MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS - 1963
We, MONSANTO COMPANY, a Corporation organised and existing under the laws of the State of Delaware, United States of America, having its principal place of business at 800 North Lindbergh Boulevard, St. Louis, State of Missouri, United States of America, hereby apply for the grant of a Patent for an invention entitled:

PROCESSING AID FOR POWDER COATINGS,

which is described in the accompanying complete specification. This application is a Convention application and is based on an application/s numbered 444,452 for a patent or similar protection made in United States of America on 21st February, 1974.

Our address for service is care of EDWIN F. WELLINGTON, Patent Attorney, 457 St. Kilda Road, Melbourne, in the State of Victoria, Commonwealth of Australia.

DATED this 19th day of February, A.D.1975

For and on behalf of MONSANTO COMPANY,

Edwin F. Wellington
Patent Attorney for Applicant Company

To: The Commissioner of Patents,
COMMONWEALTH OF AUSTRALIA.
COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952

FORM 7  REGULATION 11(2)

DECLARATION IN SUPPORT OF A CONVENTION APPLICATION
UNDER PART IV FOR A PATENT OR PATENT OF ADDITION.

In support of the Convention Application made under Part IV of the Act by MONSANTO COMPANY for a patent for an invention entitled

PROCESSING AID FOR POWDER COATINGS

1. Neal Edmond Willis, International Patent Counsel, Monsanto Company, of 800 North Lindbergh Boulevard, St. Louis, 63166, in the State of Missouri, United States of America, do solemnly and sincerely declare as follows:

1. I am authorized by MONSANTO COMPANY, the applicant for the Patent to make this declaration on its behalf.

2. The basic application as defined by Section 141 of the Act was made at the Patent Office, Washington, District of Columbia, in the United States of America on the 21st of February 1974, by Samuel Francis Joyce (U.S. SERIAL No. 444,452)

3. Samuel Francis Joyce, III, 410 Nottingham Drive, Ballwin, Missouri 63011, U.S.A.

4. The basic application referred to in paragraph 2 of this declaration was the first application made in a Convention country in respect of the invention, the subject of the application.

DECLARED at St. Louis, Missouri, aforesaid this 8th day of January, 1975.

NEAL EDMOND WILLIS

To Commissioner of Patents
COMMONWEALTH OF AUSTRALIA
The following statement is a full description of this invention including the best method of performing it known to me/us:
Background of the Invention

Recently, the use of powder coatings in industrial applications has increased greatly. Generally, powder coatings are dry coating compositions which may be applied by dip or spray methods. If used as a spray, the powder is normally applied electrostatically. As with all coating compositions, the coating must be formulated to avoid various problems such as pitting, cracking, uneven spread and other problems.

The use of processing aids to avoid the above-mentioned problems in conventional coating compositions, i.e. liquid coating compositions, is known. There are a variety of processing aids useful for such purposes, but most of such processing aids are in liquid form. The use of liquid processing aids in powder coating formulations has been attempted, but leads to further problems, since a liquid of any kind cannot be evenly distributed throughout the powder and tends to agglomerate the powder.

In the above-identified copending application, processing aids for a variety of resins are disclosed. The processing aids are liquid acrylate copolymers. Attempts have been made to use such liquid acrylate copolymers in powder coating formulations, but the resultant product is generally not satisfactory because of the agglomeration problem. Normally, manufacturers of powder coatings prefer that processing aids be added at the point of use so that the concentration of the processing aid in the powder coating resin can be varied, depending on the particular
use intended for the powder coating. Because of the difficulty of incorporating liquids into powders, however, manufacturers have had to resort to masterbatching the powder and the liquid processing aid. The masterbatch technique is, however, costly, time consuming and inconvenient. For example, masterbatching requires heating and stirring of the powdered resin and liquid processing aid for 6 to 8 hours.

Accordingly, there is a need for a liquid processing aid which is suitable for use in powder coatings.

**Summary of the Invention**

In accordance with the present invention, there is provided a liquid processing aid in powdered form which may easily be incorporated uniformly in a powder coating composition. The powdered processing aid of the present invention comprises an acrylic copolymer and a carrier therefor. The powdered processing aid is prepared by drying a mixture of the liquid acrylate copolymer and solid carrier from an organic solvent or a water dispersion or emulsion, for example, by spray drying. The resulting free flowing powder is widely applicable as a processing aid in powder coating formulations.

**Description of the Preferred Embodiment**

The active ingredient of the processing aid composition of the present invention is a copolymer of an alkyl acrylate ester with a dissimilar alkyl acrylate ester. The copolymers are normally liquids. The copolymers consist of (1) from about 5 to about 95 percent by weight of an alkyl acrylate ester wherein the alkyl constituent contains from about 1 to about 12 carbon atoms and (2) from about 95 to about 5 percent by weight of a dissimilar alkyl acrylate ester wherein the alkyl constituent contains from about 3 to about 24 carbon atoms. In a preferred embodiment of the invention, the copolymers which are an active ingredient of the processing composition of the invention consist of (1) from...
about 5 to about 95 percent by weight of an alkyl acrylate ester wherein the alkyl constituent contains from about 1 to about 12 carbon atoms and (2) from about 95 to about 5 percent by weight of a dissimilar alkyl acrylate ester wherein the alkyl constituent contains from about 3 to about 18 carbon atoms. In a yet more preferred embodiment of this invention, the copolymers consist of (1) from about 20 to about 50 percent by weight of an alkyl acrylate ester wherein the alkyl constituent contains from about 2 to about 8 carbon atoms and (2) from about 80 to about 50 percent by weight of a dissimilar alkyl acrylate ester wherein the alkyl constituent contains from about 6 to about 12 carbon atoms.

The acrylate copolymers utilized herein are characterized by their liquidity, ranging from free-flowing to viscous, at room temperature or slightly above. The copolymers have an average molecular weight of from about 2,000 to about 10,000, a preferred molecular weight range being from about 2,000 to about 6,000. A particularly useful molecular weight range is from about 3,500 to about 5,000.

Tough or rubbery acrylate materials, such as those frequently used as extenders or impact value modifiers, are unsuitable for use as processing aids for purposes of this invention.

The acrylic copolymer esters utilized herein, and their methods of production are well known to the art. The copolymers can be obtained, for example, by solution polymerization of a mixture of two monomers in an organic diluent while in contact with a peroxidic catalyst. Another method for preparing the copolymer processing aids is by mass polymerization wherein a mixture of monomeric ingredients is mixed with a polymerization catalyst in the substantial absence of a solvent at a temperature sufficient to cause polymerization. Emulsion polymerization procedures which entail polymerizing a mixture of monomers as
an emulsion are also suitable for the preparation of the pro-
cessing aids of this invention.

Exemplary liquid copolymers, prepared from dissimilar
alkyl acrylates, which are useful as processing aids (in powdered
form) for powder coatings include:

50 parts 2-ethylhexyl acrylate and 50 parts octadecyl
acrylate (Mol. Wt. = 4,040);

30 parts ethyl acrylate and 70 parts 2-ethylhexyl
acrylate (Mol. Wt. = 4,690);

30 parts ethyl acrylate and 70 parts 2-ethylhexyl
acrylate (Mol. Wt. = 3,600);

40 parts ethyl acrylate and 60 parts 2-ethylhexyl
acrylate (Mol. Wt. = 6,760);

90 parts methyl acrylate and 10 parts butyl acrylate
(Mol. Wt. = 10,230);

80 parts butyl acrylate and 20 parts lauryl acrylate
(Mol. Wt. = 5,410);

40 parts butyl acrylate and 60 parts lauryl acrylate
(Mol. Wt. = 6,790);

30 parts ethyl acrylate and 70 parts 2-ethylhexyl
acrylate (Mol. Wt. = 4,760);

90 parts methyl acrylate and 10 parts butyl acrylate
(Mol. Wt. = 5,320);

90 parts methyl acrylate and 10 parts butyl acrylate
(Mol. Wt. = 2,100);

8 parts propyl acrylate and 92 parts tetracosyl
acrylate;

85 parts hexyl acrylate and 15 parts propyl acrylate;
20 parts nonyl acrylate and 80 parts eicosyl acrylate;
25 parts heptyl acrylate and 75 parts nonyl acrylate;
35 parts amyyl acrylate and 65 parts dodecyl acrylate;
45 parts decyl acrylate and 55 parts heptadecyl
acrylate;
15 parts dodecyl acrylate and 85 parts butyl acrylate;
75 parts dodecyl acrylate and 25 parts pentadecyl acrylate;
60 parts nonyl acrylate and 40 parts decyl acrylate;
45 parts ethyl acrylate and 55 parts dodecyl acrylate;

A further innovative feature of the present invention is the discovery that alkylated benzenes and other plasticizers appear to act in conjunction with acrylate copolymers as a processing aid. Thus a mixture of alkylated benzene and acrylate copolymer may be spray dried on a carrier to afford a useful processing aid.

Although the exact nature of the action of alkyl benzenes or other plasticizers in admixture with acrylate copolymers is not known, it is believed that the plasticizers serve as a diluent for the acrylate copolymer, thus allowing the acrylate copolymer to more easily and evenly flow over the solid carrier particles. The plasticizer also functions as a processing aid.

The alkylated benzene which may be used in the powdered processing aid of the invention may be any benzene alkylate wherein the total number of carbon atoms in the alkyl portion of the molecule is from about 2 to about 20 carbon atoms. Thus, benzene may be mono-, di- or tri-alkylated so long as the total number of carbon atoms in the alkyl portions of the molecule are within the above range. Typical alkylated benzenes which may be used in the processing aid of the present invention include ethylbenzene, xylene, mesitylene, diethylbenzene, propylbenzene, butylbenzene, cumene, hexylbenzene, octylbenzene, decylbenzene, didecylbenzene, dodecylbenzene, hexadecylbenzene, octadecylbenzene and the higher alcohols, including dimer alcohols, commonly referred to as "heavy oxo ends" and which remain after distillation of crude alcohol produced by the "oxo" synthesis from olefins.
Other plasticizers which may be used in the processing aid composition of this invention include alkylated aromatic petroleum fractions, fatty acid esters, aromatic and aliphatic acid diesters, triesters of aliphatic acids, higher esters such as pentaerythritol esters, phosphates, sulfonamides, nitrogen-containing aromatics, ester-amides, nitriles, chlorinated esters and polymeric.

Specifically, fatty acid ester plasticizers which are contemplated include laurates, myristates, palmitates, oleates, stearates, ricinoleates and the like containing from about 15 to about 25 carbon atoms, e.g. ethylene glycol monolaurate, n-butyl laurate, butyl cellosolve laurate, n-butyl myristate, n-butyl palmitate, methyl hydroxystearate, n-butyl oleate, methyl ricinoleate and similar compounds.

Aromatic acid diesters include phthalates such as dimethyl phthalate, dioctyl phthalate, di-C\textsubscript{7}-C\textsubscript{11} alkyl phthalates, butyl benzyl phthalate, diphenyl phthalate, dicyclohexyl phthalate, di-2-ethylhexyl phthalate.

Aliphatic acid diesters include adipates such as diisobutyl adipate, di-2-ethylhexyl adipate, azelates, e.g. di-2-ethylhexyl azelate, sebacates, e.g. dimethyl sebacate, diiso-octyl sebacate, polyglycols, e.g. triethylene glycol di(2-ethylbutyrate) and the like.

Aliphatic acid triesters include acetates, e.g., triacetin, butyrates, e.g. tributyrin, citrates, e.g. triethyl citrate, ricinoleates, e.g. glyceryl triricinoleate and the like.

Phosphates include trialkyl phosphates, e.g. tri-n-butyl phosphate; triaryl phosphates, e.g. triphenyl phosphate, cresyl diphenyl phosphate, tricresyl phosphate, mixed alkyl-aryl phosphates, e.g. 2-ethylhexyl diphenyl phosphate, and the like.

Sulfonamides include o- and p- toluenesulfonamides, N-ethyl-p-toluene-sulfonamide and the like.
Nitrogen-containing aromatics include nitrobiphenyl and similar compounds.

Ester-amides include 2-ethylhexanoic acid diester of N,N-bis(2-hydroxyethyl)-2-ethylhexanamide and similar compounds.

Nitrile plasticizers include primary nitriles having 8-20 carbon atoms, e.g. a mixture of oleic, linoleic and 20 carbon cyclic nitriles.

Chlorinated esters include methyl pentachlorostearate and similar compounds.

Polymeric include low molecular weight polyesters and oligoesters, e.g. oligoesters having a molecular weight of 1,500-5,000 prepared from acid-alcohol mixtures.

The carrier for the mixture of plasticizer and acrylate ester copolymer may be any solid carrier and may be either inert or may be an active ingredient used in the powder coating formulation. Thus carriers such as silica aerogels, anhydrous colloidal silica, fillers, pigments and particulate resinous material may be used. The essential characteristic of the carrier is that it must have a relatively large surface area, i.e., from about 50 to about 500 square meters per gram, in order that a sufficient amount of the liquid copolymeric acrylate can be carried thereon and it should have an average particle size of from about 0.01 to about 500 microns. In most instances, the copolymeric acrylates will be adsorbed onto the surface of the carrier, e.g., silica, fillers, pigments. In other instances, the copolymeric acrylate will be incorporated into or on a different polymer or resin. For example, the copolymeric acrylate may be dry blended on powdered polyvinyl chloride in order to produce a finely divided material for use in powder coatings. Additionally, the copolymeric acrylate may be blended into the polyvinyl chloride and the subsequent composition chopped or diced and ground in order to form a material for powder coating formulations.
Generally, the carrier and the liquid mixture of plasticizer and copolymeric acrylate are mixed in order to provide a weight ratio of acrylate ester to carrier of from about 1:1 to about 0.025:1.

Normally, the processing aid composition of the present invention is added to the powder coating formulation in a concentration of from about 0.1 to about 5 percent on a weight basis of active ingredient. Preferably, the concentration is from about 0.5 to about 2 percent.

In general, the amount of processing aid composition which is added to the powder coating composition will be dependent upon the particular acrylate copolymer which is used, the carrier which is used and the powder coating formulation into which it is incorporated.

The processing aid composition of the present invention may be prepared, as stated above, by either solvent or emulsion spray drying techniques. For example, the acrylate copolymeric ester may be dissolved in an organic solvent such as methylene chloride and the carrier added to the solvent solution to give a desired concentration, for example, a 1:1 ratio of copolymeric acrylate ester to carrier. Additional solvent may then be added to reduce the viscosity to a workable level and the material is then spray dried in a dryer such as described in the Journal of Paint Technology, Volume 4, August 1972, pages 56-57.

Alternatively, the processing aid composition may be spray dried from a water emulsion of the copolymeric acrylate ester. Other means for drying, i.e., eliminating solvent or water, include evaporation, drum drying and other conventional means known in the art.

Preferably the processing aid which is coated on the carrier is prepared as an emulsifiable concentrate containing from about 30 to about 70 parts by weight acrylate copolymer
and, correspondingly, from about 70 to about 30 parts by weight plasticizer. Particularly useful is an emulsifiable concentrate of about 50 to about 60 parts acrylate and from about 60 to about 40 parts benzene alkylate.

Any conventional emulsifier is suitable for preparing the emulsifiable concentrate, i.e. any conventional nonionic, cationic or anionic emulsifier. Preferably, nonionic emulsifiers are used, i.e. ethylene oxide condensates, since the resulting emulsion has better clarity than an emulsion prepared with cationic or anionic emulsifiers.

Among the powder coating polymers and resins to which the processing aid of the invention may be added are thermosetting and thermoplastic polymers and resins, such as epoxies, polyesters, acrylic polymers, anhydride and acrylamide polymers, urethane polymers, chlorinated polyether polymers, silicone polymers; polyphenylene sulfide polymers, vinyl halide polymers, aliphatic and aromatic olefin polymers, acetal and vinyl acetal polymers, cellulosics, carbonate polymers, phenolics, polyblends of the above, rubbers and the like.

Exemplary polymers which are used in powder coatings include nylons, polyfluoroethylenes, polyurethanes, thermosetting epoxies, thermosetting polyesters, thermosetting acrylics, polyethylene, polypropylene, polyvinyl chloride, polyvinylacetel, polystyrene, cellulose acetate butyrate, thermoplastic acrylics and thermoplastic polyesters, polycarbonates, phenol-aldehyde resins, poly(cyclohexylene dimethylene terephthalate/isophthalate), ethylene/vinyl chloride copolymers, ethylene/vinyl acetate copolymers, ethylene/vinyl chloride/acrylamide terpolymers, polyblends, e.g. phenolic-epoxy polymer blends and olefin-vinyl halide polymer blends, rubbers such as diolefin-styrene, diolefin-acrylonitrile, diolefin-styrene-acrylonitrile rubbers and the like.

The processing aid of the present invention is most
suitably incorporated, at the present time, into powder coatings prepared from phenolics, epoxies, polyesters and acrylics.

The invention will be more readily understood by reference to the following examples. There are, of course, many other forms of this invention which will become obvious to one skilled in the art, once the invention has been fully disclosed, and it will be accordingly recognized that these examples are given for the purposes of illustration only and are not to be construed as limiting the scope of this invention in any way.

Example 1
Preparation of Processing Aid Composition
To 40 g. of methylene chloride there are added 20 g. of a copolymer of ethyl acrylate/2-ethylhexyl acrylate having a molecular weight of 2,100. In the solution thus formed there are dispersed 4 g. of silica aerogel. The mixture is spread on aluminum foil to evaporate some of the methylene chloride. The sticky residue is then dried in a 60°C oven. The dried powder is too tacky for use in powder coating formulations.

Example 2
Preparation of Processing Aid Composition
To 50 g. of acetone there are added 20 g. of ethyl acrylate/2-ethylhexyl acrylate having a molecular weight of 3,200. To the solution are added 10 g. of anhydrous colloidal silica. The slurry is poured on aluminum foil and dried for 2 days. The dried material is too tacky for use as a powder coating processing aid.

Example 3
Preparation of Processing Aid Composition
An amount of 10 g. of an ethyl acrylate/2-ethylhexyl acrylate copolymer having a molecular weight of 3,800 is mixed with 180 g. of methylene chloride, and 10 g. of anhydrous colloidal silica are added. Additional methylene chloride is
added to reduce the viscosity to 12 seconds on a No. 4 Ford cup. The ratio of copolymer to carrier is 1:1. The material is then spray dried on a cycle of 1/4 second on and 7 seconds off in a laboratory spray dryer. The finished material is of a fine particle size and is a free-flowing powder.

Example 4

Preparation of Emulsifiable Concentrate of Processing Aid Composition

Into a 250 ml. beaker there are weighed 10 g. of a mixture (60/40) of ethyl acrylate/2-ethylhexyl acrylate copolymer, dodecyl benzene and emulsifier (50/45/5 parts by weight). One hundred ml. water are added. 10 g. of silica aerogel are added slowly with vigorous stirring and an additional 100 ml. of water are added to reduce viscosity. The mixture is poured into an enamel pan and dried at 55°C. in a vacuum oven (20 in. vacuum). After drying the material was seived through a 100 mesh screen. The product contains 25 percent (weight basis) of acrylate copolymer and 22.5 percent alkylate.

Example 5

Preparation of Powder Coating Composition

An epoxy powder coating composition is prepared as follows:

An amount of 88.6 grams of epoxy resin¹, 50 g. filler² and 5.5 grams curing agent³ are dry blended in a ribbon blender. The dry blended material is then melt mixed through an extruder and the extruded material is granulated and then ground in a hammer mill. The ground material is seived through 200 mesh and is then ready for spraying.

¹ Dow Epoxy DER663U (Epoxy equivalent 730-840)
² Titanium dioxide
³ Dicyandiamide, 2.3 gms; Accelerated Dicyandiamide, 2.2 gms., Dow
Example 6
Preparation of Powder Coating Composition Containing Liquid (i.e. Masterbatched) Processing Aid

Example 5 is repeated but 12 g. of a masterbatch of 5% of ethyl acrylate/2-ethylhexyl acrylate (60/40) copolymer in epoxy resin is added to the initial mixture. The concentration of added acrylate copolymer is 0.6% based on the powder coating resin.

Examples 7-12
Powder Coating Compositions Containing Powdered Processing Aid

Example 5 is repeated, but 100 g. of epoxy resin are initially added and varying percentages of a processing aid prepared by spray drying a mixture of ethyl acrylate/2-ethylhexyl acrylate (60/40) and silica from methylene chloride are added to the epoxy resin.

Example 7: 0.4% processing aid (0.2% acrylate copolymer).
Example 8: 0.8% processing aid (0.4% acrylate copolymer).
Example 9: 1.2% processing aid (0.6% acrylate copolymer).
Example 10: 1.6% processing aid (0.8% acrylate copolymer).
Example 11: 2.0% processing aid (1.0% acrylate copolymer).
Example 12: 4.0% processing aid (2.0% acrylate copolymer).

Example 13
Comparative Evaluation of Liquid Processing Aid of Example 6 with Powdered Processing Aid of Examples 7-12

The compositions of Examples 6-12 are evaluated on the Gardner Multi-Angle Glossmeter as follows:
Sample steel panels are powder coated with the formulations of Examples 6-12 and cured. The sample panels are placed in the sample apertures of the glossmeter and gloss reflectance readings on a scale of 0 to 100 are taken at 20° and 60° inclination via a rotating standardizing potentiometer and microammeter.

The 20° gloss test is in accordance with ASTM D523-53T and Method 6104 of Federal Test Method Standard No. 141. These test methods are used for the gloss evaluation of plastic surfaces.

The 60° gloss test is in accordance with ASTM D523-53T and Methods 6101 and 6102 of Federal Test Method Standard No. 141. These test methods are used for the gloss evaluation of plastic surfaces.

The glossmeter consists essentially of five prefocused incandescent light sources employing achromatic lenses, five photocells, a five position switch for choosing the desired gloss scale, five receptor apertures, a standardizing potentiometer and a readout microammeter.

In accordance with the above procedure, gloss readings are taken of sample metal panels which have been powder coated with the formulations of Examples 6-12. The results are tabulated in the table, below:

<table>
<thead>
<tr>
<th>Formulation</th>
<th>%Additive</th>
<th>%Acrylate Copolymer</th>
<th>20°</th>
<th>60°</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ex. 6</td>
<td>12 (Masterbatch)</td>
<td>0.6</td>
<td>61</td>
<td>91</td>
</tr>
<tr>
<td>Ex. 7</td>
<td>0.4 Powder</td>
<td>0.2</td>
<td>52.6</td>
<td>83</td>
</tr>
<tr>
<td>Ex. 8</td>
<td>0.8 &quot;</td>
<td>0.4</td>
<td>84</td>
<td>98.3</td>
</tr>
<tr>
<td>Ex. 9</td>
<td>1.2 &quot;</td>
<td>0.6</td>
<td>84</td>
<td>97.3</td>
</tr>
<tr>
<td>Ex. 10</td>
<td>1.6 &quot;</td>
<td>0.8</td>
<td>83.3</td>
<td>94</td>
</tr>
<tr>
<td>Ex. 11</td>
<td>2.0 &quot;</td>
<td>1.0</td>
<td>86</td>
<td>94.3</td>
</tr>
<tr>
<td>Ex. 12</td>
<td>4.0 &quot;</td>
<td>2.0</td>
<td>87</td>
<td>94.3</td>
</tr>
</tbody>
</table>

*average of three readings
Note that, at both the 20° and 60° readings, the gloss of the formulation containing only 0.2 percent acrylate copolymer (Ex. 7) in powdered form is nearly the same as Example 6 which contains 0.6 percent acrylate copolymer in liquid form. Further, the gloss of the formulation containing 0.4 percent acrylate copolymer in powdered form (Ex. 8) is substantially higher than the gloss of the formulation of Ex. 6 which contains 0.6 percent liquid acrylate copolymer.

It is apparent that the powdered processing aid of the invention is substantially more efficient than the same processing aid in liquid form. The higher gloss readings indicate better leveling and coating of the powder coating composition.

That feature of the present invention wherein a plasticizer is also utilized in the processing aid (as prepared in Example 4) is described more fully in Examples 14-25, below: Examples 16, 17 and 18 contain a powdered processing aid as prepared in Example 4. The powdered processing aid of Example 19 contains only acrylate copolymer, while the processing aid of Example 14 is a liquid masterbatched acrylate copolymer.

Example 14
Preparation of Powder Coating Composition Containing Liquid (i.e. Masterbatched) Processing Aid

An epoxy powder coating composition is prepared as follows:

An amount of 177.2 g. epoxy resin, 100 g. filler, 24 g. of a masterbatch of 5% of ethyl acrylate/2-ethylhexyl acrylate (60/40) copolymer in epoxy resin and 9 g. curing agent are dry blended and prepared as in Ex. 6.

The powder is sprayed on steel panels and cured for 10 min. at 180°C.

1 Dow Epoxy, DER 663U
2 TiO₂
3 Dicyandiamide
Examples 15-19
Preparation of Powder Coating Compositions Containing Powdered Processing Aid

Example 14 is repeated, but 200 g. epoxy resin are initially added. Varying amounts of powdered processing aid are added in lieu of the masterbatch liquid processing aid of Example 6. The powdered processing aid is obtained by spray drying from an aqueous emulsion.

Example 20
Comparative Evaluation of Powder Coating Compositions Containing Liquid Processing Aid of Ex. 14 and Powdered Processing Aid of Exs. 15-19

The compositions of Exs. 14-19 are spray dried on steel panels, cured and evaluated on the Gardner Multi-Angle Glossmeter. The results are tabulated below:

<table>
<thead>
<tr>
<th>Formulation</th>
<th>PHR² Additive</th>
<th>% Acrylate Copolymer</th>
<th>Angle ²⁰°</th>
<th>Angle ⁶⁰°</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ex.14</td>
<td>12</td>
<td>0.4</td>
<td>57.7</td>
<td>89.3</td>
</tr>
<tr>
<td>Ex.15</td>
<td>1.2</td>
<td>0.4</td>
<td>66</td>
<td>88</td>
</tr>
<tr>
<td>Ex.16</td>
<td>2.4</td>
<td>0.2³</td>
<td>73</td>
<td>90.3</td>
</tr>
<tr>
<td>Ex.17</td>
<td>2.4</td>
<td>0.2³</td>
<td>83</td>
<td>94</td>
</tr>
<tr>
<td>Ex.18</td>
<td>1.8</td>
<td>0.2³</td>
<td>83.7</td>
<td>92.6</td>
</tr>
<tr>
<td>Ex.19</td>
<td>1.2</td>
<td>0.4</td>
<td>68.3</td>
<td>86.3</td>
</tr>
</tbody>
</table>

¹ Average of three readings
² Parts per hundred resin
³ Additionally contains 0.2% dodecylbenzene

It is seen that the addition of dodecylbenzene in the powdered processing aid significantly raises the efficiency of the processing aid. Similar results are obtained with other alkyl benzenes having alkyl chain lengths of 2 to 20 carbon atoms.
Examples 21-25

Preparation of Powder Coating Compositions

Examples 21-25 further illustrate the utility of a powdered processing aid which combines acrylate copolymer and an alkyl benzene. The example formulations are as follows:

<table>
<thead>
<tr>
<th>Formulation</th>
<th>Example (PHR)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>21</td>
</tr>
<tr>
<td>Epoxy resin⁴</td>
<td>200</td>
</tr>
<tr>
<td>Curing agent⁵</td>
<td>9</td>
</tr>
<tr>
<td>Filler</td>
<td>100</td>
</tr>
<tr>
<td>Masterbatch aid⁶</td>
<td>---</td>
</tr>
<tr>
<td>Powdered aid⁶</td>
<td>---</td>
</tr>
<tr>
<td>Dodecylbenzene</td>
<td>---</td>
</tr>
<tr>
<td>Powdered aid⁶</td>
<td>---</td>
</tr>
</tbody>
</table>

1 Dow, DER 663U
2 Accelerated dicyandiamide
3 Titanium dioxide
4 Five percent masterbatch of ethyl acrylate/2-ethylhexyl acrylate (60/40) copolymer in epoxy resin.
5 Spray dried on silica (5 g.) from water emulsion of 10 g. of 50/45/5 blend of ethyl acrylate/2-ethylhexyl acrylate (60/40) copolymer, dodecylbenzene and emulsifier.
6 Spray dried on silica--ethyl acrylate/2-ethylhexyl acrylate copolymer.

Example 26

Comparative Evaluation of Processing Aids of Examples 21-25

The compositions of Exs. 21-25 are electrostatically sprayed on steel panels and cured. The panels are evaluated on a Gardner Multi-Angle Glossmeter. The results are tabulated below:
<table>
<thead>
<tr>
<th>Formulation</th>
<th>$20^\circ$</th>
<th>$60^\circ$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ex. 21</td>
<td>19</td>
<td>57</td>
</tr>
<tr>
<td>Ex. 22</td>
<td>67</td>
<td>92</td>
</tr>
<tr>
<td>Ex. 23</td>
<td>64</td>
<td>89</td>
</tr>
<tr>
<td>Ex. 24</td>
<td>74</td>
<td>96</td>
</tr>
<tr>
<td>Ex. 25</td>
<td>58</td>
<td>87</td>
</tr>
</tbody>
</table>

1 Average of three readings

As can be seen from the above data, the coated panel of Ex. 21 was the least glossy (Ex. 21 formulation is a control and contained no processing aid). Exs. 22 and 23 are essentially equal (Ex. 22 containing 0.2% liquid acrylate copolymer; Ex. 23 containing 0.2% acrylate copolymer spray dried on silica). Ex. 25 contains 0.5% spray dried dodecylbenzene as sole processing aid.

Example 24 contains a 50/45/5 blend (see note (5) above) containing 0.5% acrylate copolymer spray dried on silica. Substantially improved results are seen. Similar results are obtained using other plasticizers such as di-$C_7$-$C_{11}$ alkyl phthalates, butylbenzyl phthalate, di-2-ethylhexyl phthalate, 2-ethylhexyl diphenyl phosphate, tri-$n$-butyl phosphate, triphenyl phosphate and tricresyl phosphate.

The matter contained in each of the following claims is to be read as part of the general description of the present invention.
CLAIMS
The claims defining the invention are as follows:

1. Process for preparing a powdered processing aid suitable for use in powder coating resins comprising (A) dissolving in a solvent therefor, a liquid acrylate copolymer consisting of (1) from about 5 to about 95% by weight of an alkylacrylate ester wherein the alkyl constituent contains from about 1 to about 12 carbon atoms and (2) from about 95 to about 5% by weight of a dissimilar alkylacrylate ester wherein the alkyl constituent contains from about 3 to about 24 carbon atoms, said copolymer having an average molecular weight of from about 2,000 to about 10,000, (B) adding a particulate solid carrier for said acrylate copolymer to the acrylate copolymer/solvent solution and (C) drying the mixture to thereby form a free flowing powder consisting of a solventless film of acrylate copolymer on said carrier particles.

2. Process of claim 1 which comprises (A) preparing a water emulsion of said liquid acrylate copolymer, (B) adding said particulate solid carrier for said acrylate copolymer to said emulsion and (C) drying the mixture to thereby from a free flowing powder consisting of a water-free film of acrylate copolymer on said carrier particles.

3. Process of claim 1 or 2 which comprises admixing (A) a concentrate comprising said liquid acrylate copolymer, a plasticizer and an emulsifier, (B) adding said particulate solid carrier for said acrylate copolymer to said mixture and (C) drying the mixture to thereby form a free flowing powder consisting of a solventless film of acrylate copolymer on said carrier particles.
4. Process of claim 3 wherein said concentrate contains from about 30 to about 70 parts by weight of said liquid acrylate copolymer and, correspondingly, from about 70 to about 30 parts by weight of plasticizer.

5. Process of claim 3 or 4 wherein said plasticizer is an alkylated benzene, an alkylated aromatic petroleum fraction, a fatty acid ester, an aromatic or aliphatic acid diester, a triester of aliphatic acid, a phosphate, a sulfonamide, a nitrogen-containing aromatic, an ester-amide, a nitrile, a chlorinated ester or a polymeric.

6. Process of any one of claims 1 to 5 wherein said liquid acrylate copolymer is any one of such compositions as specifically described herein.

7. Process of claim 1, substantially as described in the Examples herein.

8. Powdered processing aid obtained by the process of any one of claims 1 to 7.

9. Composition comprising a powder coating resin and a powdered processing aid which comprises a particulate solid carrier having a solventless film of a liquid acrylate copolymer consisting of (1) from about 5 to about 95% by weight of an alkylacrylate ester wherein the alkyl constituent contains from about 1 to about 12 carbon atoms and (2) from about 95 to about 5% by weight of a dissimilar alkylacrylate ester wherein the alkyl constituent contains from about 3 to about 24 carbon atoms, said copolymer having an average molecular weight of from about 2,000 to about
10,000 on said carrier particles.

10. Composition of claim 9 wherein the powdered processing aid is as claimed in claim 8.

11. An emulsifiable concentrate useful for preparing a powdered processing aid for powder coating resins comprising a liquid acrylate copolymer, a plasticizer and an emulsifier, said liquid acrylate copolymer consisting of (1) from about 5 to about 95% by weight of an alkylacrylate ester wherein the alkyl constituent contains from about 1 to about 12 carbon atoms and (2) from about 95 to about 5% by weight of a dissimilar alkylacrylate ester wherein the alkyl constituent contains from about 3 to about 24 carbon atoms, said copolymer having an average molecular weight of from about 2,000 to about 10,000.

12. Emulsifiable concentrate of claim 11 wherein said liquid acrylate copolymer is about 30 to about 70 parts by weight and, correspondingly, said plasticizer is about 30 to 70 parts by weight.

13. Emulsifiable concentrate of claim 11 or 12 wherein said plasticizer is an alkylated benzene, an alkylated aromatic petroleum fraction, a fatty acid ester, an aromatic or aliphatic acid diester, a triester of aliphatic acid, a phosphate, a sulfonamide, a nitrogen-containing aromatic, an esteramide, a nitrile, a chlorinated ester or a polymeric.

14. Emulsifiable concentrate of any one of claims 11 to 13 wherein said liquid acrylate copolymer is any one
of such compositions as specifically described herein.

15. Emulsifiable concentrate of claim 11, substantially as described in the Examples herein.

DATED this 19th day of February, A.D. 1975

MONSANTO COMPANY,
By its Patent Attorney,

[Signature]

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