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INVESTIGATION OF THE FLUID DYNAMICS OF GAS FLOW IN LARGE-DIAMETER COLUMNS

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SUMMARY

The velocity field of a gas flow immediately above the packing surface of a chromatographic column loaded and compacted by various methods has been investigated using a laser anemometer.

INTRODUCTION

The reduced efficiency of large-diameter columns is usually attributed to the appearance of the velocity profile of the carrier gas across the section of the column[1], although the experimental data concerning this profile are contradictory. Early investigations indicated an increase in velocity from the centre to the walls of the column[2,3], but later work[4–6] showed the opposite. It should, however, be noted that in the most recent work it was not the carrier gas profile that was measured, but that of the sorbing component, and the two do not necessarily coincide.

We have already suggested[7] that the decrease in efficiency with increasing column diameter may be caused by local inhomogeneities in the packing structure, resulting in irregular changes of the carrier gas velocity. Radushkevich[8] was the first to note this. Such inhomogeneities evidently explain the increase in term A of the Van Deemter equation mentioned in ref. 9. In all the above-mentioned studies the nature of the velocity field was investigated purely qualitatively. The methods used did not permit the estimation of the local velocity values because the velocity was averaged over a considerable cross-sectional area. In order to determine the nature of the velocity profile and also to verify the local changes in velocity, we have measured the carrier gas velocity immediately above the sorbent layer in a column of 140 mm I.D. Use was made of a laser anemometer, which enabled us to determine the absolute value of the linear local velocity (U) in a region with dimensions of *ca.* 0.5 mm. Our measurements confirmed that some regular changes in velocity indeed occurred, their nature strongly depending on the method used to compact the packing in the column, and evidently also on the technique used to introduce the gas into the sorbent layer. Along with such regular velocity changes, which can, in

principle, be described by some function $U = f(r)$, where r is the distance from the centre of the column, quite considerable local velocity changes of a statistical nature occur, which are evidently due to aggregation of packing particles.

EXPERIMENTAL

Investigations of the velocity field in a chromatographic column filled with a granular packing with a particle size of 0.5–1 mm were carried out with the aid of a laser anemometer. The general view of the set-up is depicted in Fig. 1. It consists of two parts: an optical part, whose operating principle is based on the Doppler effect, and a gas part built on the chromatographic principle, the only difference being that aerosol particles are introduced into the gas flow to create an optical inhomogeneity of the gas flow. A laser beam (1) is split, with the aid of a light-dividing plate (2) and a mirror (3), into two parallel beams, which, after being refracted by a lens (4), intersect at the point of investigation of the gas flow, which is 1.0–1.5 mm from the sorbent surface (5). The lens (4), with a focal length, f, of 15 cm, moves in a horizontal plane, thereby permitting measurement of the flow velocity at any point of the cross-section of the column (6). The cylindrical column, made of molybdenum glass, has an internal diameter of 140 mm and walls 2.5 mm thick; it is 90 cm long with two spherical lids at the ends. The upper lid is detachable. The gas is introduced from below through a central pipe (diameter 6 mm) that fans out towards the column, forming a funnel filled with glass wool, which prevents the packing from spilling. The gas enters the column after passing through a nozzle-type aerosol generator (7), whose inlet pressure is controlled by a standard manometer (8). In our experiments the pressure varied from 0.66 to 0.73 kgf/cm^2, depending on the method used to fill the column. The aerosol generator is filled with an ester, such as dibutylphthalate, which is dispersed by the gas delivered from a cylinder or a blower (9), to the polydisperse aerosol, which is entrained by the gas flow and enters

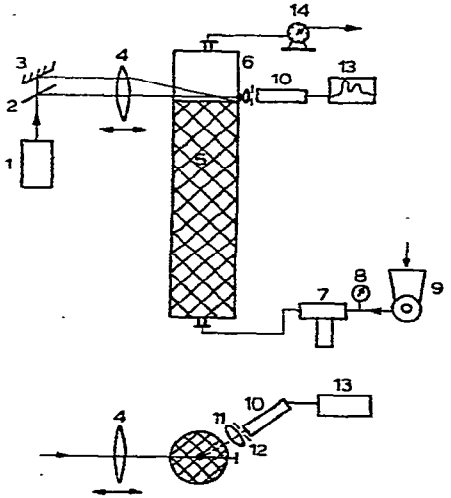

Fig. 1. Diagram of the set-up for measuring velocity profiles. 1, Laser; 2, light-dividing plate; 3, mirror; 4, lens; 5, sorbent; 6, column; 7, aerosol generator; 8, standard manometer; 9, blower; 10, photodetector; 11, objective lens; 12, diaphragm; 13, spectrum analyser; 14, flow-rate meter.

the column. The column is filled with a solid support, Spherochrom-1, grain size 0.5–1.0 mm, specific surface area 1.06 m^2/g, bulk weight 0.43 g/cm^2, porosity 0.65 cm^3/g, and effective pore radius 1.0 μm.

The motion of the aerosol particles, which have passed through the packing layer, causes, in scattered light, a Doppler shift of the frequency relative to the incident radiation. As a result of the photodisplacement of the scattered radiation from the two incident beams, a current pulsation at a frequency of f_D arises in the photodetector (10), related with the gas flow velocity, U, by the equation

$$f_D = \frac{1}{2\pi} \vec{K}\vec{U}$$

where K is the vector of sensitivity of the laser anemometer, which lies in the plane of the incident beams and is normal to the bisector of the angle of intersection, α, of these beams.

$$\vec{K} = \frac{4\pi}{\lambda} \sin\frac{\alpha}{2}$$

(λ —wavelength for He–Ar laser; $\lambda = 0.63 \cdot 10^{-6}$ m).

Because α is small (5–6° in our experiments), $\vec{K}$ practically coincides with the axis component of the linear velocity U.

The objective lens (11) and the diaphragm (12) form a space filter, so that only the light scattered directly in the region of intersection of the probing beams reaches the surface of the photocathode. From the output (10), the signal arrives at a panoramic spectrum analyser (13) C 4-12, which displays the spectrum on a screen. The velocity was estimated by the method described in ref. 10. The signals had a frequency of the order of 2000 Hz, which corresponds to a linear velocity of *ca.* 1 cm/sec. For a chromatographic column of 140 mm I.D. this linear velocity is achieved at a gas flow-rate of 13 l/min. The gas flow through the column is controlled by a gas clock (14).

Our experiments showed that when aerosols pass over a support layer with thickness of 0.5 m, there are still enough aerosol particles for measurements to be made. Because of the laminar gas flow in the support and the average aerosol particle size of *ca.* 0.5 μm, the proportion of particles caught is low.

We investigated the velocity distribution across the column using three different methods of filling: (i) free loading through a funnel; (ii) loading in four equal portions with twenty compactions of each portion and fifty compactions of the filled column by tapping it against the floor; (iii) loading the column in portions of 600 ml and compacting each portion with a plunger with simultaneous pumping of the column with a vacuum pump. In all the three cases the height of the sorbent layer was 50 cm. The velocity field was measured along a single diametral line at intervals of 5 mm. On a small section the velocity profile was taken at 1 mm intervals. After filling the column by method (iii), the entire velocity field was measured at 1 mm intervals. A second loading of the column by this method showed that, on the whole, the velocity field was reproduced fairly well. The velocity fields for each of the above methods is given in Figs. 2, 3, and 4. In order to determine the regular velocity profile

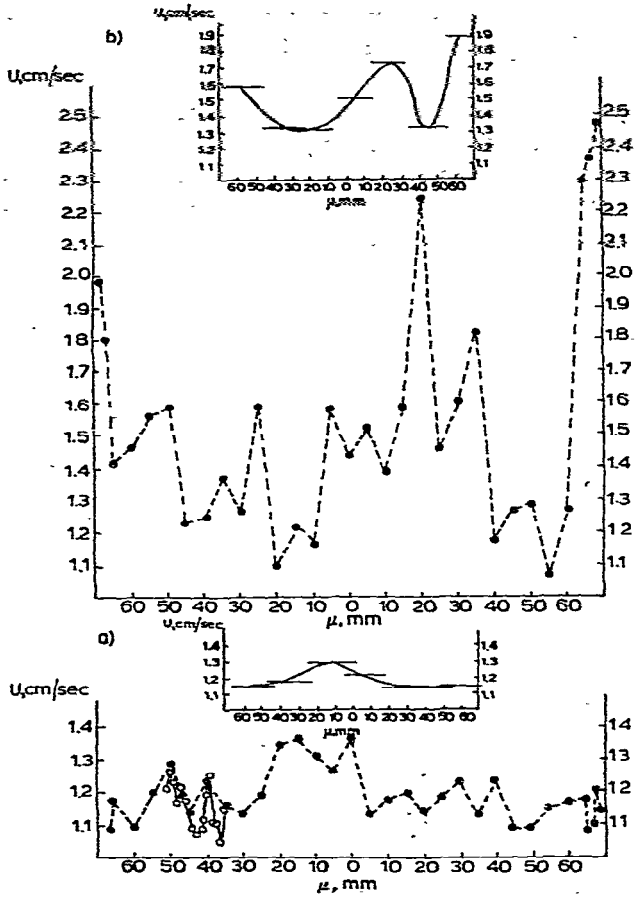

Fig. 2. Velocity field after free loading: (a) before tapping the walls of the column, (b) after tapping the walls of the column.

against the background of the local fluctuations, we calculated the average velocity on 20 mm sections of the diametral line. The average values obtained are denoted by the dashed lines in the Figures. The midpoints of these segments are joined with smooth lines, which show the nature of the regular velocity profile. The accuracy of velocity measurement at each point is $\pm 5\%$. Simultaneously with velocity field measurements, we determined the column efficiency with the same filling methods. The HETP was determined on a metal column, 1 m $\times$ 140 mm I.D., filled with 20% dinonylphthalate on Spherochrom-1, grain size 0.5–1 mm. The column was at room temperature, the sorbate *n*-pentane, the dose volume 1 ml, the carrier gas nitrogen, and the detector of the heat-conductivity type.

In order to determine the mechanical stability of the sorbent layer in the column and the nature of the disturbance resulting from tapping the column walls, we compared the column efficiency after tapping along the entire column length on four sides, the total number of impacts being about 80. The efficiency data obtained are listed in Table I.

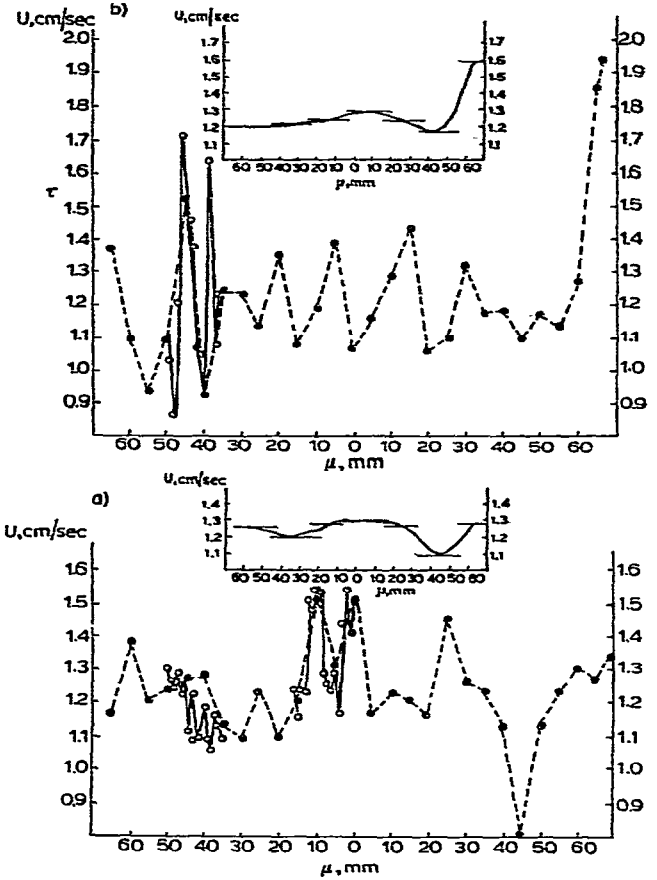

Fig. 3. Velocity field after loading in portions and compaction by tapping column on the floor; (a) before tapping the walls of the column, (b) after tapping the walls of the column.

RESULTS AND DISCUSSION

As can be seen from Fig. 2, with free packing of the column the velocity profile is very irregular; the local velocity fluctuations are also comparatively small. The slight velocity increase in the centre of the column may be attributed to the method of gas delivery. Because the gas is fed into the centre of the column, the hydraulic resistance along the axis is minimal, which may cause some velocity increase in the central part of the column cross-section. When a disk of diameter 70 mm was placed over the inlet hole, an increased gas velocity was observed near the column walls. The gas delivery technique evidently affects the velocity profile, but in order to discover exactly how experiments with a column equipped with a distributing grid need to be carried out.

The virtual non-existence of a profile and the small value of local fluctuations agree with the HETP value. For free packing this value was 4.2 mm, and was close to the values obtained using the other filling methods.

Column compaction by tapping the column walls resulted in a sharp inho-

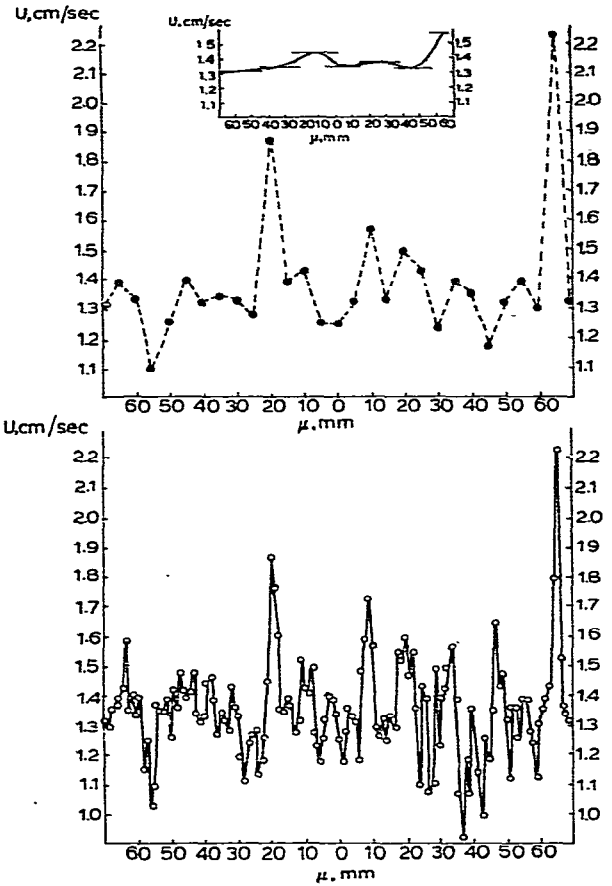

Fig. 4. Velocity field after loading followed by compaction with a plunger and simultaneous vacuum pumping.

mogeneity of the velocity field. The velocity in the region of the column walls increased abruptly, particularly on the side where the tapping was most intensive. Tapping evidently loosens the layer of sorbent next to the wall, and the hydraulic resistance in this part of the column therefore drops sharply. Besides, after the compaction of the packing, the local inhomogeneities are greatly increased.

This abrupt increase in the inhomogeneity of the packing structure raised the

TABLE I

COMPARISON OF THE EFFICIENCY OF COLUMNS FILLED BY DIFFERENT METHODS

Filling method	*HETP (mm)*	*Packing density (g/cm³)*
Free loading	4.2 (12.2*)	0.50 (0.535*)
Loading by tapping column against floor	3.9 (8.9*)	0.56 (0.57*)
Loading with compaction by plunger and vacuum pumping	4.0 (6.0*)	0.55 (0.57*)

* After tapping on the walls of the column.

HETP to 12.2 mm. Thus, compaction of the packing by tapping the column walls cannot be recommended for the preparation of effective large-diameter columns. We normally compacted the packing in columns of diameter up to 150 mm by tapping the column against the floor. It can be seen from Fig. 3 that in this case, a rather complicated velocity profile is formed. The gas velocity increases both in the centre and near the walls of the column. In the intermediate portions of the cross-section the gas velocity is lower. As has already been mentioned, the increased velocity in the centre of the column may be associated with the method of gas delivery. The velocity profile as a whole will then be concave along the direction of the flow. With this method considerable local velocity fluctuations are also observed. Despite the regular velocity profile and local velocity fluctuations, the HETP is no higher with this method of filling than with simple loading. This may be associated with the higher filling density, which must reduce the external-diffusion resistance to mass exchange.

The efficiency of a column filled by the above method is sharply reduced if the packing is additionally compacted by tapping the column wall. An investigation of the velocity field shows that after such tapping the gas velocity near the column walls abruptly increases. Thus, any impacts on the column walls adversely affect its efficiency. As it is difficult to compact the packing by tapping the column against the floor if the column diameter exceeds 150 mm, we tested the method of compaction with the aid of vacuum pumping and a plunger. In contrast to Frisone[11], we obtained a good column efficiency (HETP = 4.0 mm) and a considerably better reproducibility of the parameters of the granular layer than with the other methods. We prepared a sufficiently effective column of diameter 200 mm. An investigation of the velocity field above a packing compacted in this way revealed considerable local density fluctuations. The velocity profile obtained was practically flat, except for a slight increase in the centre of the column (a possible cause of this is mentioned above). The velocity increase at some points near the wall is probably due to loose contact between the plunger and the packing surface near the column walls.

CONCLUSIONS

1 With any of the investigated methods of packing compaction considerable local fluctuations in gas velocity across the column take place.

2 Compaction of the packing by tapping the column against the floor yields a regular velocity profile. The nature of this profile can be judged only after the column is equipped with a gas distributor at the inlet.

3 Tapping the column wall sharply increases the gas velocity near the wall.

4 When the packing is compacted with a plunger in combination with vacuum pumping, practically no regular velocity profile is observed. Despite the considerable velocity fluctuations, the efficiency of the sorbent layer is sufficiently high.

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