Ermittlung des Verlaufs der Bunttongleichheit auch im buntkräftigeren Bereich angewiesen, wobei ein spektraler Farbintegrator mit doppeltem Infeldstrahl die gesuchte Lösung bringen könnte.

Einfluss des Umfeldes. Für die bisher erwähnten Messungen war das Umfeld gleichhell mit dem Infeld, wenn am Orte des Spektrums keine Blende angebracht war (weisses Infeld). Es wurden dann weitere Messungen bei verschiedenen Umfeldhelligkeiten durchgeführt. Es ist zu erwarten, dass eine Farbe niedrigerer Helligkeit bei dunklerem Umfeld als buntkräftigst ausgewählt wird, z.B. wurden für die grüne Optimalfarbreihe (λ_d = 515,9 nm) von 4 Personen die Maximalfarben für die Umfeldhelligkeiten U = 0%, 20%, 100%, 200% (entsprechend den MUNSELL-Stufen 0, 5, 10, 12,8) ermittelt und unter Berücksichtigung der Fehlerbreite in Figur 5 im V_Y, ($V_A - V_C$)-System wiedergegeben. Dieser Versuch bekräftigt damit unsere Erwartung. Weitere orientierende Messungen an den übrigen 24 Blenden verliefen im allgemeinen ähnlich, doch liegen nicht unerhebliche Unterschiede im Ausmass des Verschiebungseffektes vor. Rein qualitativ besteht nicht nur ein Einfluss des Umfeldes auf die Helligkeit des Infeldes, sondern auch auf das Farbmerkmal «buntkräftigst».

Prinzipiell lässt sich sagen, dass beide Funktionen V_{M_r} und $V_A - V_C$ zur Bestimmung der Eigenschaft «buntkräftigst» brauchbar sind. V_{M_r} besitzt den Vorteil, ein vom Buntton weitgehend unabhängiges Mass für die Buntkraft zu liefern[21], während $V_A - V_C$ durch seine Einfachheit beeindruckt. Die Berechtigung für unsere Verwendung von $V_A - V_C$ bei der Rollblendenabstufung folgt aus der annähernden Proportionalität zwischen $V_A - V_C$ und V_{M_r} bei gleichem mischmetrischem Buntton.

Die obigen Messungen tragen einen mehr vorläufigen Charakter, da die Unvollkommenheit der Apparatur Anlass zu mehr oder weniger grossen Fehlern gab. Der Ausbau der Versuche unter Verwendung einer Xenonhochdrucklampe mit tageslichtähnlicher Spektralverteilung und einem verbesserten Aufbau des spektralen Farbintegrators ist im Gange[22].

Summary. In experiments with a sufficiently great number of observers, using our spectral colour integrator, it has been shown that *one* colour containing a maximum of chromatic power (chroma) can be chosen out of a series of optimal colours continuously changeable from white over the fullcolour to black. The determination of such maximal colours, differing in hue, shows their position between fullcolour and spectral colour in the mixing metric colour solid. Their relation to maximal properties of scale-metrically defined chromatic power functions is discussed.

K.-D. HOFMANN[23] und P. WEISENHORN

Laboratorium für Farbenmetrik, Physikalisches Institut der Universität Basel (Schweiz), 22. Juni 1962.

[21] K. MIESCHER, K.-D. HOFMANN, P. WEISENHORN und M. FRÜH, Die Farbe *10*, 115 (1961).

[22] Für die Ausführung der Zeichnungen und Berechnungen danken wir Frau M. FRÜH. – Für die Unterstützung dieser Arbeit dankt der eine von uns (K.-D. HOFMANN) der CIBA Aktiengesellschaft, der andere (P. W.) dem Fonds zur Förderung der wissenschaftlichen Forschung in Bern.

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The Increase of Solute Velocity in the Capillary and Porous Flow of Solutions

It has recently been shown[1-4] that when particles are suspended in a fluid in laminar cylindrical flow, they are displaced laterally, because of the uneven shear to which they are subjected, in the direction of a decreasing velocity gradient, that is, toward the center of the tube. Since the fluid velocity on the axis is twice the mean velocity, this lateral displacement must result in an increase of the mean longitudinal velocity of the solute in relation to the mean velocity of the solvent. The experiments reported here were made in an attempt to investigate this point.

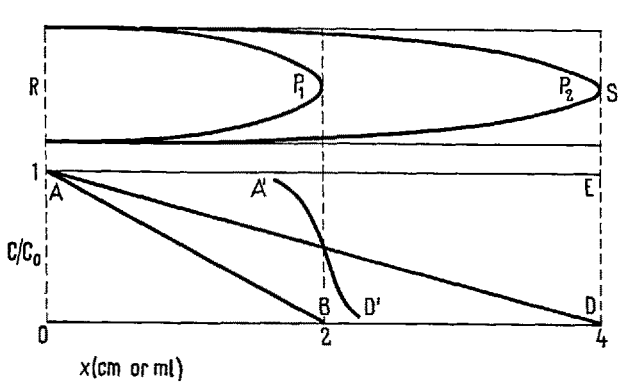

Fig. 1. Laminar flow in a tube.

Capillary Flow. Since a continuous flow of solution in a tube would not lend itself conveniently to such a study, we have alternated solvent and solution and observed the phenomena taking place at the boundaries. The experimental procedure may be described under two headings: (1) Solution follows solvent. The situation may be called a single boundary system. (2) A small sample of solution is introduced between two sections of solvent. This is a two boundary system.

The conditions under which the present work was done may be visualized with the help of Figure 1. R–S is a cylindrical pipe, 4 cm in length and 4 ml in capacity, connected to a reservoir at R and open at S. We assume that convection in the tube is due to laminar flow alone and first disregard the effect of diffusion.

Case I: At the start, the reservoir is filled with solution of concentration C_0 and the pipe with water. If 1 ml of solution flows into the pipe, 1 ml of water flows out at S; the solution penetrates into the water in the form of a paraboloid P_1, and the distribution of the solute along the pipe is given by the straight line AB, its amount by the area AOB. Introduction of a second ml of solution causes the solution to occupy volume P_2, its amount is now area AOD.

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Case II: After introduction of 1 ml of solution, the solution in R is replaced by water, and 1 ml of the latter allowed to flow in. The solution now occupies volume P_2–P_1, its amount is ABD.

As more solution (Case I) or more water (Case II) is pushed into the tube, the solute concentration of the samples collected at S is easily calculated with the help of this diagram.

Molecular diffusion completely alters the picture, as shown by TAYLOR[5]. In Case I (i.e., with only one boundary) the straight line AD is changed into a distribution curve A'D' centered on a point (coordinates $x = OD/2$, $C/C_0 = 0.5$) which moves forward at the mean velocity of the fluid; in other words, the boundary becomes much steeper. The shape of A'D' will, of course, depend on the diffusion constant of the solute and on the time of flow. In Case II (i.e., with two boundaries), provided the solution specimen is small and the distance travelled long enough, diffusion will transform the triangle ABD into the narrow and high profile of a normal distribution curve whose longitudinal displacement is assumed to proceed at the mean velocity of flow of the liquid.

Experimental. Case I: The results in Figures 2 and 3 were obtained with a plastic tube 1.00 ml in capacity, 261 cm in length, 0.70 mm in mean internal diameter. It was fitted at one end to a four-way stopcock connected to one hypodermic syringe provided with its plunger and to another syringe without its plunger which served as a reservoir whose height could be adjusted. (For fast flow, the solution was propelled by hand with the help of a second plunger.) The tube was first filled with solvent from the first syringe, then, by turning the stopcock, connected to the reservoir containing the solution under study. The specimens were collected dropwise at the open end of the pipe into tared test tubes, weighed, made up to the proper volume with water, and analyzed colorimetrically. The chemical procedures were as follows:

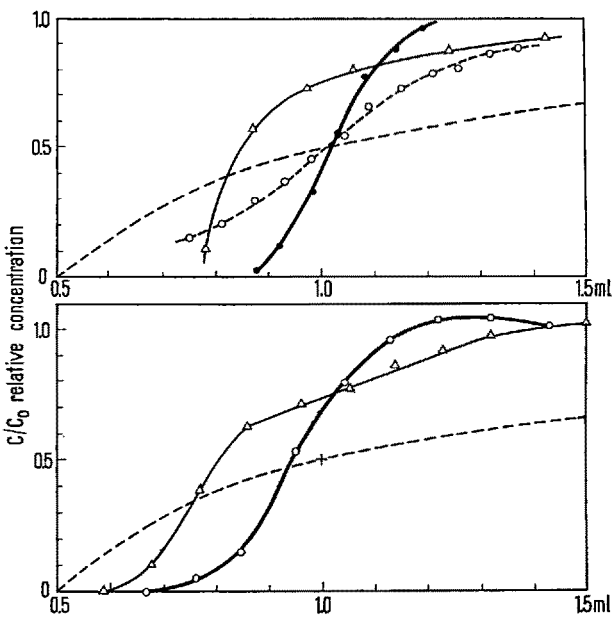

Fig. 2. Single boundary system. Triangles: 16% red cell suspension, velocity 3.3 cm sec^{-1}. Circles: 0.025 *M* KSCN, 9.9 cm sec^{-1}. Dots: 0.025 *M* KSCN, 0.28 cm sec^{-1}.

Fig. 3. Single boundary system. Triangles: 0.5% pseudoglobulin solution, 3.3 cm sec^{-1}. Circles: Same, 0.11 cm sec^{-1}.

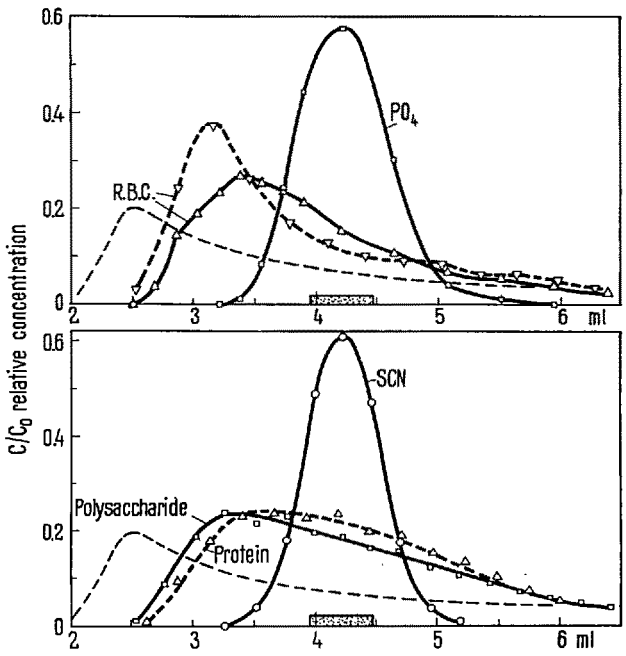

Fig. 4. Double boundary system. A (solid lines): 0.5 ml specimen containing red cells (4%) and KH_2PO_4 (0.005 *M*) in 0.14 *M* NaCl. B (broken line): 0.5 ml of 4% red cells in 30% sucrose. Velocity 3.2 cm sec^{-1}.

Fig. 5. Double boundary system. A (solid lines): 0.5 ml specimen containing a tuberculin high molecular weight polysaccharide (1%) and KSCN (0.03 *M*). B (broken line): 0.5 ml of 1% pseudoglobulin. Velocity 3.2 cm sec^{-1}.

Red cells: laking with dilute NH_4OH. Phosphate: FISKE and SUBBAROW[6]. Thiocyanate: addition of 0.2 volume of 5% $Fe(NO_3)_3$ in 2.5% HNO_3. Carbohydrate: H_2SO_4 and carbazole. Protein: LOWRY et al.[7]. In Figure 2, the abscissae give the position of the specimens in terms of ml collected at the end of the tube. For example, at $x = 1$, 1 ml had been collected, corresponding to a mean displacement of water of 261 cm, the whole length of the tube. The theoretical profile expected of a solution specimen obeying Poiseuille's law alone is given by the broken line.

In the experiment with red cells, the tube was first filled with 0.14 *M* NaCl, which may be called the 'solvent'; the 'solution' was a 16% suspension of sheep red blood cells in 0.14 *M* NaCl. The red cell curve in Figure 2 is remarkable because it is some 40% higher than expected, a difference that would decrease slowly as the graph was extended to the right. Lower concentrations of red cells gave curves in the same position, which always fell short of the theoretical origin at $x = 0.5$. This would happen if the red cells assumed an annular distribution, as observed with plastic spheres[3].

The experiments with KSCN conform to expectation. They yield sigmoid curves centered close to $x = 1.0$, $C/C_0 = 0.5$, the steeper curves being given by the longer time (because of experimental conditions, it is obvious that these two curves cannot be exactly symmetrical).

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Figure 3 illustrates the behavior of a protein (pepsin-treated diphtheria antitoxin) of molecular weight 100000 in water. In fast flow, its profile was similar to that of red cells; in slow flow, which allowed sufficient diffusion to take place, it approached the intersection $x = 1.0$, $C/C_0 = 0.5$ (marked by a cross).

The diffusion constants of the three substances (red cells, protein, thiocyanate) studied so far are roughly (in $cm^2\ sec^{-1}$) zero, 5×10^{-7} and 200×10^{-7}, respectively. The results thus characterize clearly the behavior of these solutes according to their diffusion constants. They show that when diffusion is negligible, the solute accumulates at the front in marked excess of the theoretical curve, which is precisely the phenomenon we were looking for.

Case II: The tube was 1090 cm in length, 3.96 ml in capacity, 0.68 mm in internal diameter. The experimental arrangement described above was slightly modified to permit the introduction of a 0.5 ml sample of solution between two sections of solvent. The first thing revealed by Figures 4 and 5 is that low molecular weight solutes travel at the mean velocity of the solvent. To visualize the fact, a shaded bar on the abscissae gives the position of the specimen if it had travelled along the tube like a solid plug: the apex of the distribution curve is exactly in the middle, at $x = 4.21$.

The theoretical profile (thin broken line) is different from Case I, and again the solutes of high molecular weight do not appear quite as soon as expected. There is again an excess of these solutes: in Figure 5, for example, the area under the polysaccharide up to $x = 6$ represents practically the total amount introduced, whereas the theoretical area up to this point is only 65% of the total, the other 35% extending infinitely far to the right. Since diffusion was negligible, this concentration of the solute must have been entirely hydrodynamic. The displacement of red cells (Figure 4) was further increased by increasing the relative viscosity of the medium to 3.2 with sucrose (the 'solvent' was then 30% sucrose, 0.14 *M* in NaCl; the 'solution', the same fluid containing 4% red cells).

A remarkable separation of components is achieved by this procedure. Note that in Figure 5, for example, the sample withdrawn at $x = 3.25$ contains the highest concentration of polysaccharide and no salt. This fact should be kept in mind as a serious source of error in manipulations that involve the passage of small samples of mixed solutions through fine tubes Also, it could be the basis of a new method for the separation of solutes of different molecular weights.

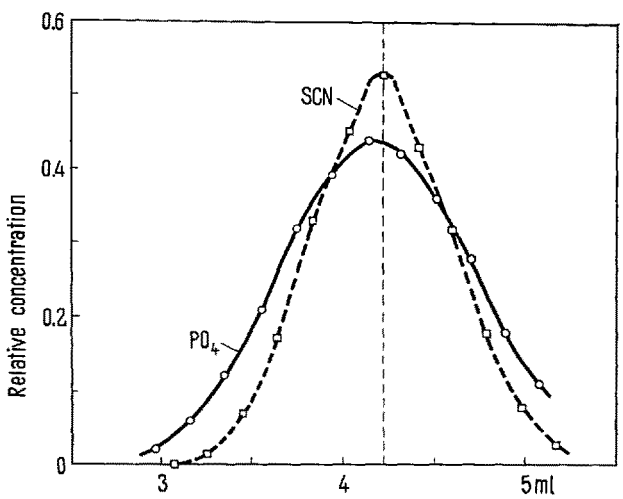

Fig. 6. Double boundary system. 0.5 ml specimen, 0.01 *M* in KH_2PO_4, 0.03 *M* in KSCN, in 30% sucrose. Velocity 3.2 cm sec^{-1}.

When mixtures of low molecular weight solutes in water were studied in this manner, no clear separation could be seen. If the relative viscosity was raised to about 3.2 by addition of glycerine (40%) or sucrose (30%) as with red cells, a slight separation became evident. In Figure 6, the apex of the SCN curve is exactly at the point corresponding to the mean velocity of the water ($x = 4.21$), whereas the phosphate curve is flatter and slightly displaced to the left, as would be expected of a salt with a lower diffusion constant. (The value of *D* for these two salts does not seem to be available in the literature.)

Porous Flow. The idea of extending this work to porous flow originated in the following observation. If a dilute solution of hemoglobin is allowed to ascend a dry strip of fairly dense filter paper, the advancing front soon turns a much deeper red than the space behind. This frontal accumulation of solute is not limited to proteins and takes place with practically any substance, as can be shown by cutting the paper afterwards in thin horizontal strips and analyzing the contents. The phenomenon obviously accounts for a trivial fact, the dark rim so often shown by dirt spots on clothing.

Since the rate of flow in the porous materials used here was infinitely slow in comparison with flow in the tubes, one would expect all curves obtained to have the appearance of distribution curves, and the point of interest would be whether the apex of these curves moved at the mean velocity of flow of the liquid, or whether it moved faster, as the above results would make one expect. It is assumed here that flow in a porous body obeys Poiseuille's law. This view is supported by the outstanding success of the filtration technique in the determination of the particle size of viruses, confirmed by other methods, in which the 'average pore diameter' of a filter is established by application of Poiseuille's law. For the materials used here, this value was about 2μ. The critical Reynolds number in porous flow is said to be between 75 and 0.1[8]. A rough estimate gives here a value of less than 10^{-4}.

Experimental. The arrangements were essentially those of descending paper chromatography. The shape of the trough and the manner in which the upper edge of the sheet was held in place insured quantitative adsorption of the specimens without loss. The bottom edge was cut to a point to permit the dropwise collection of the effluent into tared test tubes. The apparatus was enclosed in a small air-tight plexiglas box and precautions taken to avoid evaporation during the introduction or collection of samples.

Several porous materials were tried, in the form of sheets approximately 13 × 13 cm: cellulose papers, glass papers, fine nylon, thick glass cloth. The three materials most used had the following characteristics (in round numbers):

	Whatman No. 4	Whatman No. 50	Glass cloth
Imbibition (mm^3/cm^2)	20	10	18
Output (ml/h)	2.2	0.1	0.6
Flow rate (cm/h)	8	0.8	3

[8] A. E. SCHEIDEGGER, *The Physics of Flow through Porous Media* (University of Toronto Press, Toronto 1957), p. 123.

The experiments were made in two ways, the first to study the frontal accumulation of solute, the second to duplicate the tube experiments described as Case II. (1) Starting with a dry assembly, a 1 ml specimen of solution was placed in the trough and allowed to be completely adsorbed; it was then followed by water added at a continuous rate. (2) The trough first received a definite amount of water, slightly less than the maximum which the porous material could hold by imbibition. (In Figure 7B, this amount was 1.5 ml, in Figure 8, 3.5 ml; it corresponded to the 3.96 ml carried by the capillary tube.) This water was followed by a 1 ml specimen of solution, then by water continuously. As a visual aid, the boundaries that the specimen would have occupied in the total absence of friction or diffusion are shown by a rectangle. Figure 7A shows the enormous frontal accumulation that can take place with phosphate on cellulose paper. On the same medium, this phenomenon was observed with sucrose and protein as well as with all the electrolytes tested and was least with thiocyanate. On glass cloth, frontal accumulation was shown by sucrose and thiocyanate, but not clearly by phosphate.

In the second instance, when the specimen was preceded by water, there was a marked relative forward displacement of the salt (Figure 7B). This could be numerically estimated as the quotient of the value on the abscissae marking the middle of the rectangle to that corresponding to the apex of the phosphate curve. In Figure 7B, $2 : 1.4 = 1.4$ approximately. The relative displacement was in general more marked the more dilute the solution. With Whatman paper No. 4, the displacement varied from 1.4 to 1.1 for KH_2PO_4 solutions of 0.001 to 0.1 molarity.

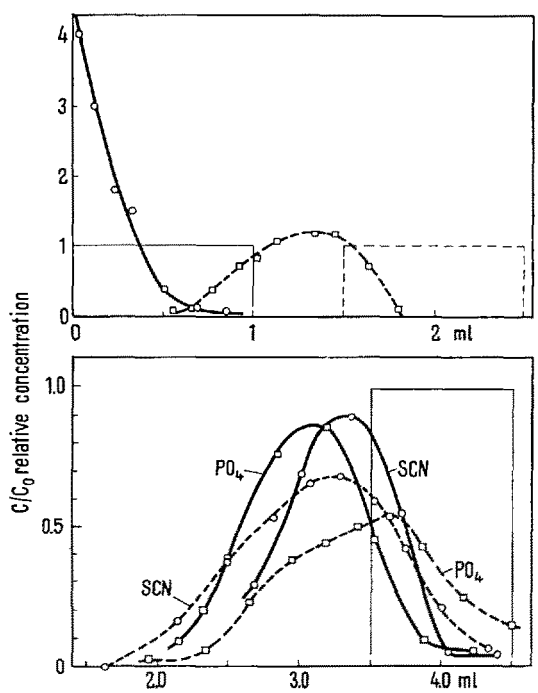

Fig. 7. Phosphate in cellulose paper (Whatman No. 50). A (solid lines): 1 ml of 0.0025 M KH_2PO_4 placed on dry paper, followed by water. B (broken lines): 1 ml of same solution introduced after 1.5 ml of water, and followed by water.

Fig. 8. Two salts on chromatography paper and on glass cloth. A (solid lines): 1 ml of a mixture of KH_2PO_4 and KSCN (0.005 M each), following 3.5 ml of water, and followed by water, in cellulose paper (Whatman No. 4). B (broken lines): Same experiment on glass cloth.

When mixtures were tested, not only relative forward displacement of the solutes but also partial resolution of the mixture were always observed (Figure 8). On cellulose paper (and nylon), phosphate preceded thiocyanate; it was the reverse on glass. On glass cloth the protein and the polysaccharide did not give the elongated profiles obtained with the capillary tube, but pointed curves which in position and shape resembled those of sucrose or salts; this is understandable since the experiments lasted hours (instead of 1 or 2 min in the tube), giving diffusion enough time to operate. The behavior of these substances on celullose was similar, but the results were spoiled by loss through adsorption.

The extent of the relative displacement varied with the porous material. It was most marked, and of about the same magnitude, in Whatman cellulose papers Nos. 4 and 50, in spite of a 10-fold difference in flow rate; somewhat less in glass cloth; very slight in one type of glass paper (Whatman GF-B).

In all experiments of type (2) the profiles had the appearance of normal distribution curves (in Figure 8, the phosphate curve on glass is one of the least symmetrical), differing only in relative displacement. The frontal accumulation of solute in experiments of type (1) seemed simply to be another manifestation of this displacement: on cellulose, the faster moving phosphate accumulated at the front; on glass, the faster moving thiocyanate. The fact that this order was reversed between cellulose and glass is not explained. In the tube, phosphate seemed to move a little faster than thiocyanate, which agrees with the difference in molecular size. In porous flow, it is most likely that secondary phenomena enter into play, such as a certain degree of adsorption-elution, which would slow down certain components, molecular orientation at boundaries[9], which would reduce the effective size of the open passages, or streaming potentials[10]. It is rather remarkable, however, that no essential difference was noted here between cellulose paper, which 'binds' water strongly[11,12], and glass cloth. The main fact is that all solutes travelled faster than the mean velocity of the solvent, in accord with expectation. In chromatographic parlance, one would say that the Rf was always greater than one.

These results have some significant implications. In the flow of blood, for example, there must be differences in the mean velocities of the components, in the decreasing order red cells–protein–salts–water, which may be of physiological importance. As a matter of fact, it has already been shown with radioactive tracers that red cells circulate *in vivo* faster than proteins[13], and many peculiarities in the viscosity of blood and of other suspensions (discussed in detail by BAYLISS[14]) can be explained by the centripetal displacement of the components during flow. In another realm, the phenomenon must operate in the movement of minerals in subsoil waters, sorting out the particles and solutes according to size, and make also more effective the mechanism of

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erosion. In Nature, the instances of capillary or porous flow are far more numerous and varied than those of turbulent flow, and the phenomenon consequently deserves some attention[15].

Résumé. On a étudié l'écoulement d'une suspension de globules rouges et de diverses solutions, soit dans un tube capillaire de 0,7 mm de diamètre, soit en milieux poreux (papiers de cellulose, papiers de verre, toile de verre) en déterminant la répartition des solutés dans les zones limites solution-solvant et en la comparant aux valeurs théoriques.

Dans le tube capillaire, les globules rouges s'écoulent toujours plus vite que la vitesse moyenne de l'eau. Protéine et polysaccharide font de même tant que la vitesse d'écoulement est assez rapide. Les sels minéraux se déplacent exactement à la même vitesse que l'eau. En milieux poreux, toutes les substances étudiées montrent une avance de 1,1 à 1,4 sur la vitesse de l'eau.

On attribue ce phénomène à la poussée hydrodynamique centripète que reçoivent les solutés dans l'écoulement laminaire cylindrique, qui les déplace vers les lignes de courant plus rapide.

J. BOURDILLON

Division of Laboratories and Research, New York State Department of Health, Albany (U.S.A.), June 12, 1962.

[15] The writer is indebted to Dr. S. BULLIVANT for reading the manuscript and making valuable suggestions.

COGITATIONES

Correlation of the Biological Activity of Organic Compounds by Means of the Linear Free Energy Relationships

During recent years, evidence has been accumulated about the possibilities of correlating the biological activity of substances with physical constants characterizing a certain part or the whole molecule under investigation. Obviously, this attempt is useful not only from the practical viewpoint (possibility of predicting the biological activity) but also for theoretical reasons, primarily for the study of the mechanism of the biological process. For example, relationships between biological activity and chemical reactivity, polarographic half-wave potentials, oxidation-reduction potentials and solubility of the active compounds have been found. This field has recently been reviewed[1]. In this connection it seems especially attractive to investigate the possibility of utilizing the linear free energy relationships well known from theoretical organic chemistry[2-5]. Hitherto, this correlation has been established in several cases by means of the Hammett equation[6-13] and the recently proposed $\alpha\beta$-equation[14-16]. It seems desirable to study this problem in a more systematic way.

Let us suppose that the biological efficiency is governed, for example, by the value of the partition coefficient p of the active compound between a polar and non-polar liquid phase. This means that the biological efficiency is in principle determined by this physical magnitude. Let us suppose, moreover, that the efficiencies $e_{i,1}$ and $e_{i,2}$ are studied of a series of compounds (i denotes the i-th member) in two biological systems (1 and 2); for the dependences on p holds: $e_{i,1} = f_1(p_i)$ and $e_{i,2} = f_2(p_{i,2})$. Evidently, a function exists fitting the relation $e_{i,1} = f(e_{i,2})$ which might be a linear one. In this way we have made the biological data 'independent' of a certain more or less arbitrarily chosen magnitude. Then we can investigate the connection with various types of constants in a more systematic way. This approach was tacitly used in the following discussion (see[14]). It is clear that the same consideration could be applied to the chemical reactivity.

Definitions of some Concepts

Biological Object: the experimental animal or the object on cell or molecular level; this object is treated with the *biologically active* substance which may be any defined organic compounds. In the present work we are concerned with the investigation of compounds (with a certain functional group) whose substituents are structurally related. *Biological system:* the system consisting of a biological object and a series of biologically active compounds in interaction.

The biological effect of the compound under study is a manifestation caused, as a rule, by a number of processes, which take place in a biological system. The process *governing* the magnitude of the biological effect (biological efficiency) is to be termed the *efficiency-determining step*. In principle, this process can be either a chemical reaction between the functional group of the biologically active compound and the reactivity centre in the biological object or a process the nature of which resembles the partition of a compound between two immiscible liquid phases[17]. In the first case we shall speak of a chemical reaction as an efficiency-determining step and in the second case of a physical process.

Linear Free Energy Relationships: These relations can be characterized by Eq. 1

$$E_i - E_r = \phi\psi_i \qquad (1)$$

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