

[54] MIXING PROCESS

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(Under Rule 47)

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[52] U.S. Cl. .... 259/9, 23/252 R

[51] Int. Cl. .... B01f 7/04

[58] Field of Search .... 259/2, 10, 9, 109, 259/110, 7, 8, 21, 22, 23, 24, 25, 26, 43, 44, 178, 68, 69, 1; 23/252

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[57]

ABSTRACT

A mixing process in which a liquid is simultaneously subjected to a combination of vertical displacement, horizontal recirculation, and horizontal roll. The process is especially well adapted for use with highly viscous fluids and can be practiced with surprisingly low power requirements. A substantially homogeneous liquid system can be produced and maintained in a relatively short mixing time.

16 Claims, 16 Drawing Figures

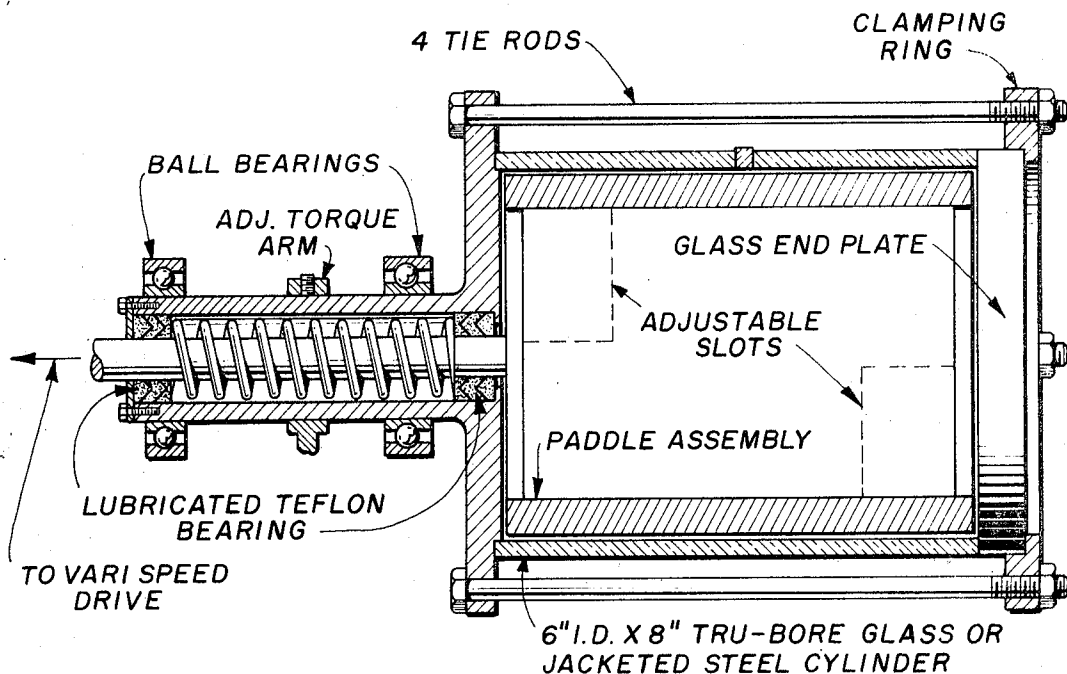
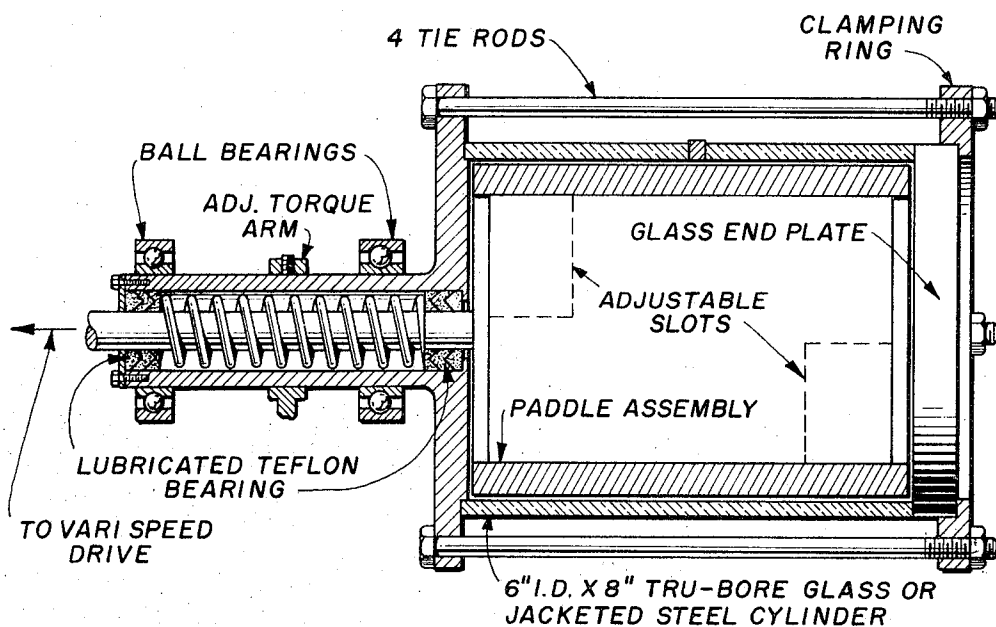


FIG. 1.



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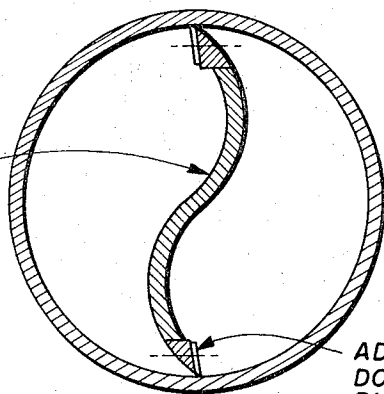


FIG. 2.

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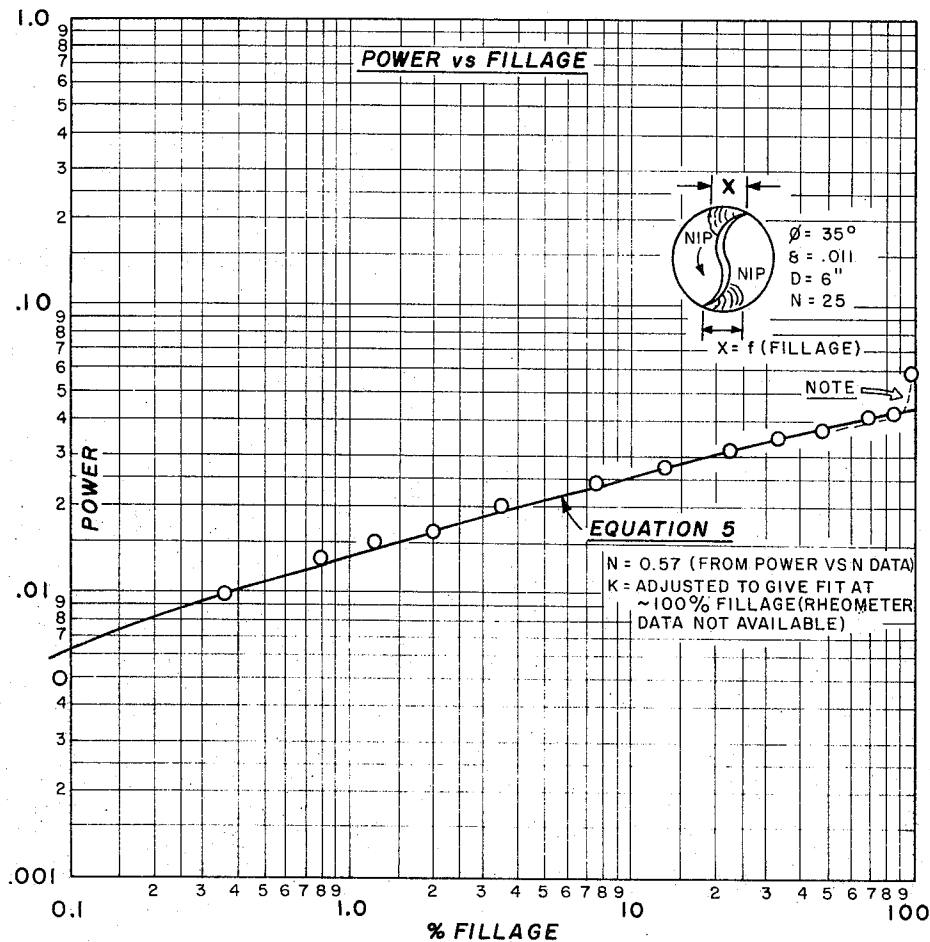


FIG. 3.

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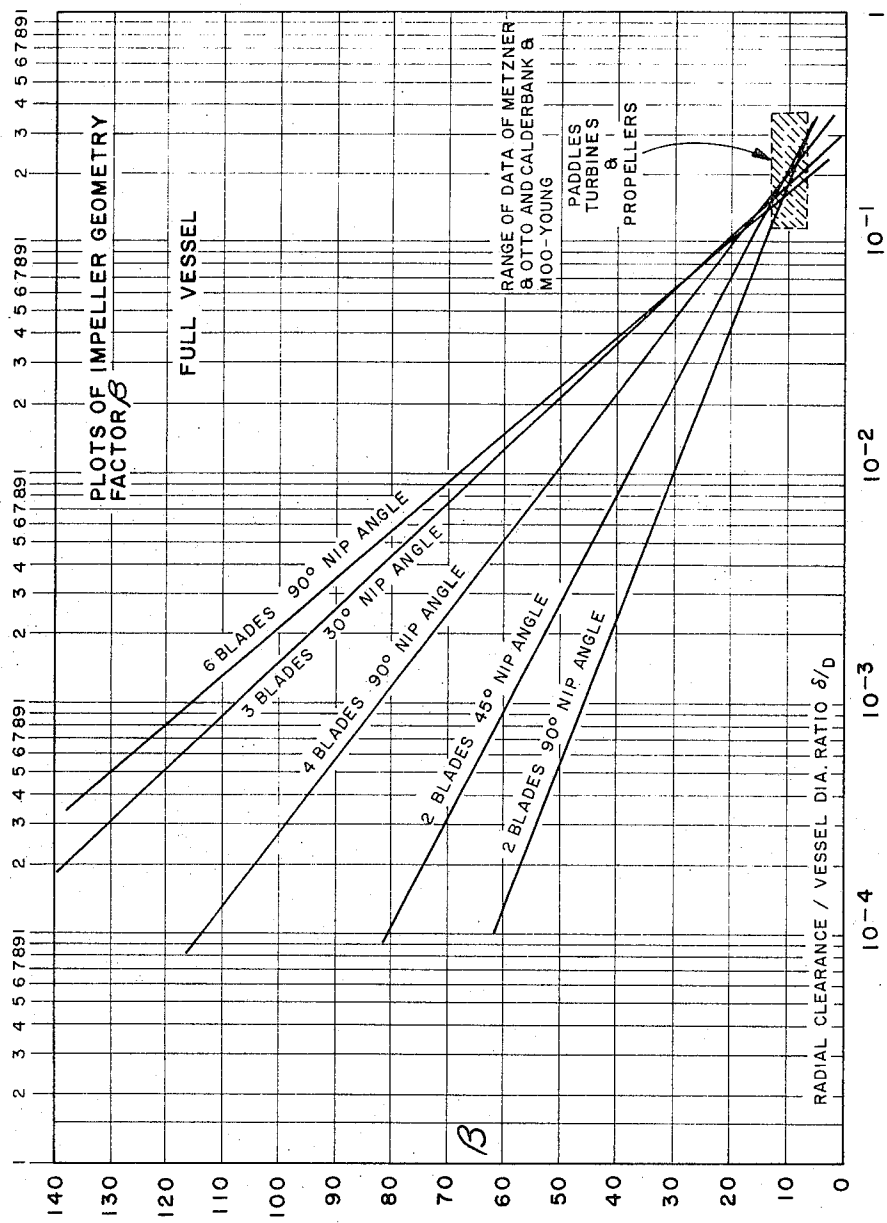


FIG. 4.

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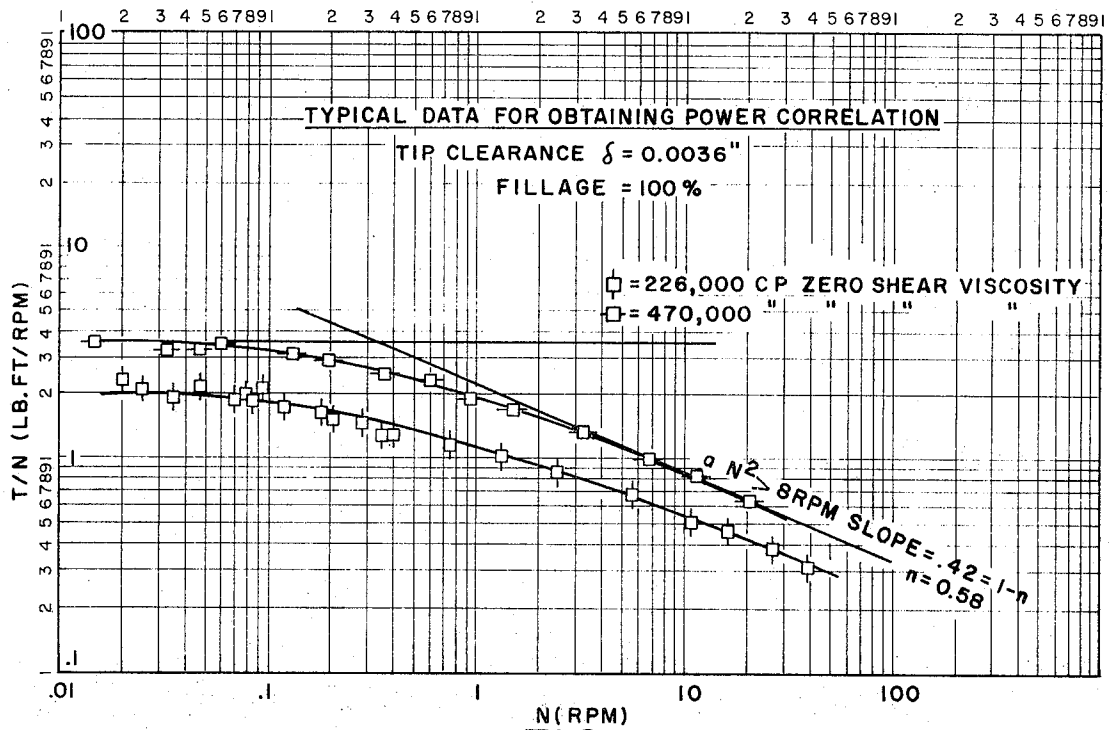


FIG.5.

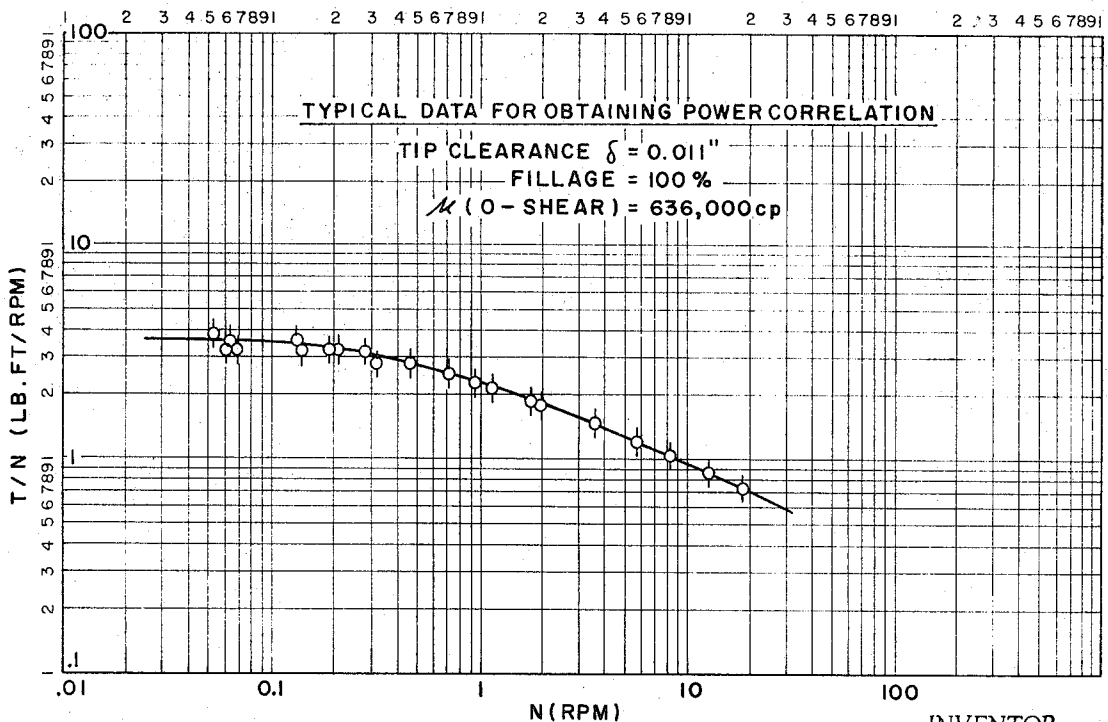


FIG.6.

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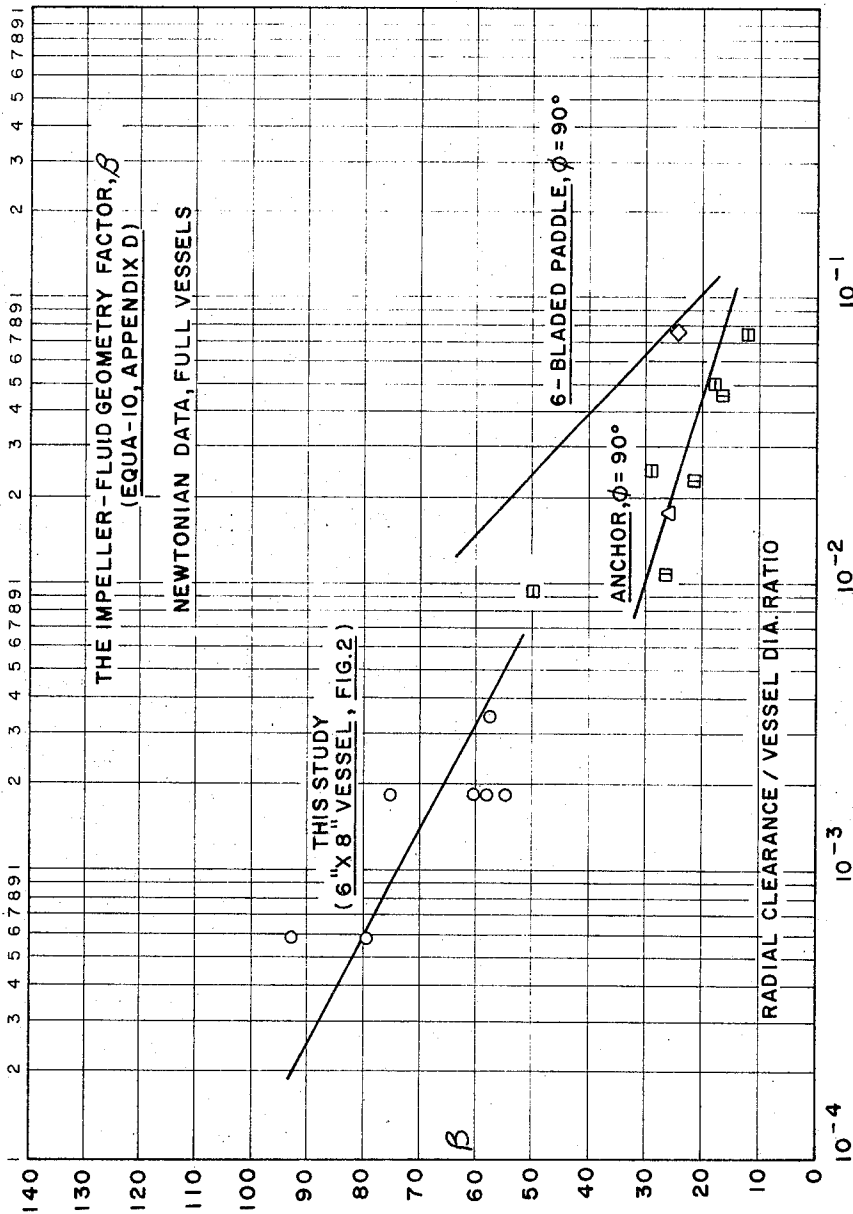


FIG. 7.

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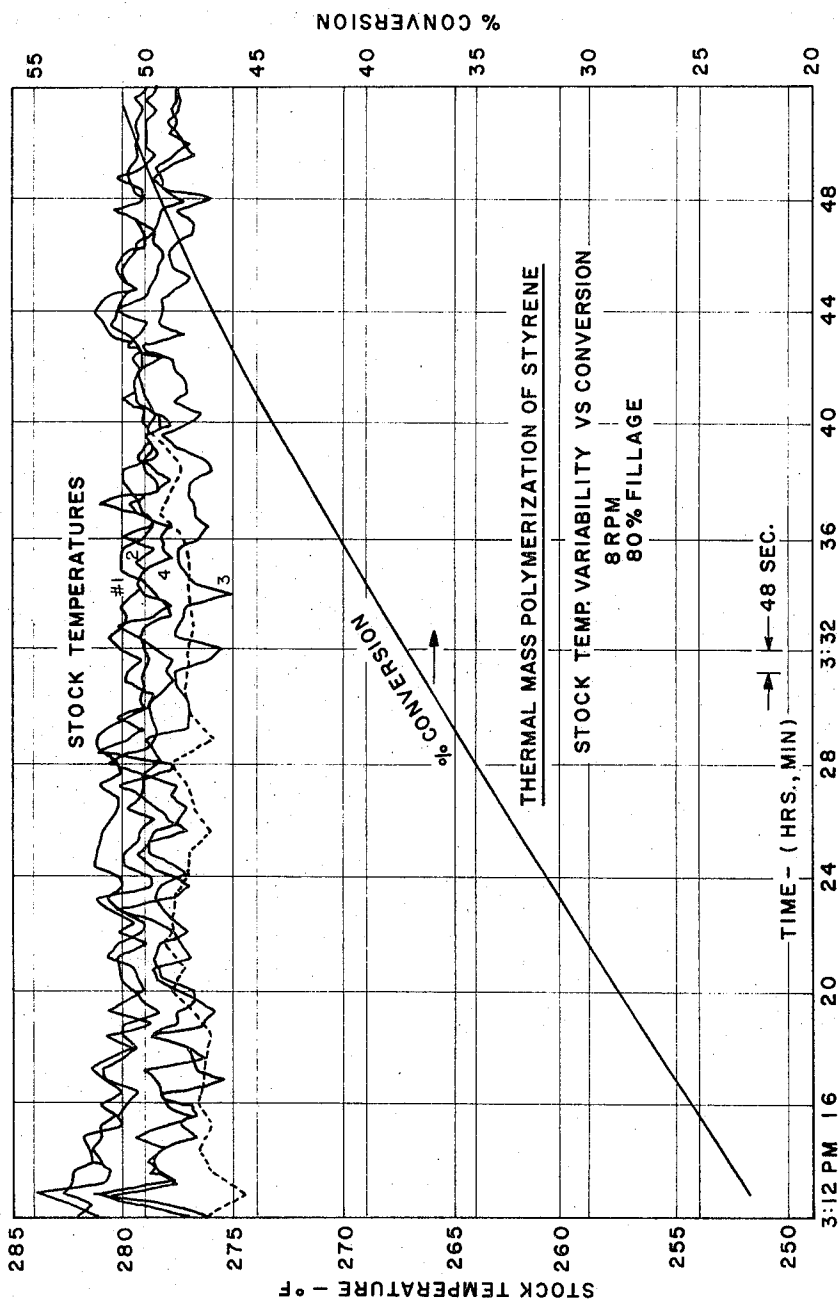


FIG. 8a.

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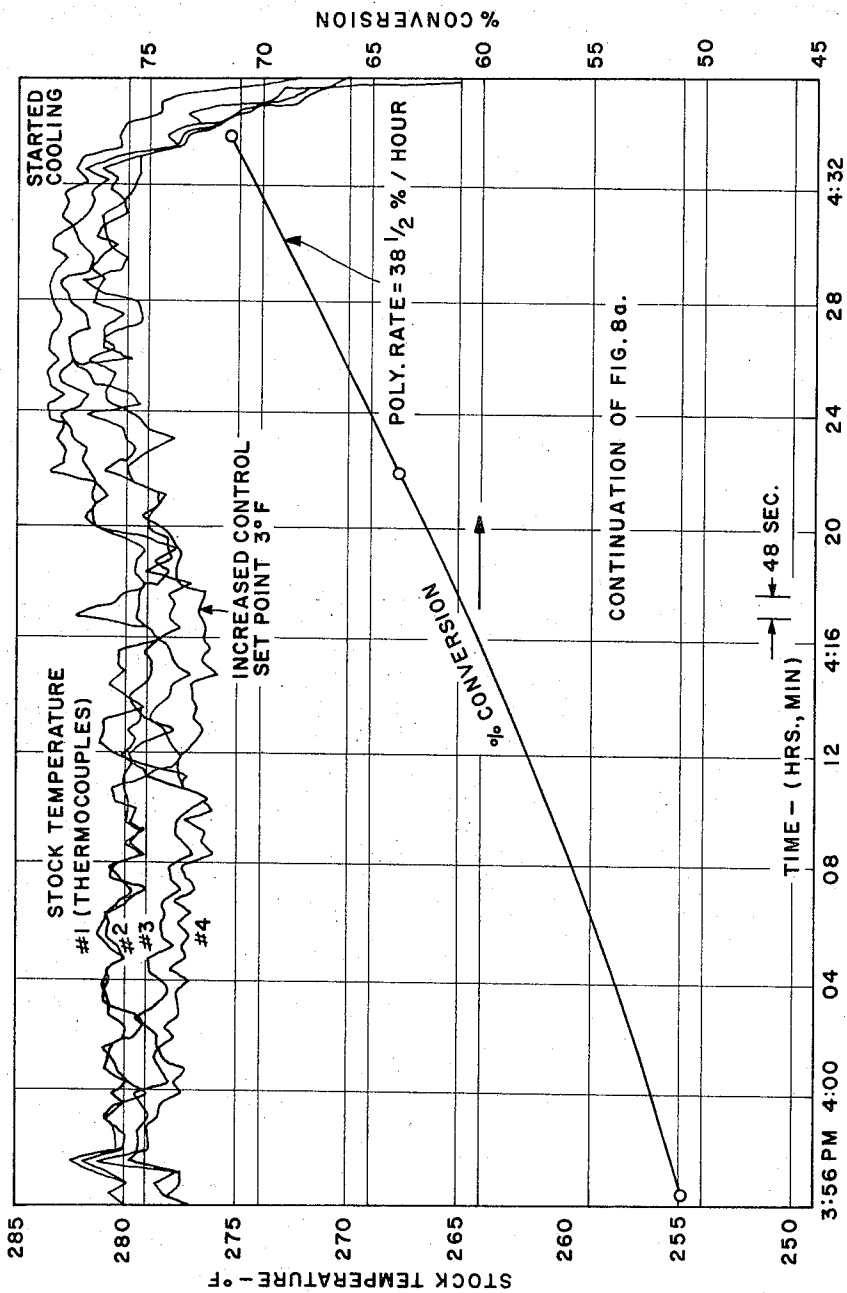


FIG. 8b.

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OVERALL WIPED-FILM HEAT TRANSFER COEFFICIENT,  $U_f$ 

(CORRELATION OF ALL DATA USING EQUATION 1)

CLOSED POINTS FROM PREVIOUS WORK:

|                                                                       |
|-----------------------------------------------------------------------|
| $\phi = 2"$ DIA. VESSEL, 2 BLADES, $\delta = .0065$ RPM'S = 7, 10, 25 |
| $\phi = 8"$ " " " 3 " $\delta = .012, .030$ " = 12, 16, 25            |
| $\phi = 16"$ " " " 3 " $\delta = 1/32, 1/16$ " = 6, 12, 16.7          |

OPEN POINTS: PRESENT STUDY USING VESSEL IN FIG. 2.

 $\delta$ 'S = .0041, .0207, RPM'S = 4, 6, 8, 20, 40, 62

VISCOSITY RANGE = 2,300 TO 360,000 CENTIPOISE

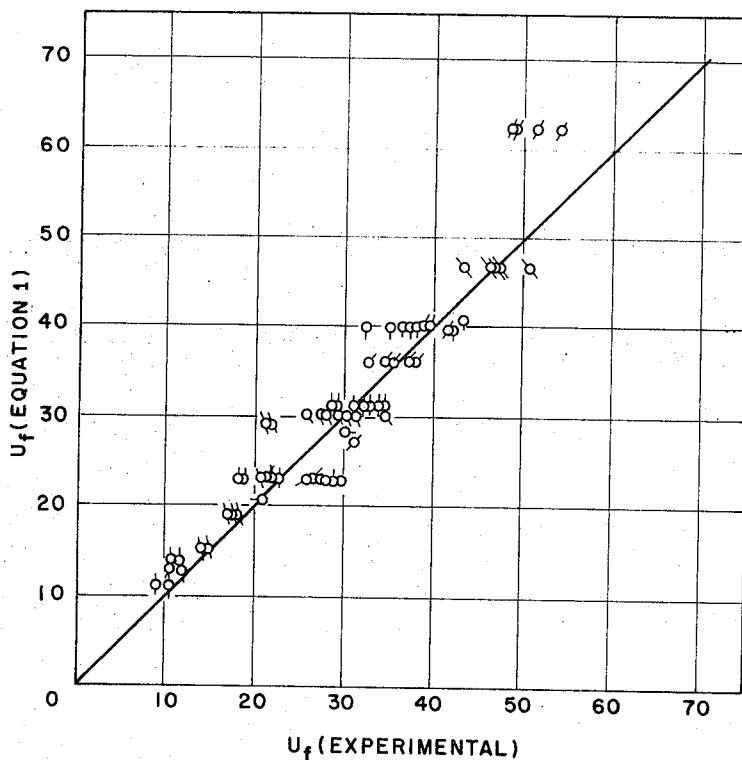


FIG. 9.

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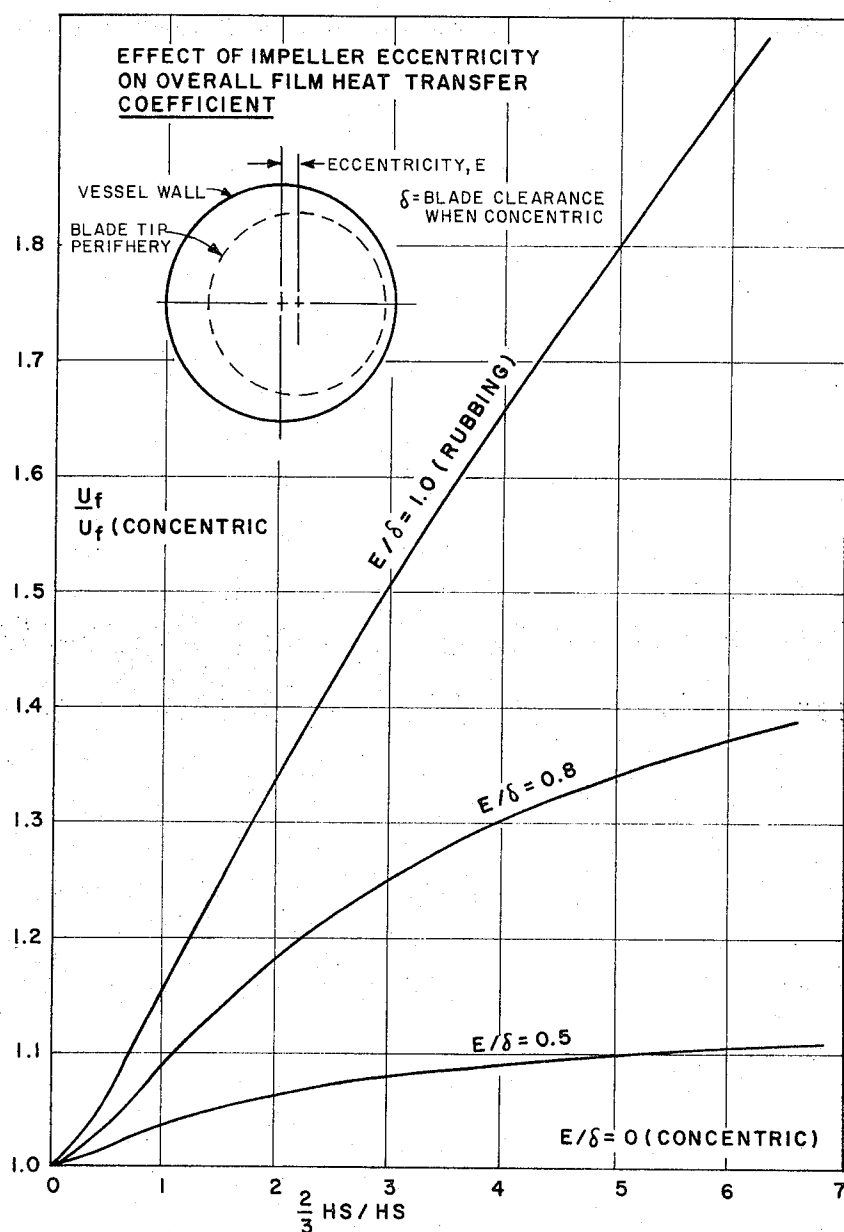


FIG. 10.

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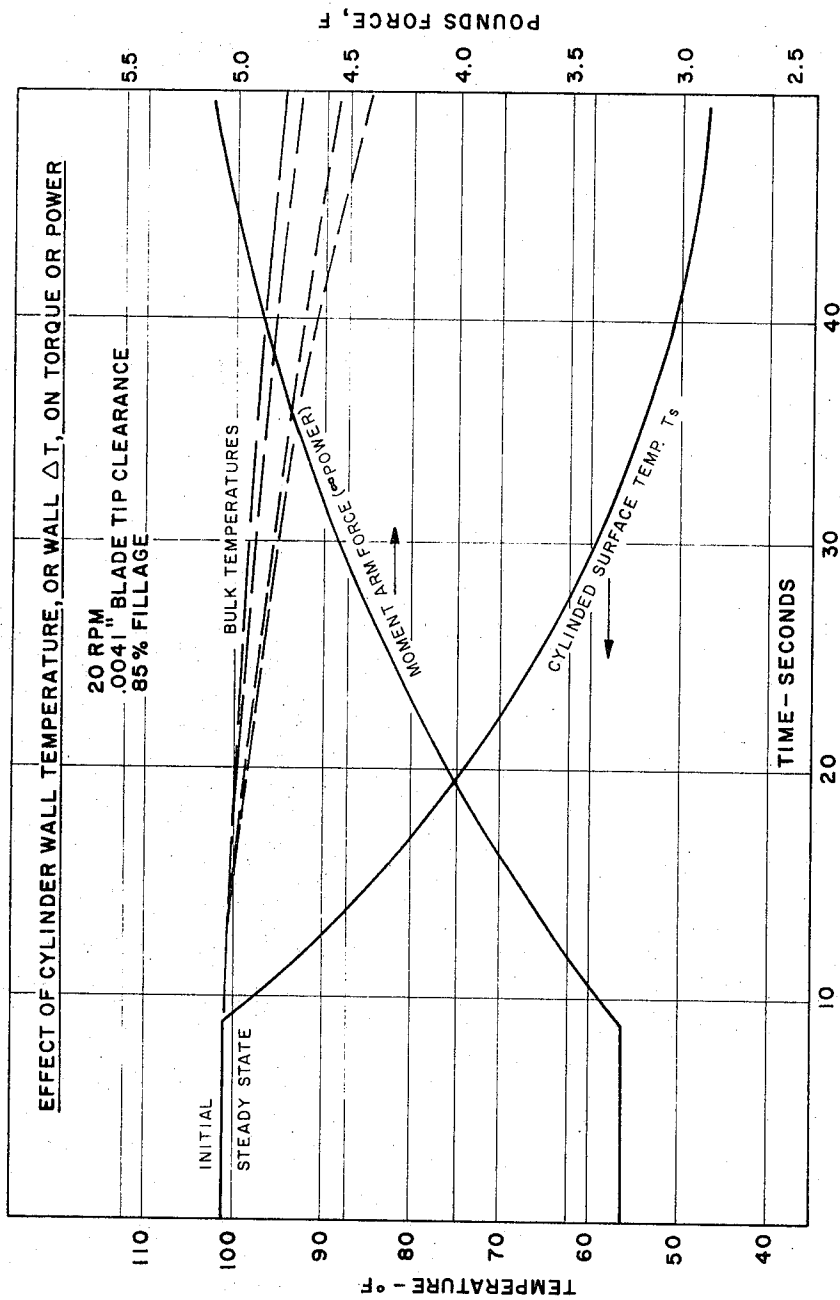


FIG. 11.

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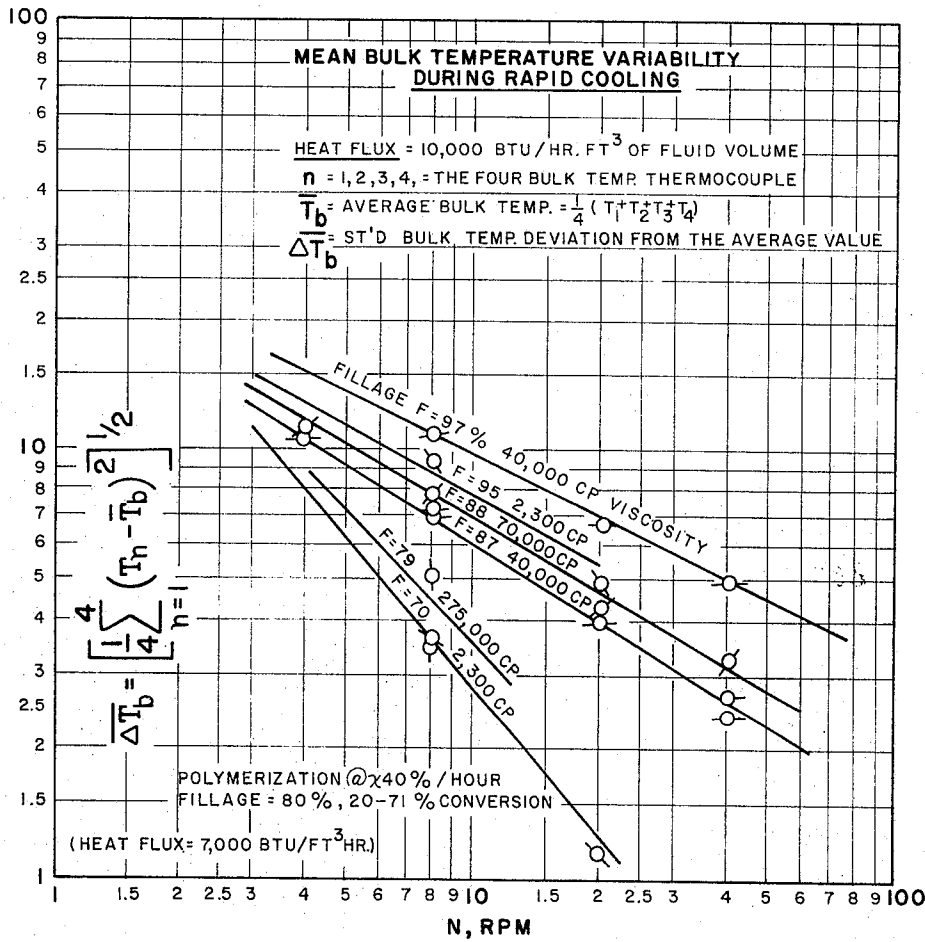


FIG.12.

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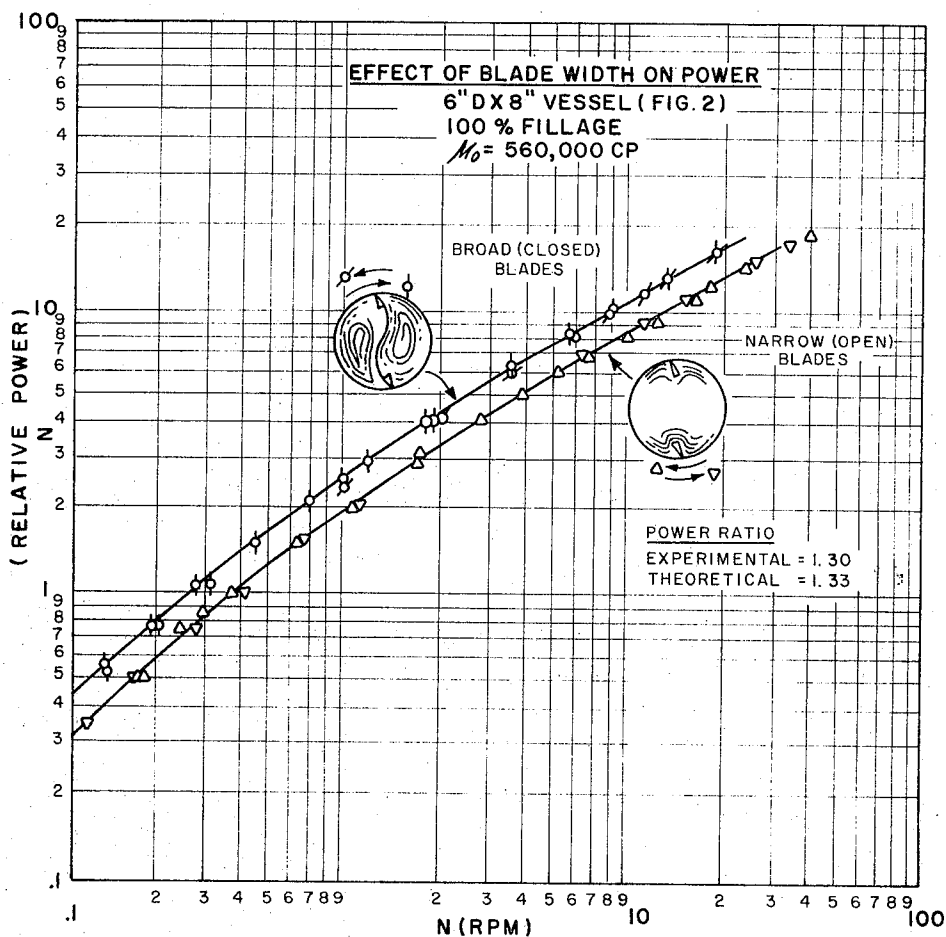


FIG. 13.

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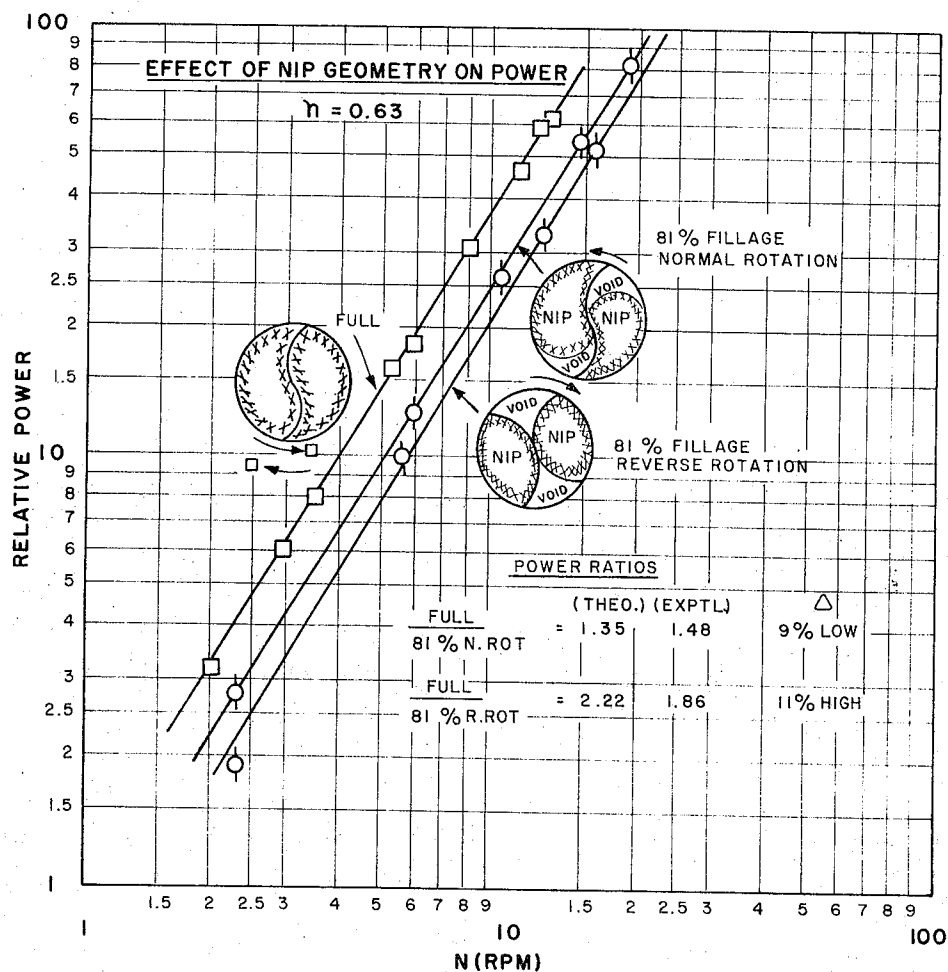
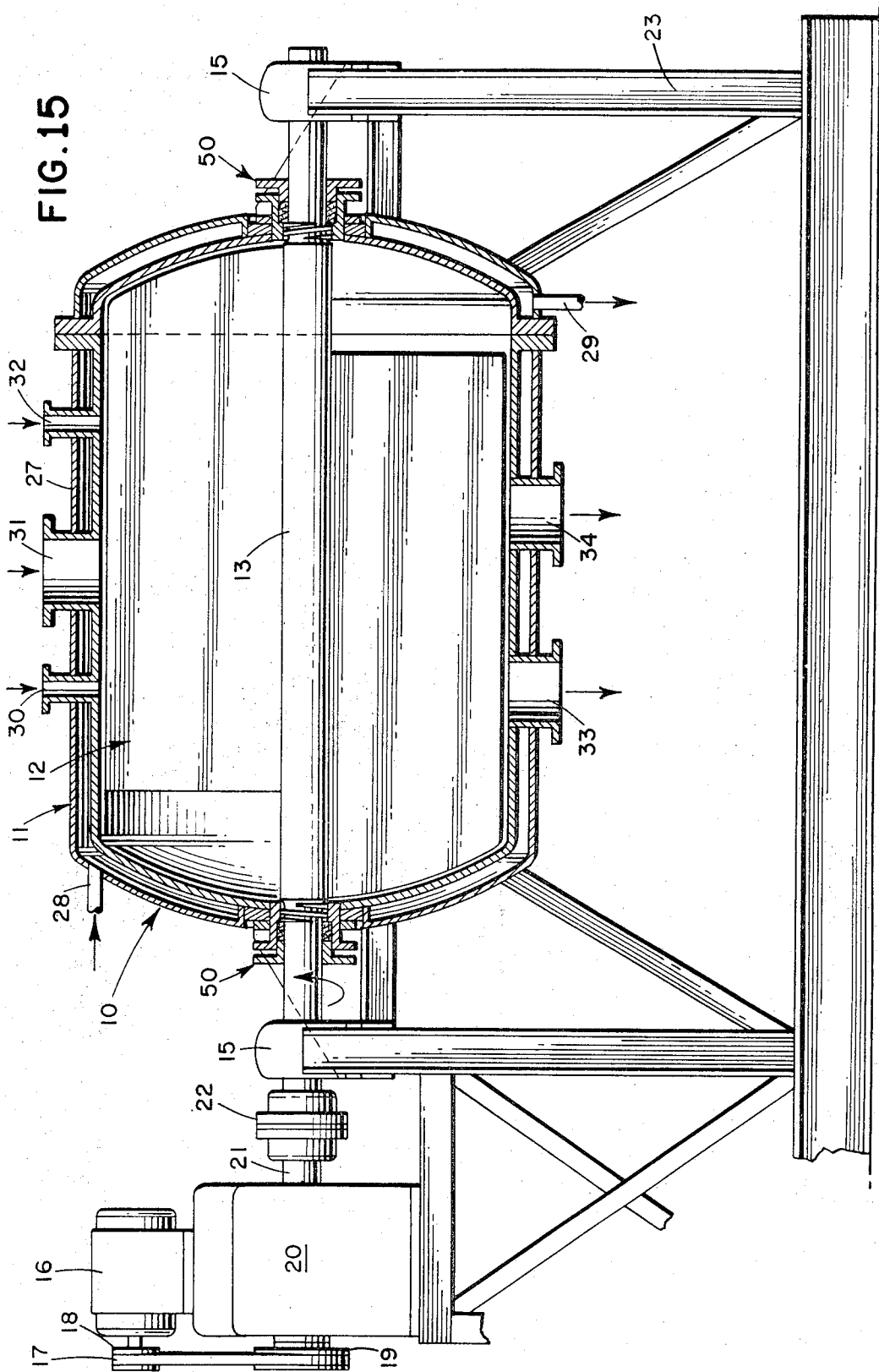


FIG. 14.

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# 1

## MIXING PROCESS

### BACKGROUND

In the art of agitating or mixing fluid and/or finely divided solid materials together, many different techniques are known. Most such techniques involve the application of one or more physical forces applied simultaneously or sequentially to a gross system in a fluid (or fluidized) state with the intended purpose of achieving a resultant mass which is homogeneous or uniform to some predetermined extent.

However, of all the known agitating methods, none has heretofore been known which produces or utilizes a combination of cyclical vertical displacement using gravitational forces, fluid recirculation in a generally horizontally extending pattern, and rolling action from pressure forces. The cyclical vertical displacement and the rolling action are preferably applied in such a way as to produce fluid randomization via fluid extension and fold-over action. There has now been discovered a new agitation process which employs such a combination. This process is very useful. For one thing, the power requirements for practicing the process tend to be surprisingly and unexpectedly low. For another, mixing times using this process to achieve uniform mixing of initially separate fluid materials tend to be surprisingly and unexpectedly short. For still another, this process can be practiced with surprisingly simple and inexpensive equipment on virtually any scale both continuously and batchwise.

One exceedingly important and highly unusual feature of this agitation process is that it enables one to mix not only relatively low viscosity liquids, but also highly viscous liquids. As those skilled in the art appreciate, the mixing or agitation of highly viscous fluids has been fraught with problems. Specialized techniques therefor are commonly considered desirable and even necessary, depending on circumstances. Operating power requirements are typically large. Uniformity and homogeneity throughout a given viscous liquid mass is commonly exceedingly difficult to achieve and maintain, owing partly to the complex fluid mechanical forces involved. With the process of the present invention, such prior art problems in the agitation of highly viscous liquids appear to be largely completely overcome, permitting one to agitate (or mix) such liquids with unexpectedly low power and in a rapid and highly efficient manner so as to produce and maintain a homogeneous and uniform liquid mass. For purposes of the present application, the terms mixing and agitation are used conventionally, synonymously, equivalently and interchangeably.

### SUMMARY

The present invention is directed to an improved process for agitating a liquid. The process is adapted to produce in the liquid so agitated substantial homogeneity and uniformity therein. The process involves subjecting a liquid which, from about 10 to 90 percent by volume, fills a generally enclosed, horizontally elongated treating zone simultaneously to a combination of three types of mixing.

One type involves cyclical vertical displacement in said zone at a cycle rate in the range from about one-half to 60 times per minute, such that:

1. first, said liquid is subjected to a vertical lifting force greater than that exerted downwardly

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thereon by gravity, and at least sufficient to move vertically at least about 10 percent of the total volume of said liquid from a gravitationally lower region to a gravitationally higher region in said zone, and then

2. secondly, such so displaced liquid is subjected to a gravitational falling force by effective removal of said lifting force therefrom, the total gravitational falling force applied thereon being at least sufficient to return substantially all of such so displaced liquid to said gravitationally lower region before said cycle is repeated on such so displaced liquid.

A second type involves rolling action in a generally peripherally located and generally horizontally extending region in said zone, said region extending circumferentially about the entire internal periphery of said zone, said region being continuously moving in a direction which is generally normal to the horizontal, such rolling action is produced by a similarly so moving band of pressure located adjacent to but following behind said region. This band of pressure exerts a force on said liquid in such region at least sufficient to cause movement of a portion of said liquid in said region along a roughly cross-sectionally circular path normally away from the adjacent internal periphery of said zone adjacent to said band of pressure towards the interior of said zone a distance which is generally less than the maximum distance across said zone at a given peripheral position and then back towards said internal periphery forwardly of said band of pressure before moving towards said band of pressure, there being a shear rate between said internal periphery and said zone of pressure of at least about 5 sec.<sup>-1</sup>, (and preferably at least 100 sec.<sup>-1</sup> and more preferably at least 1,000 sec.<sup>-1</sup>).

A third type involves horizontal displacement in said zone in a longitudinal circulatory manner at a cycle rate such that the actual volume of said liquid moved from one end region of said treating zone to the opposite end region thereof and back within one minute is equivalent to from about one-tenth to 30 times the total volume of said liquid in said zone. Such equivalent volume and the horizontal circulation rate for such liquid so moved, respectively, are each approximately proportional to such cyclical vertical displacement cycle rate in any given instance.

While such three types of mixing proceed, one continuously maintains substantially the total volume of said liquid in said zone under laminar flow conditions.

Preferable, this process is conducted so that the said cyclical vertical displacement in combination with the said rolling action produces folding action in said liquid being mixed.

This process may be practiced continuously, for example, by both continuously introducing a material to be agitated into said zone with a liquid so filling said zone being in said zone and continuously withdrawing so agitated liquid product derived from mixing said material and said liquid together from said zone. This process may, of course, also be practiced batchwise, as by first so filling said zone with a liquid to be agitated and then so agitating such liquid.

Preferably, this process is practiced by using a treating zone which is cross-sectionally circular and has a longitudinal axis which extends generally horizontally therethrough. Preferably also this process is practiced by having paddle blades rotate in said cross-sectionally circular treating zone about said longitudinal axis.

When the present invention is practiced using such a treating zone and such revolving paddle blades, it is further preferred that each of said paddle blades be further characterized by having a first region and a second region. The first region both extends axially uninterrupted at least about 50 percent along the length of said longitudinal axis, and also radially over said axial blade length uninterrupted from said longitudinal axis to a location adjacent the perimeter of said zone. The second region is axially adjacent said first region and adjacent an end of said treating zone, and said second region has discontinuities formed therethrough. Preferably, the number of such paddle blades is at least two, although more can be employed as when more than one region of rolling action circumferentially in said treating zone is desired. Preferably such regions simultaneously total no more than four though those skilled in the art might wish to use more.

This process is suitable for mixing liquids or fluids having a volume average viscosity of from about one-half to 500,000 centipoises. Preferably such a liquid has a volume average viscosity of at least about 1,000 centipoises, and more preferably a volume average viscosity of from about 10,000 to 100,000 centipoises. In other words, it is preferred to practice this invention with viscous liquids because of the superior action of mixing them generally achieved compared to other presently known mixing methods. Preferably the cycle rate for said cyclical vertical displacement ranges from about 1 to 25 times per minute and the total volume of said liquid so moved vertically is at least about 25 percent for each full revolution of a paddle assembly. Preferably, in said treating zone, in addition to the above described shear rate, the bulk average shear rate in said treating zone ranges from about 3 to 100  $\text{sec}^{-1}$ . Preferably said horizontal displacement cycle rate is equivalent to from about one-third to 10 times the total volume per minute.

Preferably, this process is practiced with a liquid so filling said zone from about 10 to 90 percent by volume having a volume average viscosity of from about 10,000 to 100,000 centipoises and the material introduced into the said treating zone for mixing with such liquid is a liquid miscible with said liquid so filling said zone and having a viscosity of from about one-half to 100 centipoises.

The present invention is suitable for mixing (agitating) virtually any fluid (liquid). Such fluid can be Newtonian or pseudoplastic, homogeneous or heterogeneous. It can be a solution, suspension, dispersion, emulsion, etc. and involve all states of matter, solid, liquid or gaseous, for example, as a dispersed phase in a continuous phase.

#### FIGURE DESCRIPTION

The invention is better understood by reference to the attached drawings wherein:

FIG. 1 is a horizontal diagrammatic view through a standardized test instrument illustrating the practice of the present invention;

FIG. 2 is a vertical sectional view through the apparatus of FIG. 1;

FIG. 3 is a graph illustrating the relationship between power consumption and volumetric percent fillage in the practice of this invention;

FIG. 4 shows a graph illustrating the relationship of paddle assembly geometry to vessel;

FIG. 5 is a graph illustrating power correlation to fluid fillage;

FIG. 6 is a graph similar to FIG. 5 but utilizing different data;

FIG. 7 is a graph illustrating relationships between paddle clearance and power;

FIGS. 8A and 8B are graphs illustrating heat removal capability using styrene batch polymerization;

FIG. 9 is a graph illustrating overall wiped-film heat transfer coefficient when practicing the present invention;

FIG. 10 is a graph illustrating effect of paddle assembly eccentricity on overall film heat transfer coefficient;

FIG. 11 is a graph illustrating effect of cylinder wall temperature on torque or power;

FIG. 12 is a graph illustrating mean bulk temperature variability during rapid cooling;

FIG. 13 is a graph illustrating effect of blade width on power consumption;

FIG. 14 is a graph illustrating effect of nip geometry on power; and

FIG. 15 is a diagram illustrating the mixer/reactor used for continuous styrene mass polymerization using the present invention.

#### EMBODIMENTS

The following examples are set forth to illustrate more clearly the principles and practice of this invention to one skilled in the art and they are not intended to be restrictive but merely to be illustrative of the invention herein contained. All parts are parts by weight unless otherwise indicated.

#### EXAMPLE A

An apparatus is constructed for test purposes as illustrated in FIGS. 1 and 2 and the following tests and evaluations are conducted:

The cylinder comprising the circumferential walls of the mixer are interchangeable with a jacketed steel cylinder through which a heated or cooled fluid is circulateable.

Experiment work is done in the 6 inches dia.  $\times$  8 inches vessel shown in FIG. 2. A glass cylinder and glass end plate are used in the visual dye tracer bulk mixing studies. A machined and jacketed steel cylinder is used for the heat transfer studies. The glass end plate is also used here in order to minimize and heat losses and to permit some visual observation of the vessel contents.

The reactor is supported by ball bearings at one end and a torque arm and scale are used to measure torque and power. Frictional losses in the impeller shaft bearings and seals are negligible compared to the torque required to turn the impeller in the high viscosity fluids.

Thermocouples are mounted on the paddle assembly. Those for measuring bulk temperatures are attached to the impeller blade and are brought out through the shaft to a set of thin, closely spaced copper slip rings which are insulated from the shaft by fiber insulators. Thermocouple wire is used for the signal pickup brushes, which are connected directly to the Speed-O-Max temperature recorder. No measurable temperature errors are observed when checked against comparison couples inserted directly into the vessel.

A sort of "tinkertoy" mixing impeller is used which could be easily modified to study the effect of various

geometry variables. It consists of a basic impeller (shown in cross-section by double lines) attached to supporting arms at the ends. This is representative of a relatively narrow bladed impeller such as an anchor type. The blade-tip inclination angles are different so that the effect of this variable can be studied by changing the direction of rotation and by varying the amount of fillage to give any degree of boundary separation behind either blade surface. Adjustable doctor blades provide accurate clearance adjustment to approximately one-eighth inch (maximum).

An S-shaped insert (shown in cross-section by single lines) is provided to convert the basic impeller to a full paddle. This shape is selected since it gives approximately the same blade-cylinder nip geometry at different fillages. Nip geometry can be varied widely by varying the fillage and direction of rotation.

Removable openings are also provided at the opposite ends of this center insert to determine whether sufficiently large axial pressure gradients could be generated in the blade nips to give rapid axial recirculation without the necessity of resorting to helical blades or screws.

All important surfaces are carefully machined. Cylinder roundness and impeller concentricity are nearly perfect.

Temperatures are recorded by a 24-point recording instrument.

#### EXAMPLE I

##### POWER CONSUMPTION VS. PERCENT FILLAGE

Using a slotted two-bladed paddle assembly whose dimensions are about 6 inches inside diameter by 8 inches in length, the apparatus of Example A is used for a series of evaluations. The paddle assembly is diagrammatically slotted and its opposite corners and each slot is sized about 1 1/2 to 2 1/2 inches. The blade is radially curved so that such an S-shaped geometry cross-sectionally in the paddle assembly causes such assembly to retain similar geometry at different fillages. The apparatus of Example A is filled to various levels with a variety of different fill percentages ranging from a full vessel down to a near empty vessel. The liquid used is a pseudoplastic fluid comprising a solution of polystyrene in ethyl benzene and has a viscosity of about 200,000 centipoises at 10 sec.<sup>-1</sup> shear rate. The paddle assembly is revolved at about 25 rpm. The data resulting are plotted as points in FIG. 3. Each data point is encircled. Low power consumption is indicated for this range of viscosities. For mixing of the type here involved in this invention, the following equation is derived (for a Pseudoplastic fluid,  $n < 1$ , where  $\tau = K\gamma^n$ ):

Power (per nip)

$$= \frac{\pi L D^2 N^{1+n} K}{(1-n)} \left[ \frac{180b \left( \frac{2n+1}{3n} \right)}{\phi} \right]^n \left[ \left( \frac{X}{D} \right)^{1-n} - \left( \frac{\delta}{D \sin \phi} \right)^{1-n} \right] \quad (4)$$

Power (total)

$$= \frac{\pi L D^2 N^{1+n} K}{(1-n)} \sum \left[ \frac{180b \left( \frac{2n+1}{3n} \right)}{\phi} \right]^n \left[ \left( \frac{X}{D} \right)^{1-n} - \left( \frac{\delta}{D \sin \phi} \right)^{1-n} \right] \quad (5)$$

where:

$D$  = vessel diameter

$L$  = blade length (vessel length)

$V$  = blade tip speed

$\delta$  = blade tip clearance (radial)

$\phi$  = blade tip inclination angle at wall

$N$  = rotational speed of paddle assembly

$X$  = "wetted" circumferential length or perimeter

$n$  = power law exponent

$\gamma$  = power-average shear rate ( $=\beta N$ )

$K$  = power law "fluidity index" ( $\tau = K\gamma^n$ )

$b$  = dimensionless constant

$\tau$  = shear stress

The summation, as above, includes all the fluid-filled blade nips. The fluid consistency index,  $K$ , is to be evaluated at the power average shear rate,  $\bar{\gamma}$ , given by:

$$\bar{\gamma} = \beta N$$

where:

$$\beta = \left[ \sum \frac{180b}{\pi \phi_i} \ln \left( \frac{X_i \sin \phi_i}{\delta} \right) \right] \left( \frac{\pi D}{\sum X_i} \right) \quad (6)$$

and where:

$\beta$  = coefficient for determining power-average shear rate. When a vessel is completely full,

$$\sum X_i = \pi D$$

Plots of impeller geometry factor are shown in FIG. 4.

The constant,  $b$ , can be evaluated from power data, using the equation:

$$P(\text{Total}) = \pi^2 L D^2 N^2 \mu_0 \sum (180b/\pi \phi_i) \ln(X_i \sin \phi_i / \delta) \quad (8)$$

and a Newtonian fluid. Thus, to determine  $b$ , the impeller shown in FIG. 2 is used with center insert in, end ports shut, and operating completely full. The power contributed by the ends was estimated at 9 percent of the total and was subtracted from the measured power to give the net cylinder power (equation 8). The following  $\phi$ 's and  $X$ 's were estimated for this impeller:

$$2 \text{ nips with } \phi = 35^\circ X = \frac{2}{3} (\pi D/2)$$

$$2 \text{ nips with } \phi = 113^\circ X = \frac{1}{3} (\pi D/2)$$

Several polystyrene syrups and blade tip clearances were used. Power data were taken at decreasing rotational speeds until the Newtonian region was reached, as indicated by constant Torque/ $N$ , see FIGS. 5 and 6. The following  $b$  values were calculated, using equation 8:

|                                           |      |      |      |      |      |      |
|-------------------------------------------|------|------|------|------|------|------|
| $\delta/D \times 10^4 = 5.83$             | 5.83 | 18.3 | 18.3 | 18.3 | 18.3 | 34.5 |
| $\mu_0 \times 10^{-3} = 2.26 \text{ cp.}$ | 4.70 | 6.40 | 5.0  | 0.26 | 2.20 | 4.40 |
| $b = 3.1$                                 | 2.7  | 2.4  | 2.4  | 3.1  | 2.3  | 2.6  |

$b$  (average) = 2.7, average deviation  $\pm 11\%$ .

The value  $b = 2.7$  is reasonable.

$K$  and  $n$  are evaluated at the power-average shear rate,  $\bar{\gamma}$  given in equation 6, for a Newtonian fluid

$$\text{Power (per nip)} = \pi^2 L D^2 N^2 \mu_0 (180b/\pi \phi) \ln[X \sin \phi / \delta] \quad (2)$$

$$\text{Power (total)} = \pi^2 L D^2 N^2 \mu_0 \sum (180b/\pi \phi_i) \ln[X_i \sin \phi_i / \delta] \quad (3)$$

$b$  = experimentally determined correction factor 2.7

for all geometries

The summation includes all the "active," i.e. fluid-filled blade nips, where  $X_i$  is the wetted perimeter which comes under the influence of blade nip of angle  $\phi_i$ . For example, in the case of a simple broad-bladed 90° anchor agitator with no fluid separation behind the blades, there are four identical nips. Therefore,  $X = \pi D/4$  and  $P$  (total) =  $4 \times P$  (per nip), where  $P$  = power.

Equation 5 is plotted as a solid line in FIG. 3.

In the data recorded in FIG. 3,  $n = 0.57$  and  $K$  is adjusted to give a fit at approximately 100 percent fillage. It is seen that agreement between equation 5 and the experimental data is excellent down to a fillage of about 0.3 per cent. No serious data deviation down to fillage levels as low as 0.09 per cent is observed. As the fillage approaches 100 per cent, the power quickly increases about 30 per cent as the fluid starts contacting rear blade surfaces. This power increase is believed to be caused by the fact that two additional high shear fluid-filled blade nips are formed when this contact is complete.

FIG. 7 shows the calculated  $\beta$  vs.  $\delta/D$  curves for several geometries (with  $b = 2.7$ , 100 percent full) and what supporting data were available from the literature, plus the data from this study. Agreement is good down to fairly high  $\delta/D$  ratios. With decreasing radial clearance to vessel diameter ratio, there is increasing power consumption.

Foresti and Liu provide the only known available power data with pseudoplastic fluids at reasonably small impeller clearances ( $\delta/D = 0.0171$ ), using an anchor agitator:

| Fluid              | K* n     | Relative Power        |                          | $\Delta$     |
|--------------------|----------|-----------------------|--------------------------|--------------|
|                    |          | This work<br>eq. 5, 7 | Foresti's<br>correlation |              |
| 5% polyisobutylene | 816 0.34 | 10.8                  | 9.1                      | 18-1/2% high |
| 10% CMC solution   | 521 0.52 | 20.5                  | 26.0                     | 21% low      |

\* (dyne sec<sup>2</sup>/cm<sup>2</sup>)

The agreement between, for example, Foresti's correlation and the power required for the present invention is considered quite good, considering that Foresti's values are based on the least-mean-square lines drawn through data points which represented four different impeller geometries and which scattered by more than 50 percent about the mean value. Since the same symbol is used for all impellers, it is impossible to tell where the anchor data fell with respect to the total average.

#### EXAMPLE 2

Using a slotted two-bladed paddle assembly in the apparatus of Example A this apparatus is employed with four thermocouples affixed to various paddle assembly locations. Under a jacket heat, input variations in the four temperatures sensed by the thermocouples are observed. The fill in this case is styrene monomer which is polymerized under mass conditions batchwise. When the stock temperature within the reactor reaches about 250°F., measurement of per cent conversion is made periodically. Results are recorded in FIGS. 8A and 8B.

Excellent mixing is illustrated by the fact that bulk temperature variability is only about 3° to 4°F. during mass polymerization at conversion rates of about 40 per cent per hour (corresponding to volumetric heat flux or evolution of about 7,000 Btu/hr.ft.<sup>3</sup>). The bulk temperature spread is just as narrow at 71 per cent con-

version as it is at 20 per cent conversion.

#### EXAMPLE 3

Using an apparatus as described in Example A above, the following evaluations were conducted:

Solutions of polystyrene in ethyl benzene having varying high viscosities are prepared and each solution is charged into the vessel so that the fill level is about 80 per cent. A dye tracer is slug injected into the vessel near the cylinder wall at the midpoint in top center. Each slug comprises typically a 0.1 weight per cent solution of ethyl benzene of anthraquinone blue dye code 357 or equivalent. Each slug is introduced in 20 milliliter units. The agitator (or paddle assembly) is revolving at a rate of about 8.5 RPM. Bulk mixing effectiveness is judged by the rate at which the dye spreads radially and axially throughout the fluid mass in the mixer to give a uniformly collected solution which is free of color striations or color gradients. The fluid mass is carefully inspected for the presence of regions of low fluid recirculation and shear. It is found that the dye tracer slug is dispersed and diffused throughout the fluid volume in the mixer in each instance most rapidly and uniformly. The solutions range in viscosity from about 5,000 to 600,000 centipoises.

Concentrated slugs of injected dye tracers got uniformly dispersed throughout the volume in about 8 revolutions, and all visible color striations disappeared within 24 revolutions (viscosities up to 600,000 centipoise). Thus:

1. 100 Percent Fillage, End Ports Open, 660,000 cp Viscosity

Good axial recirculation, but very poor cross-mixing of streamlines. Dye striations remained for a long time. Even after 10 minutes, the center core was relatively clear.

2. 80 Percent Fillage, End Ports Shut, 660,000 cp

Axial mixing was reduced practically to zero and the dye remained in a narrow 1 inch wide color band at the point of injection. Cross-sectional mixing was quite good, but some dye striations were visible after 3 minutes.

3. 80 Percent Fillage, End Ports Open, 660,000 cp

Excellent bulk mixing. Some dye reached end of vessel in about 6 seconds. Macroscopically dispersed throughout vessel in about 1 minute, i.e. dye was uniformly dispersed but striations visible. Practically homogeneous after 2 minutes. Uniform color with no striations present after 3 minutes.

4. 80 Percent Fillage, End Ports Open, 200,000 cp

Visually judged to be about same as 3.

It was found that the dye tracer slug is dispersed and diffused throughout the fluid volume most rapidly and uniformly by operating at partial fillage in a horizontal reactor, using a close-fitting full paddle with openings at the opposite ends of the paddle.

Bulk mixing effectiveness is also correlated on a somewhat more quantitative basis by measuring local bulk temperatures at four points within the fluid during rapid batch heating and cooling, and using the mean temperature deviation at a given heat flux as a relative measure of bulk mixing excellence under various conditions (see other Examples).

#### EXAMPLE 4

A comparison between experimental values and calculated values for the wiped-film heat transfer coeffi-

cient is illustrated by FIG. 9. Calculated values are derived from the following equation:

$$1/U_f = (1/h_s) + (1/h_s) + (1/h_b) + (1/h_s) + (1.5/h_s)$$

(1) 5

where

$$h_s = \text{wiped-film component} = 2(k\rho c_p NN_B / \pi)$$

$$h_b = \text{clearance film component} = k/\delta' = 4k/3\delta$$

$$h_b = \text{"buffer" layer component} = 2h_s$$

$$k = \text{thermal conductivity}$$

$$\delta' = \text{effective thickness of the clearance film}$$

$$\rho = \text{fluid density}$$

$$c_p = \text{heat capacity (thermal conductivity of fluid)}$$

$$N_B = \text{number of impeller blades}$$

$$U_f = \text{overall wiped-film fluid heat transfer coefficient}$$

$$(h_b, h_s, h_s = \text{individual fluid film heat transfer coefficients})$$

and other factors as above defined.

The scatter is appreciable; but the experimental precision probably is no better than  $\pm 10$ –15 percent. Except for one set of data which is definitely out of line, the scatter is more or less random and no significant trends due to viscosity, heating or cooling rate, etc. were noticed.

#### EXAMPLE 5

In FIG. 10, the effect of an off-center agitator is illustrated. Such an agitator results in a variable top clearance around the periphery of the vessel. Its effect is to always increase the overall fluid film coefficient over what it would be if the agitator were perfectly centered or concentric with the axis of the vessel.

The average overall fluid film heat transfer coefficient:

$$U_t = \frac{1}{\pi} \int_0^\pi U_\phi d\phi = \frac{M}{\pi} \int_0^\pi \frac{d\phi}{N - P \cos \phi}$$

This solves easily to give:

$$U_t = \frac{\frac{2}{3} h_s h_b}{\sqrt{\left(h_s + \frac{2}{3} h_s\right)^2 - \left(\frac{2h_s E}{3\delta}\right)^2}}$$

where

$$h = \text{the concentric clearance film coefficient,}$$

$$E = \text{eccentricity} \leq \delta$$

$$\delta = \text{normal concentric radial clearance, } \ll \text{ radius}$$

$$\delta_\phi = \text{radial clearance at any angle } \phi$$

$$h_\phi = \text{local clearance film coefficient at angle } \phi$$

$$P = \text{power}$$

The importance of impeller eccentricity depends on the ratio of the combined wiped-film and buffer layer coefficient to the clearance film coefficient, i.e. it is a function of  $(\frac{2}{3}) (h_s)/h$ . This relationship is given in FIG. 5 for several  $E/\delta$  values.

#### EXAMPLE 6

The effect of cold cylinder wall on power consumption is illustrated by FIG. 11 where the experimental results achieved by suddenly lowering the vessel wall temperature and recording bulk temperatures, cylinder surface temperature, and torque vs. time are noted.

The following relationship is assumed:

$$[\text{Power (at } T_s, T_b)] / [\text{Power (at } T_s = T_b)] = (\text{fluid viscosity at } T_s^x) / (\text{fluid viscosity at } T_b)$$

where

$$T_s = \text{surface temperature}$$

$T_b$  = average bulk temperature

The exponent  $x$  varied with time from about 0.3, after short time intervals, to about 0.5, after longer time intervals.

The value  $x = 0.3$  is perhaps representative of good bulk mixing at high rotational speeds where the temperature gradients are confined to a very thin surface layer.

The value  $x = 0.5$  perhaps representative of a poorer bulk mixing situation at lower rotational speeds where the temperature gradients penetrate deeper into the bulk fluid.

A cooling experiment is performed in the 6 inches = 8 inches mixer/reactor at the following conditions: 20 RPM, 0.0041 inch clearance, 85 percent fillage, using 50 percent polystyrene syrup. It is seen that if a large boundary layer temperature differential is necessary to remove reaction heat from high viscosity fluids, a substantial increase in power input is expected when using close-clearance or scraper blade agitators.

#### EXAMPLE 7

To obtain a power correlation between paddle assembly r.p.m. and fluid being mixed, experimental data as shown in FIGS. 5 and 6 are developed.

In power measurements, the vessel of FIGS. 1 and 2 is employed (supported by ball bearings and the power consumption is determined from the measured torque. The zero load torque, i.e. with the vessel empty, is checked periodically and found to be negligible compared to torque under load. Percent fillage in the vessel is calculated from the known vessel free volume and the weight and density of the charge. When operating at partial fillage, the fraction of the cylinder area in contact with fluid was measured at relatively low rotational speeds. At higher speeds, the fraction wetted contact area is estimated from the known fillage, geometry, and the observed curvature of the free surface of the fluid.

The constant correction factor,  $b$  ( $= 2.7 \pm 10$  percent mean deviation) is calculated from power measurements using equations 3 and 7. As high viscosity Newtonian fluids, polystyrene solutions are used. Torque measurements are made at decreasing rotational speeds until the Newtonian range is reached, i.e. until the ratio of torque to RPM became constant, thus indicating a constant viscosity. Unfortunately, very low rotational speeds, and hence torque, are necessary to reach the Newtonian range. This is responsible for the scatter and uncertainty in the data shown in FIGS. 5 and 6 and in the variability in the calculated  $b$  values, which are based on these data.

The overall wiped-film heat transfer coefficients is calculated from batch cooling and heating data. The thermal capacity of the impeller and steel end plate are also taken into account. The impeller temperature follows the average bulk temperature quite closely. The shaft end of the vessel insulated, and temperature measurements indicated that the thin steel end plate also followed the average batch temperature quite closely. Heat flow down the stainless steel shaft are quite small compared to the other heat effects and is neglected. Frictional dissipation calculated from torque measurements and included in the total heat flux calculation.

The overall polymer film coefficient,  $U_f$ , is then calculated using the central portion of the heating or cooling curves and the log mean  $\Delta T$ , where the batch

temperature is taken as the average of the four batch thermocouple readings (see examples above).

#### EXAMPLE 8

Mean bulk temperature variability during rapid cooling is illustrated by data recorded in FIG. 12.

The bulk mixing effectiveness was judged, for the most part, visually. A quantitative comparison, in terms of bulk temperature variability, was also obtained from rapid batch cooling data. This is shown in FIG. 11, where the bulk temperatures were taken at the point on the cooling curve which corresponds to a rate of heat flux from the melt of about 10,000 Btu/hr. ft.<sup>3</sup>

Rotational speed influences is responsible for all the mixing actions that take place.

It is also seen that viscosity, per se, is of little importance, but percent fillage is important. At fillages of 80 percent and below, the random gravity mixing action comes into play which results in a striking improvement in temperature uniformity, even at high viscosities.

The reason that the temperature uniformity is so much better during mass polymerization is that the reaction heat is liberated uniformly throughout the fluid. Also, the heat flux is appreciably less: 7,000 Btu/hr. vs. 10,000 Btu/hr. per ft.<sup>3</sup>

The effective clearance film thickness, for example, would be expected to depend to some extent on land width, clearance, speed, etc. The so-called "buffer-layer" coefficient, also, was introduced to account for imperfect bulk mixing, and may not be strictly proportional to the wiped-film coefficient (i.e. proportional to rotational speed and number of impeller blades) over a wider range of variables than were covered in this work.

If there is any eddy motion behind the blades, the actual overall film coefficient would be greater than that predicted from equation 1, especially at large tip clearances. Based on the familiar Rushton power number versus Reynolds number plots, it is generally agreed that eddy motion or turbulence begins at Reynolds numbers of about 10, at which point the slope begins to deviate from -1. Therefore, the present heat transfer correlation should not be used at Reynolds numbers greater than 10. In fact, it should be used with caution at Reynolds numbers somewhat less than 10 since some eddy motion may take place in the regions behind the blade tips before it shows up in the power plots.

#### EXAMPLE 9

The effect of blade width on power consumption is illustrated in FIG. 13. It is seen that the effect of blade width on power input is relatively small, as one might expect since most of the frictional dissipation takes place in the nip region formed by the blade and cylinder wall. Agreement with theory is excellent.

A moderate increase in power in the case of a broad blade is more than compensated by the superior, far-ranging shear field, which makes the present invention desirable for use situations where good bulk mixing is important.

#### EXAMPLE 10

The effect of fluid nip geometry on power is illustrated by FIG. 14. Here, for illustration, are used 4 nips (2 high shear, 2 lower-shear) versus 2 nips (high shear) versus 2 nips (lower shear). The theoretical power ratios agree fairly well with the experimental values.

#### EXAMPLE 11

To a mixer adapted for the practice of the present invention and equipped for use as a reactor, the mixer being of the type illustrated in FIG. 1 having a paddle assembly of the type likewise shown in FIG. 1 is continuously charged, in liquid spray form, styrene monomer through an input port. The fresh monomer charge rate is about 50 pounds/hour and the temperature of the so-charged liquid styrene monomer is about 60°F. Concurrently, after steady state conditions are achieved, there is continuously withdrawn from the reactor through output ports 33 and 34 a polymerized melt product at a flow rate of about 70.1 pounds/hour. The polymerized melt product comprises approximately 70 weight per cent polystyrene having a number average molecular weight of about 115,000 (about 55,000 Staudinger) dissolved in the balance up to 100 weight per cent styrene monomer. The polymer has a dispersion index of about 2.5. The polymerized melt product withdrawn from the mixer/reactor has a viscosity of about 40,000 centipoises at about 300°F. at a shear rate of about 10 sec.<sup>-1</sup>. Hold-up time in the mixer/reactor is about 4.6 hours and the conversion rate of monomer to polymer in the reactor is about 22 pounds of polymer made per hour per pound of hold up.

The mixer/reactor is maintained at about a 65 per cent volumetric fillage level based on the substantially unexpanded liquid phase at 300°F. and the paddle assembly rotates therein at about 12 RPM. The contents of the mixer/reactor are maintained in a substantially homogeneous and substantially isothermal condition at about 300°F. The mixer/reactor is jacketed and the fluid circulated in the jacket is maintained at about 300°F.

The mixer/reactor is equipped with reflux condenser which is interconnected with the mixer/reactor at input port 31. Vaporized styrene monomer is removed from the upper vapor phase portion within the mixer/reactor and passed into this condenser (thus passing by an input spray head in input port 31) at about 300°F. The monomer vapor is condensed and sub-cooled to about 60°F. in the reflux condenser and is then returned to the mixer/reactor. The rate of monomer vapor removal is adjusted so as to maintain the temperature in the mixer/reactor interior at about 300°F. and so as to maintain the pressure in the mixer reactor at about 13.4 psia. At this pressure, the volume of the viscous fluid mass in the mixer/reactor is found to be expanded by bubbles of monomer vapor therein to an extent of about 15 per cent over the volume of this mass, when for example, the pressure thereof is maintained momentarily at about 15 psia. (but at about 300°F.) when it is observed that there are substantially no vapor bubbles entrained in the mass. In the reactor, the shear rate is about 10 sec.<sup>-1</sup>, the horizontal displacement rate is about 8 times the equivalent total volume of the liquid phase per minute, and cyclical vertical displacement is about 24 times per minute.

The mixer/reactor maintains the materials therein in a substantially homogeneous, uniform condition and the temperature conditions in the mixer are substantially isothermal. The horse power consumed in each instance when the paddle blade revolves at 12 RPM under steady state conditions is estimated to be about 38.

Referring to FIG. 15, there is seen the mixer/reactor assembly used in this Example which assembly is herein designated in its entirety by the numeral 10. Assembly 10 is seen to comprise a vessel 11 having an impeller assembly 12. Impeller 12 extends through vessel 11 on a shaft 13. Where it passes through vessel 11, shaft 13 is sealed by seals 50 (paired). Shaft 13 is journaled for rotational movements by a pair of bearing assemblies 15.

A motor 16 is connected by a belt 17 over sheaves 18 and 19 to a transmission or reducer 20. Transmission 20 has a drive shaft 21 which interconnects with shaft 13 through a coupling 22. The mixer/reactor assembly 10 is supported by a frame 23.

Vessel 11 has an inner wall 26 and spaced therefrom, an outer wall 27, with the space therebetween serving for circulation therethrough of a heating or cooling fluid, as through an input conduit 28 and an output conduit 29. Material (not shown) for mixing and/or reacting may be fed into vessel 11 through conduits 30, 31, and/or 32 continuously or discontinuously and may be removed therefrom through conduits 33 and/or 34 in conventional ways as those skilled in the art will appreciate. For example, if assembly 10 is to be used as a reactor for continuous mass polymerization of a monomer such as styrene, conduit 31 may be connected to a reflux condenser assembly (not shown); styrene may be continuously sprayed into vessel 11 through a conduit 30, mass polymerized in a partially filled vessel 11, and then continuously removed from vessel 11 through conduit 34, agitation being accomplished by the revolution of impeller 12.

It will be appreciated that, while the embodiments of the present invention as shown and described herein are necessarily limited to a few forms of the present invention, many variations and modifications thereof are feasible and practical without departing from the spirit and scope of the present invention disclosed and claimed herein.

What is claimed is:

1. In an improved process for agitating a liquid and adapted to achieve and maintain substantial homogeneity and uniformity therein, the step which comprises subjecting a liquid which, from about 10 to 90 per cent by volume, fills a generally enclosed, horizontally elongated treating zone simultaneously to a combination of:

A. cyclical vertical displacement in said zone such that at a cycle rate in the range from about one-half to 60 times per minute,

1. first, said liquid is subjected to a vertical lifting force greater than that exerted downwardly thereon by gravity, and at least sufficient to move vertically at least about 10 per cent of the total volume of said liquid from a gravitationally lower region to a gravitationally higher region in said zone, and

2. secondly, such so displaced liquid is subjected to a gravitational falling force by effective removal of said lifting force therefrom, the total gravitational falling force applied thereon being at least sufficient to return substantially all of such so displaced liquid to said gravitationally lower region before said cycle is repeated on such so displaced liquid,

B. rolling action in a generally peripherally located and generally horizontally extending region in said zone, said region extending circumferentially about

the entire internal periphery of said zone, said region being continuously moving in a direction which is generally normal to the horizontal, said rolling action being produced by a similarly so moving band of pressure located adjacent to but following behind said region, said band of pressure exerting a force on said liquid in said region at least sufficient to cause movement of a portion of said liquid in said region along a roughly cross-sectionally circular path normally away from the adjacent internal periphery of said zone adjacent to said band of pressure towards the interior of said zone a distance which is generally less than the maximum distance across said zone at a given peripheral position and then back towards said internal periphery forwardly of said band of pressure before moving towards said band of pressure, there being a shear rate between said internal periphery and said zone of pressure of at least about  $5 \text{ sec.}^{-1}$ ,  
C. horizontal displacement in said zone in a longitudinal circulatory manner at a cycle rate such that the actual volume of said liquid moved from one end region of said treating zone to the opposite end region thereof and back within one minute is equivalent to from about one-tenth to 30 times the total volume of said liquid in said zone, such equivalent volume and the horizontal circulation rate for such liquid so moved, respectively, being approximately proportional to said cyclical vertical displacement cycle rate in any given instance, while continuously maintaining substantially the total volume of said liquid in said zone under laminar flow conditions.

2. The process of claim 1 wherein said cyclical vertical displacement in combination with said rolling action produces folding action in said liquid.

3. The process of claim 1 practiced continuously by both continuously introducing a material to be agitated into said zone with a liquid so filling said zone being in said zone and continuously withdrawing so agitated liquid product derived from mixing said material and said liquid together from said zone.

4. The process of claim 1 practiced batch-wise by first so filling said zone with a liquid to be agitated and then so agitating such liquid.

5. The process of claim 1 wherein said zone is cross-sectionally circular and has a longitudinal axis which extends generally horizontally.

6. The process of claim 4 wherein paddle blades rotate in said zone about said longitudinal axis.

7. The process of claim 6 wherein each of said paddle blades is further characterized by having:

A. a first region which both extends:

1. axially uninterruptedly at least about 50 percent along the length of said longitudinal axis, and  
2. radially over said axial blade length uninterruptedly from said longitudinal axis to a location adjacent the perimeter of said zone,

B. a second region which is axially adjacent said first region and adjacent an end of said zone, said second region having discontinuities formed there-through.

8. The process of claim 7 wherein the number of said blades is at least two.

9. The process of claim 1 wherein said liquid has volume average viscosity of from about one-half to 500,000 centipoises.

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10. The process of claim 9 wherein said liquid has a volume average viscosity of at least about 1,000 centipoises.

11. The process of claim 1 wherein said liquid has a volume average viscosity of from about 10,000 to 100,000 centipoises.

12. The process of claim 1 wherein the cycle rate for said cyclical vertical displacement ranges from about 1 to 25 times per minute.

13. The process of claim 12 wherein said bulk average shear rate ranges from about 3 to 100 sec.<sup>-1</sup>.

14. The process of claim 12 wherein said horizontal displacement cycle rate is equivalent to from about one-third to 10 times the total volume per minute.

15. The process of claim 3 wherein the liquid so filling said zone has a volume average viscosity of from

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about 10,000 to 100,000 centipoises and wherein said material so introduced is a liquid miscible with said liquid so filling said zone and having a viscosity of from about one-half to 100 centipoises.

16. The process of claim 1 wherein:

A. said liquid has a volume average viscosity of from about 10,000 to 100,000 centipoises and fills said zone from about 30 to 75 per cent by volume,

B. the cycle rate for said cyclical vertical displacement ranges from about 1 to 25 times per minute,

C. the cycle rate for said cyclical vertical displacement ranges from about 3 to 100, and

D. the horizontal displacement cycle rate is equivalent to about one-third to 10 times the total volume per minute.

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