INTRODUCTION

Drop breakup and the criteria for drop breakup in viscous flows has been the subject of vigorous research since the early defining work of G.I. Taylor (1932, 1934) concerning drop deformation and its effect on emulsion rheology in flow. The preponderance of work in the area has been focussed on drop deformation and breakup in steady, linear flows. In this context "breakup" refers to the nonexistence of a bound drop shape, since experiments generally show drop fragmentation only in the case of very extreme drop elongation or, more commonly, when the flow is abruptly altered. There have been a host of review articles detailing studies of drop deformation and unbound growth in steady linear flows. Most of these articles describe the delineation of flows as "strong" based on their ability to create unbound growth or distortion of the drop, or, in other words, cause the nonexistence of a bound shape. Most of this work also has been at least tacitly described as bearing on drop breakup in the flow through porous media.

In related studies, researchers have developed simple deterministic models for drop breakup in flow through porous media. These include the flow through a straight capillary tube, flow through a contraction, and the flow through a converging diverging tube. This previous research does not include the stochastic element of the disordered flow fields present in most porous media.

There is a growing body of literature, however which suggests that, indeed, drop breakup in time-varying flows may be qualitatively different than the "breakup" witnessed in steady flows. For example, Stone and coworkers have shown that drop fragmentation can be incurred by drops in time varying linear flows (primarily by abruptly altering the flow field) even if the flow field remains "weak" throughout its duration by the criteria established for steady flows. Moreover a recent study of drop breakup in chaotic flows by Tjahjadi and Ottino (1991) demonstrates that fragmentation can be readily induced, primarily by the "end-pinchoff" mechanism found in time dependent linear flows by Stone et al.

Our thesis then in the present research is that there is significant evidence that the breakup of drops in disordered, Lagrangian unsteady flows may very well be more important (i.e. more common) and governed by very different criteria than that governing the unbound growth of drops witnessed in steady elongation, for example. The mechanisms of breakup in disordered, unsteady flows are then only beginning to be examined. Criteria necessary to designate a given disordered flow field as strong, have therefore not been developed. This is the focus of the present research.

NUMERICAL SIMULATIONS

To examine drops and drop breakup mechanisms in disordered flow fields, first, we have completed large scale numerical simulation of drop breakup in disordered, Lagrangian unsteady flows as models for the flow through fixed fiber beds. The flow fields chosen were a class of anisotropic Gaussian flows, which have been shown by Shaqfeh and Koch to be equivalent to the flow through a disordered, dilute fixed fiber bed if one can neglect the near field interactions of fiber and drop. Individual realizations of these flow fields were created spectrally via a large sum of Fourier modes following a modified version of the procedure developed by Kraichnan to simulate model turbulent flows. The drop evolution in these flows was then modeled using small deformation theory for the evolving drop surface. To be specific O(Ca), O(Ca^2), and O(Ca^3) small deformation theory models were used to simulate the evolution of the drop shape as it traversed the porous bed (i.e. anisotropic Gaussian flow field). Note in this context that it has been shown elsewhere, that these models have been very useful and are very accurate for predicting drop breakup in steady linear flows. This is true even though the drop shape and deformation near breakup are not well predicted by these models, since the deformation is very large.

The results of these simulations were very interesting and are contained in a more detailed publication elsewhere. First it was demonstrated that "breakup" (defined within the context of the small deformation theory as a singularity in the deformation parameter with the latter defined originally by Taylor (1934)) did occur in the flow through the model fixed bed. In fact it was found that there was a "critical" value of the pore size Capillary number where 1 out of 2000 drops broke in flow. More complete examination of this critical condition revealed that there were rare events which caused isolated breakup events at smaller values of the critical Capillary. Therefore, a more accurate way to define a critical condition in this flow was to define a very small, standard rate of drop breakup per unit time and then calculate the critical condition based on the ability of the bed to cause drops to break at a faster rate than the chosen small standard. Based on this definition, we found for example that the critical capillary number was approximately 0.16 for a volume fraction of solids \( \phi = 0.025 \) in the bed. This value was determined using the second order small deformation theory, and was somewhat smaller using the third order theory, although as is well known the third order the-
ory does not include all terms at the same order of approximation. We also found that the critical condition was weakly decreasing as the solids volume fraction increased. Note that these values of the critical condition are comparable to those found for isolated drops in pure straining motion.

The most interesting results from these simulations were the predictions about the rates and the mechanisms associated with how the drops broke-up. First, as shown in Fig. 1, the breakup events were isolated and fairly short lived events. The deformation parameter, \( D_0 \), for single drop in a given realization of the stochastic flow, would typically oscillate in a disordered fashion until, in one to three pore lengths (or correlations lengths of the stochastic flow), it would rapidly become unbound, indicating a breakup event.

Most of these events appeared to be twist breakups where the drop would reach a state of deformation that would not allow it to undergo the next deformation that the stochastic flow imposed without fracture. Vorticity was very often associated with breakup. Of note here is that the upon examining the drop distribution of those drops which did not fracture we found that breakup was not favored in any particular region of the probability density of deformation. It seemed to occur with almost equal probability over the whole range of deformations. Thus the distributions became almost Gaussian at large Capillary numbers, cf. Fig. 2.

A second notable characteristic of these breakup events was the local flow field in which they typically occurred. We examined the statistics of the type of linear flow that the drop experienced in the one to two time steps before the breakup event. The summary of this study is presented elsewhere where we plot a histogram of the parameter, \( L \), which has been used by Olbrigtn et al. to characterize steady flows as strong or weak. Values of \( L > 0 \) contain more extensional character than vortical flow and are classified as "strong" since they will break drops at a critical steady flow strength. Those for \( L < 0 \) are alternatively classified as "weak". Our results show that this classification is of little use for the stochastic flow examined here, since drop breakup was noted just as readily in the weak flows as in the strong. In fact, it would appear from our visualizations of the breakup events that the local flow in which the drop breaks is not a useful test for predicting breakup events, since the deformation before the breakup event is as important as the flow in which the drop ultimately breaks. This is a key point to which we shall return in our discussion of ongoing work.

**EXPERIMENTS**

In parallel to the simulation work described in the preceding section, we have developed an ongoing experimental investigation of drop breakup in dilute fixed fiber beds. We have been motivated in this work.
by recent experiments by Vinckier et al.\textsuperscript{16} and Yang et al.\textsuperscript{17} on the shear behavior of PDMS/polyisobutylene emulsions. These researchers created emulsions by vigorously mixing these immiscible liquids at concentrations of PDMS of approximately 1%. At that concentration, coalescence and breakup under shear act to create a steady drop size distribution which was reproducible. Upon creating this drop distribution these researchers used turbidity, dichroism and small angle light scattering measurements to examine the deformation and breakup of drops under steady simple shear. They found that form dichroism measurements were a sensitive measure of the breakup process.

We have chosen to use the same materials for our emulsions, however since our work is initially focused on the breakup of single drops, we have chosen to use a much more dilute emulsion at PDMS concentrations of 0.01%. We have chosen to use the same light scattering and polarimetry techniques as the Vinckier et al.\textsuperscript{16} and Yang et al.\textsuperscript{17} however we have added microscopy of the emulsions, which has allowed us to determine the evolution of the drop size distribution during the flow fields examined.

The emulsions were examined in two flow fields: simple shear and the flow through a dilute fixed bed of fibers. The latter flow was generated in an existing fiber bed that was constructed with known statistics at solids volume fraction of 2.5%. The bed has been used in two previous studies by our group in examinations of particle orientation and polymer stretch in fixed bed flows.\textsuperscript{18,19} In simple shear flow, we reproduced the salient features of the experiments by Vinckier et al.\textsuperscript{16} and Yang et al.\textsuperscript{17}. To be specific, turbidity, small angle light scattering, and microscopy indicated that drop breakup occurred even at low values of the Capillary number (Ca < 0.07) primarily because of breakup of the large drop tail in the distribution. After breaking this tail, the distribution achieved a steady state in the flow at a given capillary number which was characterized by some degree of anisotropy in the SALS and dichroism. We could correlate directly with an average deformation parameter in the flow.

The measurements in the simple shear flow acted to benchmark our results in the flow through fixed beds. These measurements showed remarkable similarities to the shear flow measurements. Breakup from the large drop tail was found to occur at pore size Capillary numbers that were very low (again Ca < 0.07). However in this instance, we used time-dependent measurements to reveal that breakup was nearly uniform throughout the bed and occurred after only one or two pore lengths of flow. This is in agreement with our numerical simulations. Initial measurement of the dichroism in the flowing suspension however, did indicate that it reached a steady state in the flow. This suggests that the deformation of the drop population which has not broken reaches a steady drop size distribution and then breakup events occur which do not significantly distort the distribution. The deformation parameters calculated from the dichroism in the fixed bed flow indicate deformation which may be well in excess of that in simple shear flow (cf. Fig. 3). These deformations were in excess of those calculated from our direct numerical simulations of the process and at present we are working to determine the cause of this discrepancy.

![Figure 3](image)

**Fig. 3** Measured deformations in the flow of a PDMS emulsion through a dilute fixed fiber bed. Shown are also the simulations for fixed bed flow and the theory for shear flow.

**ONGOING WORK**
At present we are focusing on two parts to the research outlined above. First, we are modifying the simulations in a number of important ways. Rather than use the small deformation theories to predict drop breakup we are writing a full, boundary integral code to examine full drop deformation and change in these stochastically fluctuating fields. We feel this is important for two reasons. First, the preponderance of twist breakups in our initial simulations might be an artifact of the small deformation theory since it is well known that the detailed shapes achieved by the drops as calculated from this theory are not accurate near breakup. Thus other important mechanisms of drop breakup may indeed be excluded by this small deformation theory including end pinching and capillary wave instability. Both of these mechanisms were important in the available work on breakup in chaotic flows. This may be part of the reason for the reduced drop deformation parameters in our simulations as compared to the experimental work. A second reason for modifying our simulations is to focus on determining a reasonable criterion to predict breakup in these flows. It is now our feeling that any of the possible two point rate of deformation correlation functions may be useful. The reasons for this are made manifest from our previous work: these correlations contain important information concerning the strain that a drop experiences before entering a new flow region.

The second general area in which we are focusing our energies is in improving our experimental work. At present we are constructing a large fiber bed which will be index-of-refraction matched to the fluid. Thus we will be able to examine single drops as they pass through the bed, and thus witness breakup events as they happen. It is our purpose here to understand whether these twist breakups (or more generally, short time breakup events) are the major breakup mechanism in these disordered flows.

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REFERENCES