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A New Separation Concept Based on a Coupling of Concentration and Flow Nonuniformities

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A New Separation Concept Based on a Coupling of Concentration and Flow Nonuniformities

Described here is a new separation concept which theory shows to have considerable potential for analytical-scale work. Like chromatography, separation is achieved by differential migration in a narrow tube through which fluid is flowing unidirectionally. Unlike chromatography, separation is brought about in a one phase (or at least a continuous phase) system. It should not only be applicable to chromatography-type separations, but, more important, it should exhibit a distinct advantage in separating macromolecules and colloids, because of its essentially one-phase nature.

The present concept can be illustrated by reference to a special case in which thermal diffusion causes enrichment. If we imagine a narrow tube with a fluid mixture inside, the application of a lateral temperature gradient will introduce a unique maldistribution of each component with respect to the gradient axis x. Since viscosity also depends on temperature, flow velocity will also vary along the x axis. Each component will therefore travel at a characteristic velocity whose value will be determined by the particular maldistribution of that component's concentration.

It is instructive to consider the generality of the above concept. First we note that there are many sources of enrichment besides thermal gradients; there are electrical, magnetic, and centrifugal forces, etc., any of which might be applicable in certain cases. Second, one need not depend on a temperature gradient for flow inequalities. Nonuniform velocity profiles accompany all normal flow processes because of viscous and other effects.

The field-flow fractionation method proposed here, like cascade methods in general, considerably multiplies the effect of any of the basic forces which lead to enrichment. Very strong fields can be applied over the small cross sections employed. Simple theory shows that under favorable conditions the final separation is roughly equivalent to that which would be obtained if the strong lateral force were acting directly over the entire tube length. In a coiled or spiral configuration, the tube can be made exceedingly long, limited only by flow resistance. Thus one would hope to get the resolution characteristic of great migration lengths while maintaining reasonable separation speed by virtue of the intense fields.

By way of example, a 1-volt drop across a channel 1 mm in width and 10 m in length should be equivalent to a single-stage electrophoretic separation employing 10,000 volts along a 10-m strip. A 100°C temperature drop would equal a hypothetical drop of 10^{60}C over the same 10-m length. It is immediately apparent that some present limitations can be disposed of. Strong centrifugal fields, which ordinarily act over no more than a few centimeters, could be extended to an effective length of many meters. Nonuniform electrical fields, whose enriching effect is limited by breakdown phenomena, could be, in effect, multiplied many times.

The application of this concept to macromolecules might be especially interesting. These materials are difficult to separate chromatographically because of slow equilibration and a precarious distribution between two phases (even the gel filtration or permeation method has not totally solved the problem). The enriching force of the proposed method, analogous to phase distribution, could be made as gentle or as severe as necessary. If macromolecular enrichment were to be achieved with thermal diffusion, it would be pertinent to note that a temperature drop of only a few degrees suffices to give a significant concentration bias. If one were to apply a total lateral temperature drop of 30 or so °C, most high-molecular-weight components would move almost entirely to the neighborhood of the cold wall. In the steady state, diffuse layers would extend out from the wall a short distance, the precise thickness fixed by molecular weight and other properties. While the variable thickness would lead to differential migration, the over-all thinness of such layers would lead to rapid equilibration, with an attendant rapid separation, and also to a wide spectrum of retention times.

A number of variations can be conceived for this technique. An analogue of programmed-temperature chromatography, in which the temperature gradient (or other force) is gradually diminished, could be used. This would release large molecules, in an order depending on the molecular weight, from the slowly flowing wall region. Other variations in gradient and/or flow are apparent.

One might multiply the effect through an alternating flow; in one flow direction the gradient would be fully applied, and in the opposite direction it would be removed or otherwise altered so as to prevent the loss of resolution already achieved. For slowly diffusing substances (especially macromolecules), it might be desirable to halt the flow near the beginning to allow the achievement of a steady-state distribution.

Other variations might involve controlling the flow pattern through changes in channel geometry or the introduction of granular material; the use of frontal as well as zonal techniques; changes in the composition of the carrier, giving a gradient elution or comparable effect; the use of gases as well as liquids for carriers; and the use of a sorbent packing, thus combining the present method with chromatography. These examples are by no means exhaustive, but they do convey an idea of the possible scope of the proposed method. Work is now in progress concerning the theoretical characteristics and experimental implementation of these concepts.

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