LETTER TO THE EDITOR

The Concept of Chemical Capacitance: Is It Necessary or Meaningful?

Dear Sir:

In the November 1986 issue of the *Biophysical Journal* an article by Okajima and Hong (1) appeared which reported on displacement photocurrents from purple membranes attached to thin Teflon films. In agreement with the results of other investigators, Okajima and Hong reported that the photocurrent was composed of a fast and a slow component (B1 and B2, respectively) of opposite polarities. The authors conclude that "the time course of the B1 signal is completely predictable by an equivalent circuit containing a chemical capacitance" (1). The concept of a "chemical capacitance" was orignally introduced by Hong et al. (2,3), however I challenged the physical meaning of the "chemical capacitance" (4).

A peculiarity in the early experiments was the partial illumination of the membrane: as argued explicitly in reference 4, the magnitude of the "chemical capacitance" was just that of the illuminated area of the bilayer. Okajima and Hong (1) now report a control experiment: they compared the fast B1-photocurrent under complete and partial illumination of the membrane by progressively focusing the laser beam. The photocurrent amplitude and time course then could be superimposed on each other (Fig. 8 in reference 1). It is important to note the authors statement that in this experiment the laser energy was held constant and that the photoresponses were in the linear range. The authors interpret this result to mean "the chemical capacitance is not the ordinary membrane capacitance in disguise" (1). This conclusion may be rejected for the following reason.

As can be easily calculated from the molar extinction coefficient of bacteriorhodopsin at the excitation wavelength (ϵ_{590nm} = 50,000), one needs 5×10^{15} photons/cm^2 to excite 67% of all bacteriorhodopsin molecules in a thin film (OD < 1). This photon density corresponds to 2 mJ/cm^2. In comparison, according to reference 1, the energies used for partial illumination ranged from 2 J/cm^2 to 33 J/cm^2. Hence the experiments under discussion were carried out under conditions of extreme oversaturation which must have resulted in multiple absorption effects. (This may also be the reason for the discrepancies mentioned in reference 1 between results from reference 1 and those of other authors.) Although under all excitation conditions the bacteriorhodopsin is saturated, these multiple absorption effects may well be linear over the range of flash energies applied.

If one takes for granted the experimental finding of the linear increase of the photocurrent with the flash energy, then it is trivial that the photocurrent was the same whether generated over a large or small area, as the photocurrent depends solely on the number of absorption processes. The number of absorption events is determined by the number of photons in a flash that was constant. Hence the experimental result can be completely explained without assuming a "chemical capacitance" and does not represent control experiments with respect to the critique I raised in reference 4.

A different argument against the concept of a "chemical under all capacitance" results from the nature of the equivalent circuit of Fig. 1 and the numerical values calculated for the "chemical capacitance." On the one hand, the equivalent circuit given by Okajima and Hong does not describe the system appropriately, as it fails to account for the series connection of the purple membranes with the capacitance of the Teflon film support. The latter is a membrane that is physically distinct from the attached photoactive purple membranes. Therefore, it should be incorporated into the equivalent circuit as a separate entity, rather than writing it off as "an integral part of the membrane" (1).

On the other hand, to be meaningful, a "chemical capacitance" should behave as some kind of interfacial capacitance (similar to the Gouy-Chapman ionic double layer capacitance). Specifically, its magnitude should depend on the photoactive membrane (here purple membranes) and on the ionic conditions, but not on the material and the thickness of the supporting membrane. What, however, is the meaning of a "chemical capacitance" that amounts to micro-Farads/cm^2 in the case of lipid bilayer membranes (2,3) and pico-Farads/cm^2 in the case of a Teflon film support (1)?

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