so as to insure uniform dispersion. The stirring is then continued until no resin particles remain (30-60 minutes). The finished blend can be pumped directly to coating equipment or can be cast into slabs for subsequent remelting prior to use.

Example II

The method of Example I is used to prepare a blend containing the following proportion of materials:

	Parts
Wax A	53
Wax B	35
Copolymer X	10
Polyethylene (low m.w. 1500)	2

Example III

The method of Example I is used to prepare a blend containing the following proportion of materials:

	Parts
Wax A -----	46
Wax B -----	30
Copolymer X -----	20
Polyethylene (low m.w. 1500) -----	4

Example IV

The method of Example I is used to prepare a blend containing the following proportion of materials:

	Parts
Wax A -----	42
Wax B -----	28
Polyethylene (low m.w. 1500) -----	5
Copolymer X -----	25

The excellent properties of the wax-copolymer blends given in the above examples are summarized in Table V. Each of these compositions will be especially desirable for particular end uses, which uses will be obvious to one skilled in the art.

Table V

PROPERTIES OF COATING COMPOSITIONS

	Example			
	I	II	III	IV
Petrolatum M.P., ° F.-----		168.5	171	181
Viscosity, cs., at 210° F.-----		94.5	1,030	2,700
Viscosity, cs., at 250° F.-----	122.5	57.7	523	1,160
Tensile Strength at 40° F., p.s.i.-----	535	670	764	816
Elongation at 40° F., in.×10 ⁻³ -----	36	48	134	108
Scuff, ² mgs./100 in. ² -----	14.6	7.0	<1.6	<1.6

¹ Brookfield viscosity in cps. using Model LVF, #2 Spindle at 60 r.p.m.
² 3-M Tri-M-ite polishing paper with a 2,000 gm. load.

It is understood that this invention is not limited to the specific examples, which examples have been offered merely for the purpose of illustration, and that modifications may be made thereof without departing from the spirit of the invention.

What is claimed is:

1. An improved wax composition consisting essentially of about 42-55% of a paraffin wax having a congealing point of about 151° F., a refractive index of about 1.4282

at 212° F. and a kinematic viscosity at 210° F. of about 5.6 cs., about 28-37% of a semi-microcrystalline wax having a congealing point of about 154° F., a refractive index of about 1.4309 at 212° F. and a kinematic viscosity at 210° F. of about 7 cs., about 6.5-25% of an ethylene-vinyl acetate copolymer, said copolymer having a comonomer ratio of ethylene to vinyl acetate of about 72 to 28, an ASTM melt index of about 15 and a ring and ball softening point of about 276° F., and about 1.5-5% polyethylene having a molecular weight in the range of 1,000 to 20,000.

2. The composition of claim 1 consisting essentially of about 55% of said paraffin wax, about 37% of said semi-microcrystalline wax, about 6.5% of said ethylene-vinyl acetate copolymer and about 1.5% polyethylene of about 1,500 molecular weight.

3. The composition of claim 1 consisting essentially of about 53% of said paraffin wax, about 35% of said semi-microcrystalline wax, about 10% of said ethylene-vinyl acetate copolymer and about 2% polyethylene having a molecular weight of about 1,500.

4. The composition of claim 1 consisting essentially of about 46% of said paraffin wax, about 30% of said semi-microcrystalline wax, about 20% of said ethylene-vinyl acetate copolymer and about 4% polyethylene having a molecular weight of about 1,500.

5. The composition of claim 1 consisting essentially of about 42% of said paraffin wax, about 28% of said semi-microcrystalline wax, about 25% of said ethylene-vinyl acetate copolymer, and about 5% polyethylene having a molecular weight of about 1,500.

6. An article of manufacture comprising a surface coated with the composition described in claim 1.

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