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E. D. SEITER

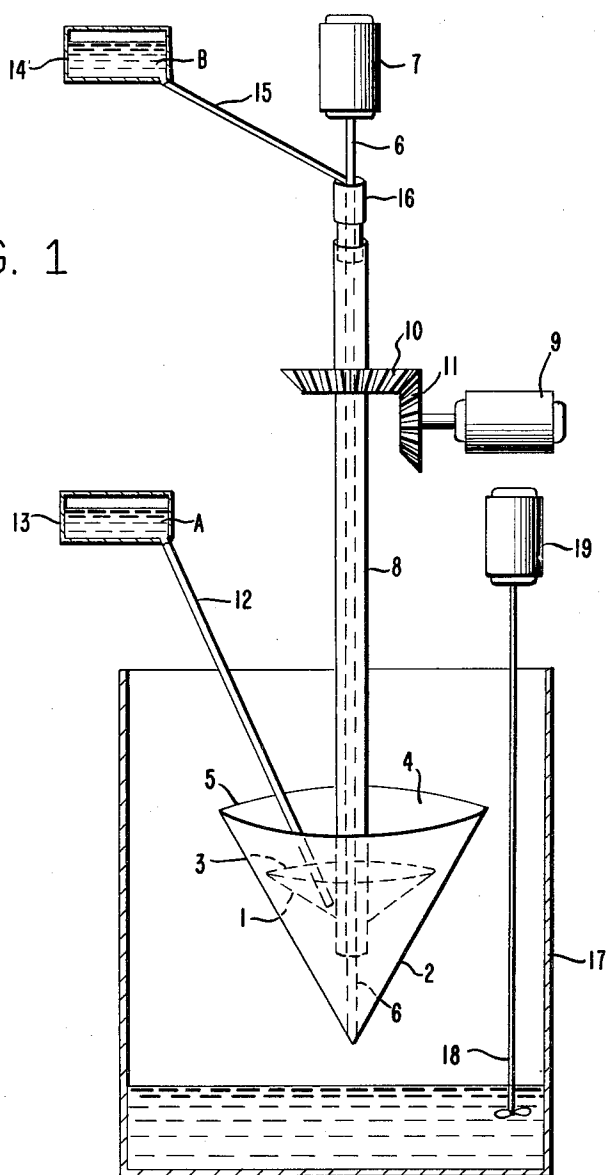
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APPARATUS FOR MIXING FLOWABLE MATERIALS

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2 Sheets-Sheet 1

FIG. 1



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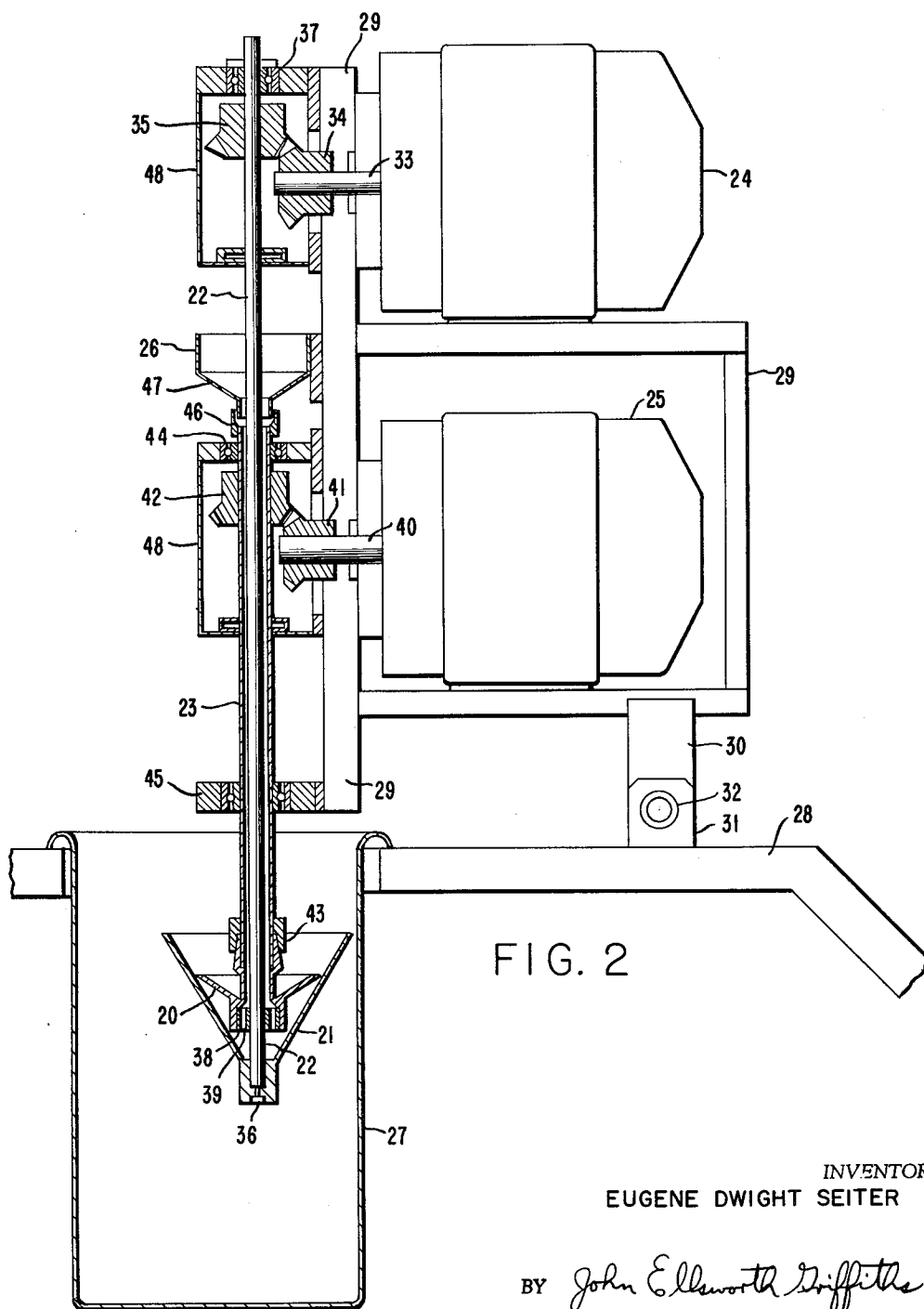
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This invention relates to apparatus for mixing fluids and more particularly to a means for effecting controlled uniform blending of a plurality of flowable materials. Still more particularly, it relates to apparatus by the use of which two or more materials are brought together as thin superimposed layers.

An object of this invention is an improved apparatus for mixing materials. Another object is to provide such apparatus which can be easily and accurately controlled. Still another object is to provide such apparatus in which rapid mixing is obtained.

Another object is to provide improved apparatus for mixing two or more separate materials whereby maximum surface areas of the materials are presented for contact with each other. Yet a further object is to provide such apparatus which is economical and simple to construct and easy to maintain under continuous operating conditions. Still further objects include providing apparatus for reacting two or more starting materials without exposing the product of the reaction to an excess of the starting materials, and providing such apparatus wherein the environment for the reaction is constant. Still other objects will be apparent from the following description of the invention.

In its broader aspects, this invention is directed to forming a thin layer of a flowable material by rotating the material about an axis, which is preferably vertical, and effecting interfacial contact of the layer with another thin layer of a flowable material similarly formed. The layers may be of the same but preferably are of different materials.

"Flowable material" as used herein is meant to include liquids, e.g., solutions, dispersions and emulsions, as well as particulate finely divided solids, and combinations of these. It is essential only that the material be sufficiently fluid so that it will flow. It will be apparent that materials which can be rendered fluid by heating are within the scope of this invention since a simple application of heat to the material at the beginning of or throughout the process is an obvious expedient modification.

The thinness of the layers will depend on such variables as the materials being processed, the viscosity of the materials, the concentration including solids content of the materials, the operating temperature, the degree of product uniformity desired, the apparatus utilized, etc. Optimum operating conditions will be readily determinable by persons in the art in accordance with the teachings set forth herein. In general, the highest degree of product uniformity will be obtained by having the layers as thin as possible. The process has been found to be most effective for liquids, such as aqueous solutions.

In a specific preferred embodiment of this invention, a thin layer of an aqueous solution of a silver salt, such as silver nitrate, is formed and superpositioned onto a thin layer of an aqueous solution containing a photographic binder, such as gelatin, and an alkali metal halide, such as potassium bromide. Light-sensitive photographic emulsions made according to this process have been found to exhibit excellent photographic properties.

In one aspect, this invention comprises rotating a first flowable material about an axis to form a first thin layer by centrifugal action, rotating a second flowable material about the same axis to form a second thin layer by cen-

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trifugal action, and bringing the first and second layers together while rotating to provide maximum surface contact between the layers.

In an apparatus according to this invention, separate liquids are introduced simultaneously into separate zones in which the liquids are rotated about a common axis to form separate thin rotating layers under the action of centrifugal force. The rotating layers are continuously rotated in a spinning manner to fling them radially outward from the axis in such a manner as to bring them into essentially coplanar intersurface contact with one another in another zone, through and from which the combined material centrifugally passes so that the combined material does not remain at the location of first contact, and the combined material is continuously received from the last mentioned zone from where it may be collected, stored, or utilized as by further processing or introduction into a product or combination with other materials.

The apparatus of this invention is particularly advantageous in the blending of liquids of different viscosity wherein the rotative motion given to the more viscous liquid is at a greater speed than that of the less viscous liquid.

The invention will be more clearly understood by the following description of novel mixing apparatus according to this invention, which description will now be given with reference to the accompanying drawings, wherein:

FIG. 1 is a schematic elevation view, partly perspective, illustrating one type of apparatus of this invention; and

FIG. 2 is an elevation view, partly in section, showing further details of the apparatus of FIG. 1.

Referring now to the drawings, the principal elements of the apparatus of this invention include a hollow inner conical vessel 1 positioned with apex lowermost and a hollow outer conical vessel 2 similarly positioned. Vessels 1 and 2 are in nesting relationship with each other, i.e., their concave receptacle portions both face upwardly and their apexes both point downwardly when the axis of rotation is vertical, as in the illustrated embodiment. The inner vessel 1 is positioned so that its upper edge or lip 3 is close to but does not touch the inner wall surface 4 of the outer vessel 2. As illustrated, the lip 3 in vertical displacement is lower than the corresponding edge or lip 5 of the outer vessel.

Vessels 1 and 2 are independently mounted for independent rotation about a common axis. Thus, outer vessel 2 is conveniently secured at its apex to vertical shaft 6 which is driven by any suitable means, e.g., motor 7. Inner vessel 1 is secured to the lower part of hollow vertical shaft 8 which is driven by any suitable means, e.g., motor 9 through cooperating bevel gears 10 and 11.

The inside diameter of shaft 8 is larger than the outside diameter of shaft 6 in order to provide a passage therebetween for flowing material, as hereinafter described. The shafts are maintained non-contacting with each other by suitable means, one example of which is described below and shown in detail in FIG. 2 of the drawings. Shafts 6 and 8 in their concentric relationship are supported by any suitable means, e.g., motors or bearings which in turn may be secured to or mounted on a suitable frame, directly or through other members.

One material A is introduced into the lower portion of inner vessel 1 by tube 12 leading from a source 13. Material B from source 14 is introduced into the lower portion of outer vessel 2 by tube 15 which feeds into funnel member 16 and downwardly through the passage between shafts 6 and 8. The feed rates of materials A and B may be conveniently controlled by any suitable means (not shown), such as pressure, constant or variable head, valves in tubes 12 and 15 or elsewhere, or other metering devices. Alternatively, the funnel mem-

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ber 16 may be adapted to serve as a reservoir which feeds material at a desired rate. If materials A and B are the same, it is obvious that feed lines from a common source will be satisfactory. The sources and other feeding elements useful within this invention are not critical and various modifications will readily occur to persons in the art.

As explained below, operation of this apparatus causes the combined materials to flow outwardly over the lip 5 of the outer vessel 2. Provision can be made for catching the materials, as by collecting reservoir 17, wherein the combined materials may be stirred or otherwise agitated if desired by propeller mixer 18 driven by motor 19. Reservoir 17 may be of any convenient size and configuration, and may be provided with obvious modifications without departing from the present invention. For example, the reservoir may be jacketed and heated or cooled by any suitable means. Suitable outlet may be provided in the lower part of the reservoir to withdraw the combined product either batchwise, as by a faucet or similar tap, or continuously, as by a pipe or other channeling means leading to other operations, further processing, storage, etc.

Additional details of one embodiment of the apparatus of this invention are illustrated in FIG. 2 of the drawings. The principal elements of the apparatus shown in FIG. 2 correspond to those of FIG. 1 and include inner vessel 20, outer vessel 21, shafts 22 and 23 with associated driving motors 24 and 25 respectively, funnel member 26, and collecting reservoir 27.

Referring to FIG. 2, the illustrated apparatus is shown conveniently supported by pedestal member 28 upon which is mounted a frame 29 carrying motors 24 and 25. The frame 29 may be secured to pedestal member 28 by brackets 30 and 31 which are connected to pivot rod 32. As can be seen, the frame and mixing apparatus can be readily swung back on its pivotal mount to permit access to the collecting reservoir 27 which is shown inserted in an aperture in pedestal member 28.

The drive shaft 33 of motor 24 interfits with miter gear 34 intermeshing cooperatively with miter gear 35 secured to vertical shaft 22. Shaft 22, to the lower end of which the outer vessel 21 is fixedly attached by any suitable means such as screw 36, is rotatably suspended near its upper end by a bearing 37 mounted on the frame 29. Shaft 22 is additionally positioned near its lower end by bearing 38 having vertical holes 39 to permit the downward flow of material B from funnel member 26 into the outer vessel 21. The number of holes is not critical. Bearing 38 serves to maintain shafts 22 and 23 in concentric non-contacting relationship permitting independent rotation.

The drive shaft 40 of motor 25 interfits with miter gear 41 intermeshing cooperatively with miter gear 42 secured to vertical shaft 23. Shaft 23, to the lower end of which inner vessel 20 is fastened by any suitable means such as a tapered sleeve clamp 43, is rotatably suspended near its upper end by a bearing 44 mounted on the frame 29. Shaft 23 is additionally positioned by similarly mounted bearing 45.

Shaft 23 may be conveniently fitted with a tapered bushing 46 to accommodate a lower extension 47 of funnel member 26.

The bevel gears or miter gears referred to above are preferably enclosed in suitable separate housings 48. At the lower end of each housing a suitable seal, such as a split felt seal, is inserted to prevent lubricating greases or oils from the gears from accidentally contaminating the materials A and B.

In a preferred method of operation of the illustrated equipment, with particular reference to FIG. 1, inner vessel 1 and outer vessel 2 are each rotated by their drive systems in the same direction. Material A is fed directly from source 13 into inner vessel 1 near its apex. Material B is fed from source 14 by way of funnel member 16 and the interior of hollow shaft 8 the outer ves-

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sel 2 near its apex. The rotation of outer vessel 2 spreads material B by the action of centrifugal force into a thin film or layer which climbs the inside wall 4 of the vessel 2 to its lip 5, over which the material radially passes. The rotation of inner vessel 1 spreads material A similarly into a thin film or layer which climbs the inside wall of vessel 1 to its lip 3, over which the material moves. The film or layer of material A being discharged from the inner vessel 1 effects contact with the film or layer of material B in a circular zone on the upper part of the wall 4 of the outer vessel 2. The combined materials then pass over the lip 5 of the outer vessel 2 into the collecting reservoir 17.

It can be seen that the described operation brings together the separate materials while each is in the form of a thin layer. The layers are not thrown at each other in a helter-skelter fashion, but rather are effectively moved smoothly into interfacial contact. Increasing the speed of rotation of vessels 1 and 2 serves to reduce the thickness of the layers of materials, thus providing maximum surface exposure of each material to the other.

The dimensions of the various parts of the apparatus, and the materials of construction, are not particularly critical, and may readily be selected by persons in the art to meet specific requirements.

In a practical apparatus of the type illustrated in FIGS. 1 and 2, a $\frac{1}{4}$ inch stainless steel shaft about 22 $\frac{1}{2}$ inches long may be used for shaft 6. A hollow stainless steel shaft $\frac{1}{2}$ inch in inside diameter and 13 inches long may be used as shaft 8. A stainless steel cone having an apex angle of 60° and 4 inches in diameter at its base may be used for vessel 2. A smaller cone with an apex angle of 120° and 2 $\frac{3}{4}$ inches base diameter may be used as vessel 1. Conventional variable speed motors may be used to drive the shafts. Long-wearing, corrosion resistant materials, such as tetrafluoroethylene resin, are preferred for the bearings. Either or both vessels can be heated or cooled as desired in any suitable manner. It is of course desirable to utilize for all parts construction materials inert to the particular fluids being processed.

The operation of the apparatus of this invention will be further explained but is not intended to be limited by the following examples:

Example I

Utilizing apparatus of the type shown in FIG. 1 having the dimensions cited above, material A consisting of 167 ml. of a 3-normal aqueous solution of silver nitrate and 333 ml. of distilled water was placed in a tank source leading to the apex of the smaller cone. Material B consisting of 350 ml. of a 3-normal aqueous solution of potassium bromide, 13.3 grams of inert gelatin and 150 ml. of distilled water was placed in a source leading through the passage in the hollow shaft to the apex of the larger cone. Two hundred ml. of distilled water was placed in the collecting reservoir. The smaller cone was rotated at about 350 r.p.m. and the larger cone was rotated in the same direction at about 900 r.p.m. Materials A and B were fed simultaneously into the respective cones until simultaneously completely used in a period of about one minute. Agitation of the material in the collecting reservoir was continued by a propeller mixer. The material collected in the collecting reservoir was a light-sensitive emulsion which was then conventionally ripened, washed, and coated onto a cellulose acetate film base and dried.

Examination of the coated emulsion under an electron microscope showed that the obtained emulsion was exceptionally uniform. It is believed that this exceptional uniformity results because the entire precipitation reaction which takes place upon bringing the starting materials together occurs in a small mixing zone in a ring at the confluence of the fluid layers. The environment for each nascent silver bromide grain is uniform because the con-

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centration of reactants is maintained essentially constant in the mixing zone throughout the entire operation. This constant concentration of reactants contrasts with the prior art method of preparation of photographic emulsions by propeller type mixing wherein the concentration of the reactants constantly changes as the precipitation reaction proceeds. The emulsion exhibited excellent photographic properties when exposed and processed by conventional methods.

Example II

Utilizing the apparatus of Example I, material A consisting of 500 ml. of 0.0105 molar aqueous sulfuric acid was placed in a tank source leading to the apex of the smaller cone. Material B consisting of 500 ml. of 0.0108 molar aqueous barium hydroxide was placed in a source leading through the passage in the hollow shaft to the apex of the larger cone. The smaller cone and the larger cone were rotated in the same direction at about 400 and 900 r.p.m. respectively. Materials A and B were fed simultaneously into the respective cones until simultaneously completely used in a period of about one minute. A precipitate of barium sulfate in water was collected in the collecting reservoir and examined under an electron microscope.

For comparative purposes, 500 ml. of each of materials A and B were additionally prepared and poured simultaneously into a container in which a conventional propeller type mixer was positioned. This mixer was operated at about 700 r.p.m. for one minute. This precipitate of barium sulfate are also examined under an electron microscope.

It was observed that the largest particles of barium sulfate in the propeller-mixed precipitate measured 28 microns in diameter while the largest particles of barium sulfate in the precipitate prepared according to this invention measured only 12 microns. Smaller particle size is evidence of more complete and uniform blending of the starting materials and is a desideratum in many chemical reactions.

Example III

In order to demonstrate the high speed mixing action obtainable with the apparatus of this invention, an acid-base neutralization was carried out using 500 ml. of 0.70-normal aqueous sulfuric acid as material A and 500 ml. of 1-normal aqueous sodium hydroxide as material B. The latter contained a small amount of phenolphthalein as a color indicator. As the materials were fed into their respective rotary vessels, the material throughout the entire mixing zone on the inner upper wall of the outer cone was observed to be a constant red color at all times during the operation, thus indicating the practically instantaneous and complete mixing of A and B.

For comparison purposes, this neutralization was also carried out using identical starting materials A and B in the following manner: The acid was placed in a tank container in which a propeller type mixer was rotating at about 500 r.p.m. The base containing the indicator was added by pouring at a uniform rate into the acid during a period of one minute. It was observed that a uniform red color was obtained only after 43 seconds of mixing had taken place, clearly indicating that the products of the neutralization reaction were exposed to the acid in the container for 43 seconds.

Example IV

This example will illustrate the carrying out of an extremely rapid (split second) series of consecutive reactions, as well as the versatility and other advantages of this invention. Utilizing the apparatus of Example I, material A consisting of 167 ml. of 3-normal aqueous silver nitrate and 333 ml. of distilled water was placed in a tank source leading to the apex of the smaller cone. Material B consisting of 30 ml. of 0.5-normal aqueous potassium iodide, 13.3 grams of inert gelatin and 470 ml. of

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distilled water was placed in a source leading through the passage in the hollow shaft to the apex of the larger cone. Material C consisting of 125 grams of solid potassium bromide and 200 ml. of distilled water was placed in the collecting reservoir. The smaller cone and the larger cone were rotated in the same direction at about 900 and 300 r.p.m. respectively. Materials A and B were fed simultaneously into the respective cones until simultaneously completely used in a period of about one minute.

On the upper portion of the inner wall of the larger cone, the materials A and B reacted to form silver iodide in a split second reaction occurring in a narrowly defined ring-shaped reaction zone. A split second later, the combined materials dropped into the collecting reservoir where silver bromide was formed in another split second reaction. The product was a gelatino silver iodobromide light-sensitive emulsion which was then conventionally ripened, washed, coated onto cellulose acetate film base and dried. The resultant photographic film and emulsion exhibited excellent sensitometric and uniform properties similar to the emulsion and film of Example I.

The process of this and the above examples is carried out on a continuous basis by providing continuous sources of starting materials and continuously removing the emulsion from the collecting reservoir.

As indicated by the preceding examples, this invention is useful in preparing mixtures of liquids or in carrying out chemical reactions between liquids. However, other materials such as finely divided solids could be substituted for one or more of the liquids.

The invention readily lends itself to various modifications. The apparatus can be easily adapted to mix more than two materials. Additional shafts, rotating centrifuging vessels, and motors may be used. One or more of the lower centrifuging vessels may be mounted and driven from below using suitably sealed apparatus. A particular embodiment of the apparatus of this invention adapted especially for carrying out the multiple step reaction process of Example IV includes a suitably mounted and driven apparatus of three vertically nested centrifuging vessels. The uppermost vessel is of course the smallest, the lowermost vessel is of course the largest, the two uppermost vessels are preferably mounted as illustrated in the accompanying drawings, and the lowermost vessel is preferably mounted and driven from underneath.

The configuration of the rotating vessels or cones is not critical within broad modifications. Cones, discs, cups, and any suitably concave saucer-shaped vessel can be used which provides an inner wall which slopes generally upwardly and outwardly. The inner vessel could be perforated thus partially dividing the feed material in that vessel into radial jets. The vessels may be provided with vanes or baffles which may be full or partial, parallel or perpendicular or askew to the rim of the outer vessel, and may comprise solid or perforated plates or rings. Any of the centrifuging vessels may be more convex or concave than any other of the vessels. It is preferred, however, that each relatively lower and outer vessel have an apex angle smaller than each next adjacent inner and higher vessel. The apex angle of each vessel is preferably less than 180°, and it is preferred that the apex angle of each cone is smaller than the next adjacent higher cone.

The nesting centrifuging vessels are rotated preferably in the same direction but may be rotated in opposite directions. Where greater turbulence with its attendant advantages is desired, the cones are preferably rotated in opposite directions. It has been noted that when the cones are rotating in opposite directions, the reaction products move along the surface of the outer cone spiraling upward and away from the unused reagents at the reaction site, thus suppressing unwanted side reactions. This advantageous feature may be useful in increasing yields of organic reactions, precipitation of pigments, etc.

The driving power for the centrifuging vessels may come from separate power sources as illustrated or may

conveniently be derived through suitable power take-off connections from a single source, using appropriate gearing where desired. The drive may be continuous or intermittent to one or more vessels and changes in the direction of rotation of one or more vessels may be effected as desired. The vessels may be rotated at the same or at different speeds, as disclosed above.

Although the axis of rotation in carrying out the process and utilizing the apparatus according to this invention is preferably vertical, it will be understood that such is not essential, particularly at high speeds of rotation, wherein the centrifugal force acting on the material exceeds the force of gravity by a relatively large amount.

Automatic controls for continuous operations can be utilized effectively in this invention. For example, starting or metering the flow of one or more of the starting materials can be readily accomplished by using an actuating signal from the effluent. This signal can be a function of the pH, the turbidity, color or other chemical or physical property of the product.

This invention is particularly advantageous in its versatility in carrying out mixing operations of the type illustrated by Example IV above. For example, the order of introduction of reactants can be changed so that the silver bromide rather than the silver iodide will be precipitated in the first stage. The gelatin, metal ions, or other ingredients can be introduced at any desired stage. In this way, by effecting control of the grain size and structure of the precipitate by controlling the thickness of the layer of reacting material by selecting a desired speed of vessel rotation, the silver iodide and the silver bromide grain structures can be separately controlled.

An advantage of the invention resides in the exceptional degree of uniformity obtained in the mixed product. Another advantage is that the materials to be mixed are passed into a narrowly defined mixing zone which contains only an extremely small amount of each of the materials at any instant. Yet another advantage is that more intimate contact between the materials takes place due to the greater exposed surface area per unit volume of the materials. This latter advantage is particularly important in view of the fact that many precipitation reactions go to completion in bulk mixing before a homogeneous mixture can be obtained. Still another advantage is that mixing according to the present invention can be carefully controlled by adjusting the speed of rotation of the centrifuging vessels, thereby controlling the thickness of the fluid films. A further advantage is that the apparatus of this invention is simply constructed and inexpensive to make and manufacture.

An additional advantage is obtained by being able to precisely delineate the mixing zone. In this manner, the actual starting point of a reaction can be precisely determined and thus the novel process and apparatus of this invention make it possible to carry out reactions which have critical reaction rates. Since the starting point of the reaction can be easily controlled, the mixer is also useful in studying reaction rates.

Yet a further advantage resides in the fact that energy is applied to the system during operation exactly where the energy is required, namely, in the spreading of the fluid into thin films just prior to bringing them into contact with one another. This obviates the need for high fluid pressures, typically required in other types of mixers where a large amount of energy is brought in by way of the fluid pressure required to introduce the fluids into the system. Yet a further advantage is that this invention permits the individual independent control of each of the fluids being mixed simply by adjustment of the speed of the centrifuging vessel on which the fluid is being distributed. Thus, the more viscous fluids can be spun at relatively higher speeds in order to obtain a layer of desired thinness to produce the mixing effect desired.

Another advantage is that the shear and turbulence of the material on each of the centrifuging vessels is adjustable simply by variation of the speed of the vessel, which fact greatly improves the versatility of the operation as compared with other types of mixing and reaction processes and apparatus.

From the above description, it will be clear that the subject of this invention is particularly useful, in addition to physically mixing materials, in chemically mixing or reacting materials. Reactions which may be cited for purposes of illustration include the neutralization of acids and bases, oxidation and reduction reactions, e.g., oxidation of such compounds as aniline with various reagents to obtain quinone, azobenzene, nitrobenzene, liquid phase nitration of hydrocarbons, halogenation, e.g., of diazonium salts, and the hydrolysis of esters and alkyl halides with water, dilute acids or bases. The apparatus of this invention may also be used to carry out a polymerization reaction, e.g., between a dibasic acid halide and a diamine.

Still other advantages and uses will readily be apparent from the above description of the invention, which is intended to be limited only as set forth in the following claims.

The invention claimed is:

1. An apparatus for admixing a plurality of flowable materials and removing the admixture by centrifugal force comprising a plurality of conical vessels facing in the same direction and disposed in nesting relationship with each other, each of said vessels mounted for rotation about a vertically disposed axis and having generally upwardly and outwardly sloping walls forming a smooth, uninterrupted surface, said vessels being of different sizes with the smaller vessel positioned completely within each progressively larger vessel so that the top part of each vessel is lower than the top part of each progressively larger vessel and the top part of said smaller vessel is positioned in close proximity to said progressively larger vessel, means for continuously feeding flowable material into each said vessel adjacent its lowermost part, driving means for said vessels, and collecting means for material issuing from the outer vessel.

2. Apparatus as set forth in claim 1 wherein said vessels are mounted for independent rotation.

3. A mixing apparatus comprising a first conical centrifuging vessel mounted for rotation about a vertically disposed axis, a second conical centrifuging vessel mounted for rotation about said axis, each of said vessels having generally upwardly and outwardly sloping walls forming a smooth, uninterrupted surface providing an upper supporting surface for a flowable material, said second vessel being smaller than said first vessel and being positioned inside said first vessel in a nesting, non-contacting relationship therewith such that the uppermost part of said second vessel is below the uppermost part of said first vessel and the top part of said second vessel is positioned in close proximity to said first vessel, the area on the inner said supporting surface of said first vessel vertically located between said uppermost parts defining a ring-shaped mixing zone.

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