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INTERFEROMETRIC STUDIES OF DIFFUSIVE UNSTIRRED LAYERS GENERATED IN GRAVIOSMOTIC SYSTEMS

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Using the interferometric method, the effect of the force of gravity on diffusive layers formed around the membranes of a graviosmotic system was studied. The results obtained were in the form of interferograms. These give direct information about the layers, such as their thickness and solute concentration drops on them. To analyse the interferograms, theoretical models of graviosmotic systems were developed filled with solutions of density decreasing and increasing with concentration. The present work is crucial with respect to all basic problems connected with the near-membrane layers of graviosmotic systems. It solves all the basic problem pertaining to physical interpretation of the graviosmotic phenomenon, which up to now was studied mainly in its biophysical aspect.

INTRODUCTION

Graviosmosis is one of the basic membrane phenomena determined by the force of gravity. Is was first observed in two-membrane systems in 1971 (Kargol, 1971, Przestalski & Kargol, 1972). It consists in the occurrence of volume flows, called grawiosmotic flows, as a result of proper reorientation of a membrane system with respect to the direction of the force of gravity. Membrane systems that give rise to such flows have been called graviosmotic systems (Kargol, 1971, 1978; Przestalski & Kargol, 1972).

A graviosmotic flow can be directed upwards or downwards, depending on whether the solution used is of density increasing of decreasing with concentration.

A number of effects is connected with the graviosmotic phenomenon, such as: pumping of water against gravitation force circulation of water, asymmetry and amplification of the graviosmotic transport (Kargol, 1978, 1981, 1992; Kargol, Dworecki & Przestalski, 1979). Based on that phenomenon as well on the related effects, a hypothesis has been developed by Kargol (1978, 1992) of graviosmotic xylem water transport in plants. According to the hypothesis the elevation of water along the xylem vessel elements could occur not only by the mechanisms of the cohesion-transpiration theory, or due to rot pressure, also by using the graviosmotic mechanisms.

Since the graviosmotic phenomenon was discovered a number of attempts have been made to give it a physical interpretation and describe the graviosmotic flows analytically (Kargol 1971, 1978, 1981, 1985; Przestalski & Kargol, 1972, 1987; Ślęzak, 1983). All those attempts are based on a postulate put forward by Kargol & Przestalski (Kargol, 1971; Przestalski & Kargol, 1972). According to the postulate the force of gravity has either destructive or a stabilising effect on the near-membrane layers unstirred layers generated near the membranes of a graviosmotic system. Up to now, there was no sufficient experimental evidence in support of the postulate. The present work is bridging this gap. In it the layers were investigated by using the interferometric method (Dworecki, 1984, Kargol *et al.*, 1986; Dworecki, Kargol & Przestalski, 1987; Lerche & Wolf, 1971) with a Mach-Zehnder interferometer (Francon, 1966).

The results of the studies performed are in the form of interferograms, which help solve all the basic problems concerning near-membrane layers in graviosmotic systems. The analysis of the interferograms permitted the formulation of an exhaustive physical interpretation of graviosmosis, and the elaboration of theoretical models of the membrane systems we are interested in. The models may constitute the starting point for elaboration of a full analytical description of graviosmotic volume flows.

RESULTS AND DISCUSSION

The investigation of the near-membrane diffusive layer generated in graviosmotic systems was conducted with an experimental setup based on Mach-Zechnder interferometer (Francon, 1966; Kargol *et al.*, 1986). A schematic of the setup is shown in Fig. 1. The membrane systems studies with plane-parallel

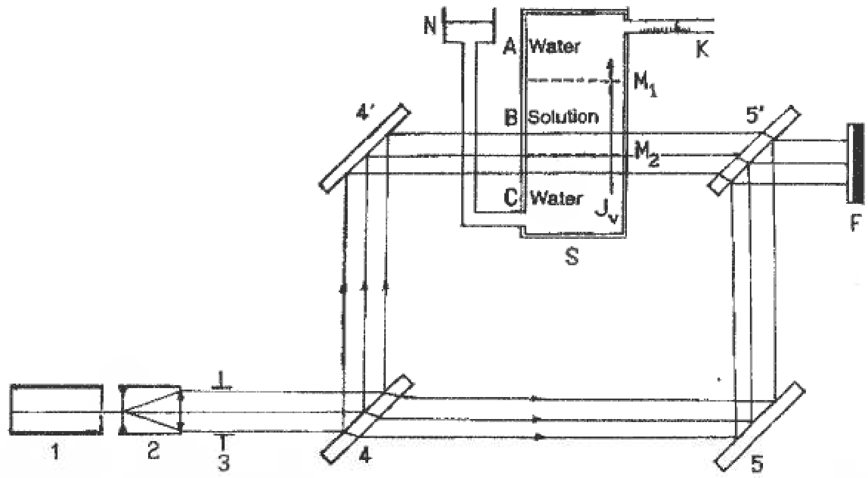

Fig. 1. Measuring setup: 1 - laser, 2 - telescope, 3 - diaphragm, 4, 4', 5, 5' - plane - parallel plate, F - film camera, S - graviosmotic system (M_1, M_2 - membranes, A, B, C - departments, J_v - graviosmotic flux, N - vessel).

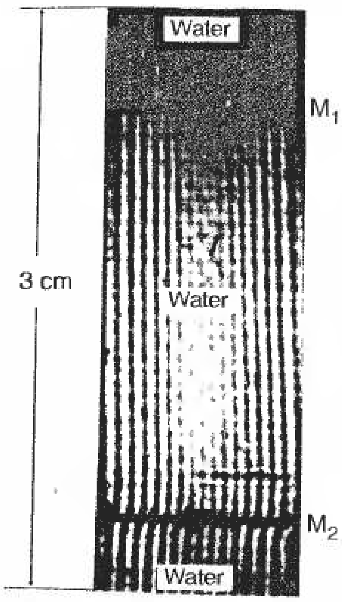

Fig. 2. Test interferogram.

side walls was placed in the path of two laser beams, between mirrors 4' and 5'. The beams were so formed as to encompass the entire graviosmotic system, the vicinity of membranes M_1 and M_2 inducing. The investigation results were obtained in the form of interferograms recorded with a camera F (photographic or TV).

With this setup a study was performed of the layers of interest in various graviosmotic systems, using solutions of density increasing as well as decreasing with concentration.

The sample results reported in the present paper are concerned with a graviosmotic system with two neprophane membranes, in which were employed water solutions of ethanol (as solutions of density decreasing with concentration) and glucose solutions (as solutions of density increasing with concentration). Each membrane had the filtration coefficient L_p=5·10^{-12} m^3·N^{-1}·s^{-1} and the following reflection coefficients: σ_e= 0.028 (for ethanol) and σ_g =0.068 (for glucose). In all experiments the side compartments (A and C) of the membrane system were filled with pure water, and the middle compartment (B) - with the chosen solution of concentration C_B.

In order to check whether the interferometric system had been correctly aligned, a test interferogram was made. That inerferogram, taken when all the three compartments (A, B and C) were filled with water, is shown in Fig. 2. The setup is correctly aligned when the interference lines are straight and parallel.

With the middle compartment filled with the solutions studies, interferograms proper were made. A selection of these is shown in figures 3, 4, 5 and 6.

Interferograms shown in figures 3, 4 and 5 were made for water solutions of ethanol of concentrations 0.125 [M], 0.25 [M] and 0.5 [M]. The unit [M] expresses the number of moles of sulote in one litre of solution. Interferograms (a) in those figures were recorded after 10 minutes of formation of the near-membrane layers, and interferograms (b) - after 20 minutes of formation.

Analysing all the interferograms, it is easy to note that the interference lines are curved slightly on both sides of the upper membrane M_1 encompassing fairly narrow regions of the solutions. Thickness of those regions, which are just the near-membrane diffusive layers, are constant and do not depend on the concentration C_B of ethanol in the solution. They also do not depend on the time of formation. The slight bend of the interference lines within the near-membrane lay-

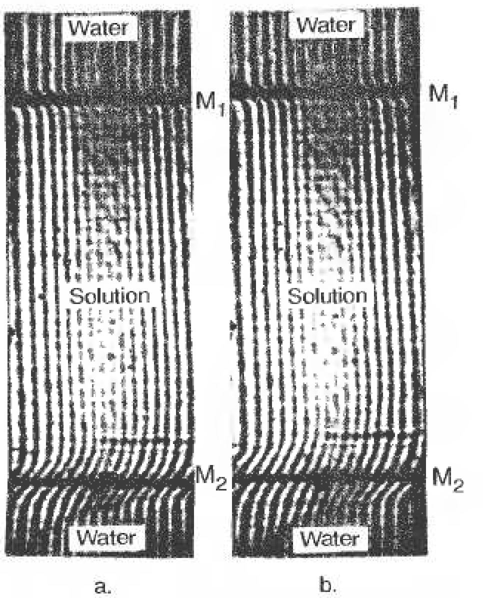

Fig. 3. Interferograms obtained with 0.125 [M] ethanol solution. The unit [M] expresses the number of moles of solute in one litre of solution.

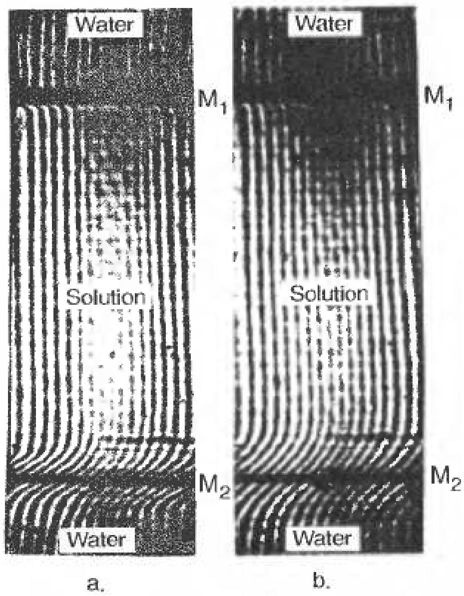

Fig. 4. Interferograms obtained with 0.25 [M] ethanol solution.

ers indicates that there is a relatively small concentration drop on them. Thus, the concentration difference on the membrane must be sufficiently great, as must also be the osmotic pressure difference.

The situation is different near the lower membrane (M_2). Here the line curvature is markedly greater and encompasses markedly broader regions of the solutions on both sides of the membrane. That means that the thickness of the diffusive layers are relatively great (markedly greater than those of the layers generated near membrane M_1). The concentrations drops on them are also sufficiently great, and small is the concentration difference on the membrane as well as the osmotic pressure difference. From the bend of the lines it follows that thickness of the diffusive layers and concentration drops on them increase in time. They are the greater the greater the ethanol concentration C_B of the solution in the middle compartment (B) of the graviosmotic system.

In general, one can say that the osmotic pressure difference on membrane M_2 is considerably smaller than the difference on membrane M_1. Thus there occurs in the system a certain resultant difference in effective osmotic pressures.

The interferograms are different when solutions of density increasing with concentration are employed in the graviosmotic membrane system. Such sample interferograms are shown in Fig. 6. They were obtained with a system of two nephrophane membranes and glucose solutions. Interferograms (a) and (b) in this figure were obtained with the glucose solution C_g equal to 0.025 [M]. The first of them was recorded after 10 minutes, the second after 20 minutes, of unstirred layer formation. The last interferogram is for glucose concentration $C_g=0.05$ [M]. It illustrates the state of layer formation after 10 minutes of their formation.

Now, analysing these interferograms it is easy to infer from the bending of the interference lines that here near-membrane layers of small thickness are formed at the lower membrane (M_2). Thickness of those layers depend neither on the solution concentration nor on time of formation. The concentration drops on them are not great. Correspondingly great must then be the difference between concentrations on membrane M_2, and thus the difference in osmotic pressure.

Layers of relatively great thickness are, however, formed at the upper membrane (M_1). Their thickness and concentration drops on them are great and increase in time, as can be inferred from the curvature of the interference lines. Therefore, the difference in concentrations at membrane M_1 must be small and so is the osmotic pressure difference. Thus, in the case of solutions of density increasing with concentration, a certain resultant osmotic pressure difference also develop.

THEORETICAL MODELS OF GRAVIOSMOTIC SYSTEMS

Based on the results presented above of interferometric investigations of the unstirred layer in gravios-

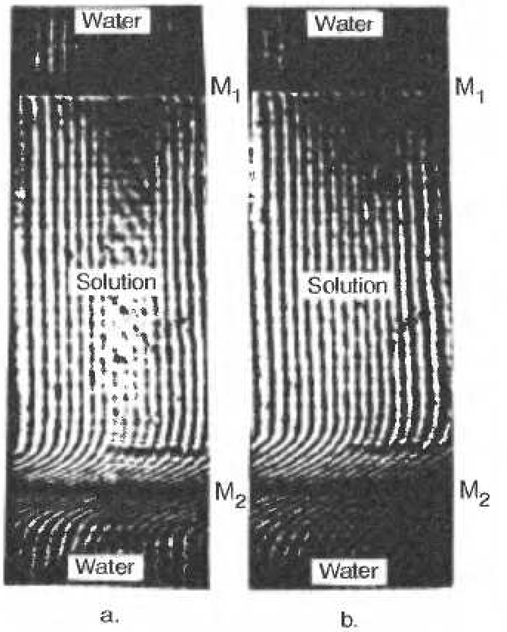

Fig. 5. Interferograms obtained with 0.5 [M] ethanol solution

motic systems, we have constructed two theoretical models (a) and (b), of those systems. They are presented in Fig. 7 a, b.

The first one (a) refers to systems with solutions of density decreasing with concentration, the second one (b) - systems with solutions of density increasing with concentration. We begin their presentation with model (a) shown in Fig. 7a.

It was assumed in the model that concentration C_e of the middle compartment (B) is higher than the concentration C_{oe} of solutions filling the compartments (A and C) of the graviosmotic system. In the situation presented, in the vicinity of the upper membrane (M_I) there develop two relatively thin diffusive l_e layers and l_{eB}. Their thickness does not depend, as already states, either on concentration C_e on or time of formation. This is confirmed by the interferograms in Figs. 3, 4 and 5.

In order to explain that state of affairs let us take a closer look at the diffusion of the solute upwards from compartment B to A. The molecules leave the layer l_{eB}, permate the membrane M_1 and the enter the compartment A forming here the layer l_e. As a result of such movement of the solute molecules, the mean concentration of layer l_{eB} becomes smaller than concentration C_e in the bulk of compartment B. However, the density of the layer will be greater than the density of solution C_e. In the case of layer l_e the situation will be different. This layer will have a higher concentration than C_{eo} in compartment A. Its density will thus be lower than that of solution C_{eo}. In the field of gravity both the layers become unstable. Therefore, convective current KK will develop in compartments A and B, resulting in a destruction of the diffusive layers.

The intensity of convective currents will be the greater the diffusion of solute molecules, and thus the greater the concentration C_e, in the middle compartment. This results in constant thickness of layers l_e

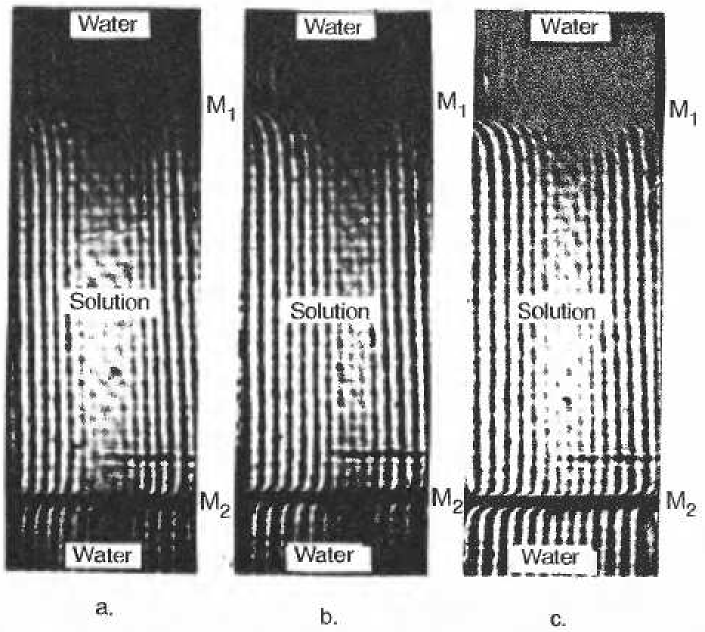

Fig. 6. Interferograms obtained with 0.25 [M] glucose solution (interferograms a and b) and with 0.05 [M] solution (interferogram c).

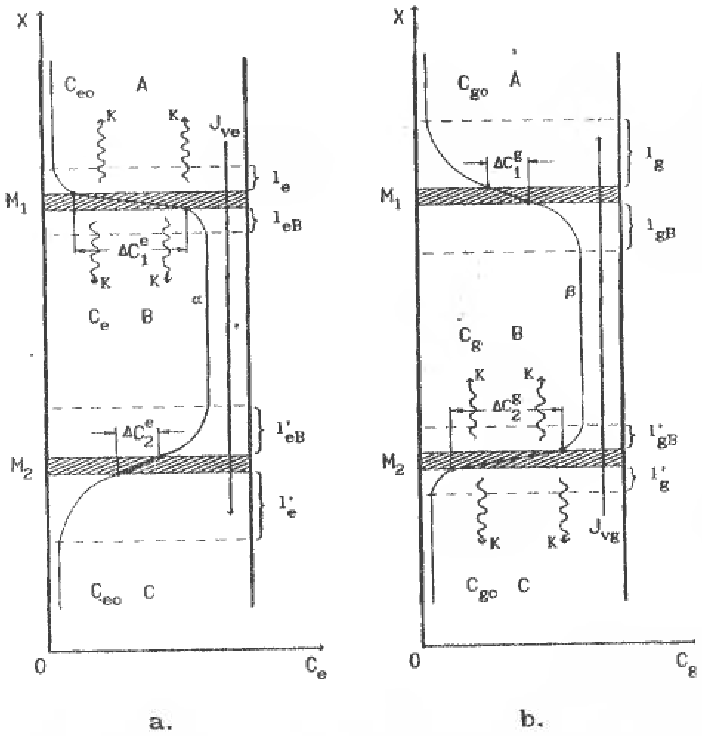

Fig. 7. Theoretical models of graviosmotic systems (a - model of a system filled with a solution of density decreasing with concentration, b - model of a system filled with a solution of density increasing with concentration, and - concentration profiles).

and l_{eB} - not dependent on concentration C_e, which is confirmed by the interferograms in Figs. 3, 4 and 5. These interferograms indicate also the thickness of the layers in the stationary state do not depend on time.

Due to the generation of convective flows (KK) the layers in compartments A and B undergo destruction to a certain extent. As a result the difference ΔC_1^e in concentration on the thickness of membrane M_1, and thus the difference in osmotic pressures $\Delta\Pi_1^e = RT\Delta C_1^e$, will be relatively great.

The surroudings of the lover membrane differ. Here the solute diffusion that goes from compartment B to C results in the formation of stable layers l'_e and l'_{eB} (see Fig. 7a). This is because the mean density of layer l'_{eB} is greater than the density of solution in the whole of compartment B, while the mean density of layer l'_e is lower than the concentration C_O of compartment C.

In the situation presented, the force of gravity stabilizes the layer. Thickness of the layers and drops in concentration in them increase with time. Analysis of interferograms shown in Figs. 3, 4 and 5 confirms this. Over the membrane thickness there will occur a relatively small concentration difference ΔC_2^e and thus not a great osmotic pressure difference $\Delta\Pi_1^e = RT\Delta C_2^e$.

In general we can say that in the field of gravity the system undergoes osmotic polarisation. A certain resultant osmotic pressure difference occurs:

$$\Delta\Pi_e = \Delta\Pi_2^e - \Delta\Pi_1^e$$

which induces volume flow J_V of the solution, called graviosmotic flow. That flow is directed downwards, since $\Delta\Pi_1^e >> \Delta\Pi_2^e$.

Generation of such flows has been confirmed by a relatively large number of earlier works (Kargol 1971, 1978, 1992, Kargol et al. 1979, Przestalski & Kargol 1972, 1987 and others).

In the case of model (b) shown in Fig. 7b the situation with the unstirred layers is different. This model is for graviosmotic systems where the solution employed is of density increasing with concentration. Here, as is easily understood, in the vicinity of the upper membrane M_1 stable layers l_g and l_{gB} develops in the field of gravity. Their densities and concentration drops on them increase in time, as follows from the interferograms of Fig. 6a, b and c.

Correspondingly, the concentration difference ΔC_1^g on the membrane decreases to small values.

Thus, in the stationary state, the osmotic pressure difference $\Delta\Pi_1^g = RT \cdot \Delta C_1^g$ on that membrane in relatively small. However, near the lower membrane (M_2) we deal with unstable layers $l_g^{'}$ and $l_{gB}^{'}$. They induce convective currents KK in both compartments B and C. The form of gravity exerts a destructive effect on the layers. Their thickness as well as concentration drops on them are not great, which is confirmed by the interferograms Figs. 6a, b, c.

Thus, the concentration difference ΔC_2^g is relatively great, and consequently, the osmotic pressure difference $\Delta\Pi_2^g = RT \cdot \Delta C_2^g$ on the membrane (M_2).

In this case, the 2-membrane system becomes polarised cosmetically in the field of gravity. The osmotic pressure difference that remains:

$$\Delta\Pi_g = \Delta\Pi_2^g - \Delta\Pi_1^g$$

induces graviosmotic flow J_V directed upwards (because $\Delta\Pi_1^g >> \Delta\Pi_2^g$). This state of affairs has been confirmed by experimental studies (e.g. Kargol, 1971, 1978, 1980, 1992; Kargol et al., 1979; Przestalski & Kargol, 1972; Ślęzak, 1983).

The models presented above, being highly adequate to the reality, may constitute a very good starting point for a full analytical description of graviosmotic flows.

It should be further added, that the convective currents KK can be observed visually when a solution of fluorescent is employed, or else by using electrolytes - due to irregular oscillations of electrical potential (Kargol, 1988).

CONCLUSIONS

Using the interferometric method direct investigations of the near-membrane diffusive layers that are generated in graviosmotic systems were performed. The studies were performed with solutions of density decreasing and increasing with concentration. Typical in this respect were solutions of ethanol and glucose. The results of the study were obtained in the form of interferograms. The report presents selected interferograms of systems filled with water solutions of ethanol and glucose.

By analysing these interferograms, definitive theoretical models of graviosmotic systems were created that take into account the near-membrane stable and unstable layers generated by the force of gravity. The unstable layers induce convective flows. As a result of these differing layers and convective flows the graviosmotic system undergoes osmotic polarisation, and graviosmotic flows develop.

The models presented are able to represent the real state of the near-membrane layers very well and in a convincing manner. They may be used for elaboration of a full analytical description of graviosmotic flows.

The study contains direct evidence for the existence and state of near-membrane diffusive layers. It also solves all the basic problems concerning the layers and the problems connected with physical interpretation of the graviosmotic phenomenon.

In conclusion, it should be added that that phenomenon has, up to now, been studied mainly in its biophysical aspect. Based on that, as mentioned earlier, a graviosmotic hypothesis of xylem elevation of water in plants has been worked out (Kargol, 1978, 1992). That hypothesis is not in conflict with either the transpiration-cohesion or the root pressure theories. More than that - it supplements those theories.

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