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ACOUSTIC FIELD-FLOW FRACTIONATION

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SUMMARY

Field-flow fractionation (FFF) in a standing acoustic wave field with the use of radiation pressure was studied. With this force field across the channel, values characterizing the effectiveness of the separation were obtained. In this situation the separation process resembles conventional FFF. Particles may either be pressed against the channel wall or focused in the region of maximal flow velocity, depending on the sign of the adiabatic compressibility difference and/or the density of the particles and the medium. Particles may be separated according to density, adiabatic compressibility and diffusion coefficient. Optimal conditions for separation were found and results of preliminary experiments are presented.

INTRODUCTION

The principal ways to exploit field-flow fractionation (FFF) are being widely investigated, both theoretically and experimentally[1–3]. The aim of these investigations is the development of a new means for increasing the effectiveness of the existing variety of methods and the quest for transverse forces that will allow the separation of particles according to new physical parameters.

In this paper we discuss the possibility of particle fractionation with the use of radiation pressure working on the particles scattering an acoustic wave. If the standing acoustic wave is excited across a flat channel, then this force equals[4]

$$F(x) = \frac{\pi}{\lambda} vf \frac{\rho u^2}{2} \sin\left(\frac{4\pi}{\lambda} x\right) \tag{1}$$

where λ is the wavelength, v is the volume of particles of dimensions much less than the acoustic wavelength, u is the ultrasonic oscillating velocity amplitude, x is the cross-sectional coodinate and

$$f = 1 - \frac{\beta^*}{\beta} + \frac{3\,(\rho^*/\rho - 1)}{2\,\rho^*/\rho + 1} \tag{2}$$

where β^* and β are the adiabatic volume compressibilities of a particle and a medium, respectively, and ρ^* and ρ are the particle and medium densities, respectively.

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The space separation of particles across the channel and, consequently, the difference in the linear velocities of their migration in the flow velocity profile formed in the flowing liquid is the basis for their field-flow fractionation in terms of ρ^* and β^*. The most advantageous is the case where the half-width h of the channel, situated between two identical transducers, is equal to $\lambda/8$ (ref. 5).

The flat channel and acoustic transducers form an acoustic resonator in which a plane standing wave is excited. Depending on the number of a harmonic in which a resonator is excited, the pressure nodal plane of the standing acoustic wave or the pressure antinodal plane of that may coincide with the symmetry plane of the channel. Under these conditions the particles with $f > 0$ are concentrated in the channel symmetry lane, where the flow velocity is close to the maximal value, u_0, and the particles with $f < 0$ are focused near the channel walls, or *vice versa*. The primary parameters characterizing the elution curve, that is, the average velocity of particle distribution peak, $\bar{U}$, the hydrodynamic diffusion coefficient, D_{eff}, which defines the root-mean-square width of the peak, and the transversal Peclet number, ε_0, for particles focused near the channel walls are given by the expressions[5,6]

$$\bar{U} = \frac{2\,u_0}{\varepsilon_0} \tag{3}$$

$$D_{\text{eff}} = \frac{8\,u_0 h^2}{D\varepsilon_0^4} \tag{4}$$

and

$$\varepsilon_0 = \frac{\pi v \rho u^2}{16\,kT}\,|f| \tag{5}$$

where D is the particle diffusion coefficient and kT is the thermal energy. At $\rho = 1$ g/cm^3, $v = 10^{-14}$ cm^3, $u_0 = 50$ cm/s, $kT = 10^{-21}$ J (room temperature) and $f = 0.1$, $\varepsilon_0 = 10$. This is in agreement with the typical FFF values of the transversal Peclet number.

For particle focused near the channel symmetry plane, the corresponding equations are[5]

$$\bar{U} = u_0 \cdot \left(1 - \frac{1}{2\,\varepsilon_0}\right) \tag{6}$$

$$D_{\text{eff}} = \frac{u_0^2 h^2}{8\,D\varepsilon_0^3} \tag{7}$$

$$\varepsilon_0 = \frac{\pi^2 v \rho u^2}{64\,kT}\,|f| \tag{8}$$

The separation of particles with $f > 0$ from particles with $f < 0$ must be very effective as a result of the large differences in the average fraction velocity. In this

instance a process of eluting a single fraction requires a time of the same order as the relaxation time, namely $h^2/D\varepsilon_0$. Under the observed conditions and at the channel half-width of 50 μm, this time is about 100 s.

The maximum Peclet number increases with increase in the dimensions of the separated particles proportionally to their volume and for micron sized particles may reach 10^3–10^4.

As the radiation pressure force of ultrasound depends strongly on the channel cross-section coordinate, it is possible to create some other homogeneous field in the channel, *e.g.*, a gravitational field. In this instance, under the effect of transversal gravity and radiation pressure, it is possible to concentrate particles in the planes where the gravity equals the radiation pressure force:

$$\frac{\pi}{\lambda} vf \frac{\rho u^2}{2} \sin\left(\frac{4\pi}{\lambda} x\right) = gV(\rho^* - \rho) \tag{9}$$

As is evident from eqn. 9, the coordinate of the particle concentration plane does not depend on the particle volume and, consequently, the elution time also does not depend on the particle volume, but depends only on the ratio of f to $(\rho^* - \rho)$.

APPARATUS

The first stage of development of the system designed for acoustic field-flow fractionation was the demonstration of the transversal force of the acoustic radiation pressure effect on the velocity of samle mobility in the channel. The experimental set-up is shown schematically in Fig. 1.

The carrier fluid (distilled water) was released from a reservoir supplied with a filter (pore diameter 0.22 μm) into the channel of the separator. The effluent was mixed with 0.9% sodium chloride solution and entered the conductimetric particle counter. The fluid level in the tank containing water exceeded that in the tank containing sodium chloride solution and was maintained almost constant. This ensured a constant fluid flow velocity in the system. The signals from the particle counter were recorded on an X–Y recorder.

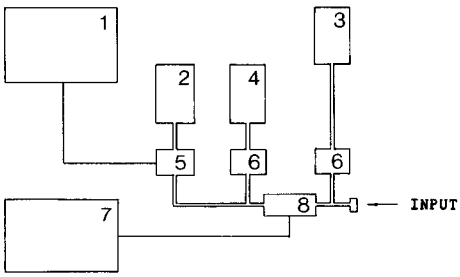

Fig. 1. Experimental design for field-flow fractionation with a transversal force of acoustic radiation pressure. 1 = Recorder; 2 and 3 = carrier fluid tanks; 4 = tank containing electrolyte; 5 = conductimetric detector; 6 = filters; 7 = driving electronics; 8 = acoustic separator.

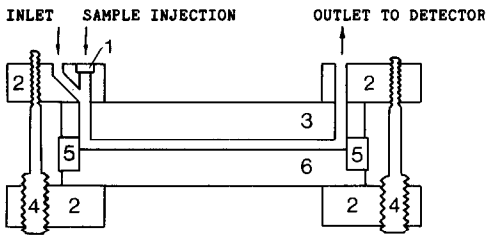

Fig. 2. Design for particle separation in the transversal force field of acoustic radiation pressure. 1 = Plug for sample injection; 2 = brass frame; 3 = matching plate; 4 = adjustment screws; 5 = rubber gaskets; 6 = ultrasonic transducer.

The separator itself is shown in Fig. 2. A flat channel measuring 10 × 30 × 0.4 mm was forced by the surface of the ceramic ultrasonic transducer and the matching glass late with a rubber gasket fixed aroud its edges. Carrier fluid was injected and released from the channel through the inclined openings in the matching plate. A sample was introduced into the channel by a microsyringe through the rubber diaphragm. The matching plate and the acoustic transducer were fixed in a massive frame. The necessary homogeneity of the channel cross-section was adjusted with he help of four screws and was controlled acoustically by the purity of the mode of acoustic oscillations of the channel.

RESULTS AND DISCUSSION

The registration techniquc used allows operation only in the micrometre particle range and hence with diffusion coefficients of 10^{-9}–10^{-8} cm^2/s. Transversal Peclet numbers of 10^3–10^4 are necessary for obtaining an acceptable time for the establishment of an equilibrium concentration distribution of particles across the channel. For particle dimensions of 3.8 μm, as used in this experiment, the transversal Peclet number in the natural gravity field is $2 \cdot 10^3$. Therefore, in the absence of a radiation pressure field, these particles sediment at the bottom of the horizontal channel. Unfortunately, the time to establish an equilibrium concentration distribution in the natural gravity force field is of the order of 10^3 s, although in a stronger field it may be considerably reduced.

The matching plate used allowed the adjustment of the antinodal points of the oscillation velocity on the upper wall of the channel and of the points of maximum radiation pressure at a distance of 0.9 times the channel width from its lower wall.

The experiments were carried out the following way: 1 min after sample injection, the flow of carrier fluid in the channel was stopped and then resumed with a maximal velocity of 0.1 mm/s with in 10 min. The dependences of the number of particles moving through the detector, counted per unit time (10 s) at different transducer voltages, are presented in Fig. 3. It can be seen that at low voltage the sample particles adhere to the channel walls under the influence of gravity. Then a small peak, representing the non-adherent particles under the influence of radiation pressure (curve 1 in Fig. 3), was observed. The other peaks in Fig. 3 correspond to the particles focussing in different channel planes, and their location is determined by eqn.

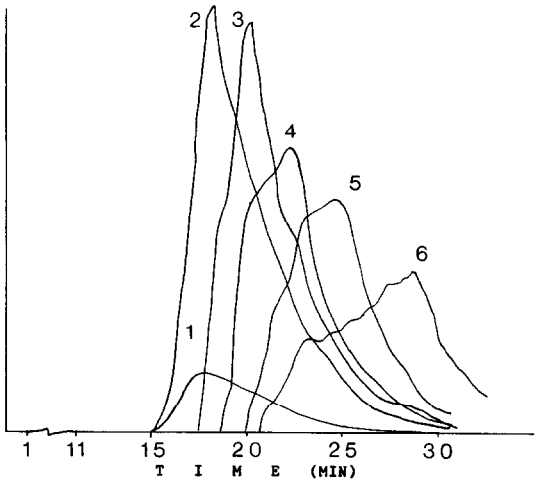

Fig. 3. Acoustic field-flow chromatograms for a sample containing polyacrylic latex particles at different ultrasonic transducer driving voltage amplitudes. Curves: 1, $V_d = 2.50\ V_{rms}$; 2, $V_d = 3.54\ V_{rms}$; 3, $V_d = 4.33\ V_{rms}$; 4, $V_d = 5.00\ V_{rms}$; 5, $V_d = 5.59\ V_{rms}$; 6, $V_d = 7.07\ V_{rms}$.

9. At high transversal Peclet numbers, the time of particle output from the channel is defined, in agreement with eqn. 9, by the expression

$$\tau = \frac{L}{u_0}\left\{1 - \left[\frac{2.5}{\pi}\arcsin\left(\frac{6.25}{V_d^2}\right)\right]^2\right\}^{-1} \tag{10}$$

where L is the channel length and V_d is the transducer driving voltage amplitude. The time interval defined by eqn. 10 is in satisfactory agreement with the time of peak output at different transducer voltages in Fig. 3.

Although these experiments are only preliminary, we believe that they demonstrate convincingly an opportunity for particle separation in an acoustic pressure force field. Further development of the suggested technique is possible at increased resonator operating frequencies. Therefore, by preserving the magnitude of the Peclet number, the channel width could be decreased and as a result the time of transversal equilibrium establishment and velocity of the carrier fluid could be decreased, thus substantially improving the resolving power of the fractionation. The force of acoustic pressure allows particles to be separated by FFF according to their density and adiabatic volume compressibility. It is unlikely that acoustic FFF could compete with various forms of centrifugation or with sedimentation flotation flow fractionation, as the force obtained by centrifugation is greater than the force of radiation pressure. Particle separation according to volume compressibility seems to be unique and may provide new information about the structures of the separated particles.

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