Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

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Version of record first published: 24 Sep 2006.

To cite this article: La. Valkova , C. Betrencourt , A. Hochapfel , I. V. Myagkov & L. A. Feigin (1996): Monolayer Study of Monensin and Lasalocid in the Gas State, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 287:1, 269-273

To link to this article: http://dx.doi.org/10.1080/10587259608038763

Mol. Cryst. Liq. Cryst., 1996, Vol. 287, pp. 269–273
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Monolayer Study of Monensin and Lasalocid in the Gas State

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(Received November 22, 1995; in final form January 19, 1996)

Langmuir films of monensin and lasalocid sodium salts have been studied in the gas state, where they present positive surface pressure up to high molecular areas. The results are analysed in terms of $\pi A - \pi$ coordinates, where π and A are the pressure surface and the molecular area respectively. The obtained linear curves show that the films have a behavior similar to a perfect gas. The results are consistent with molecular associations at the interface.

INTRODUCTION

The carboxylic polyethers monensin and lasalocid isolated from *streptomyces* are known to form liposoluble cation complexes in biomembranes and mimetic bilayers.[1] The antibiotic activity is considered as a consequence of the ionophore properties of these complexes. Although their mecanisms of actions are similar, there are differences in ion selectivity and in insertion mode in the biomimetic membranes.[2,3] The two compounds, mainly used as sodium salts (MON-Na and LAS-Na), are applied in veterinary medecine and recent studies have shown their efficiency against MDR (multi drug resistance) in cancer therapy.

Among the different configurations of these molecules, micellar aggregates have been detected in solution under certain conditions.[4] There is also an aggregation tendency at the air–water interface when the compounds are incorporated in phospholipid monolayers.[5]

LAS-Na and MON-Na can be spread and compressed as Langmuir films on an aqueous subphase due to their particular amphiphilic character which might be classified as "facial".[6,7]

The compression isotherms (surface pressure π versus molecular area A) of both antibiotics show characteristic and almost constant surface pressures within a wide molecular area range in the part where the molecules are in the gas state. The purpose of this study has been to make use of this effect in order to detect association phenomena and also to find an explanation for the positive surface pressures.

EXPERIMENTAL

MON-Na (90–95%), LAS-Na (90–95%) and DPPC (99%) were from Sigma Chemicals. The solvents chloroform and hexane were of chromatographic quality and potassium chloride of analytical grade.

The experiments with MON-Na and LAS-Na monolayers on air–water interface was carried out with the equipment of film balance MDT (Russia), where the surface pressure was measured with a Wilhelmy plate with an accuracy better than 0.01 mNm^{-1} for the surface pressure, and $\pm$ 5 mV for the electric potential jump ΔV.

We have prepared the monolayers on the surface of doubly distilled water (pH = 6) from antibiotic in hexane-chloroform (3:2 by volume). The monolayer was compressed after 5 minutes delay so as to allow the solvent to evaporate at the rate of 5–7 cm^2min^{-1}. The monolayer was compressed at 10^{-3} nm^2sec^{-1} per molecule. The temperature of the water subphase and the air was 20 °C.

RESULTS AND DISCUSSION

In order to investigate the aggregation behaviour of the antibiotic molecules on the air–water interface, π – A diagrams were recorded. The isotherm and the curve of the electric potential jump for MON-Na and LAS-Na are shown in Figure 1 and Figure 2 respectively. The compression isotherms of the pure antibiotics have already been studied elsewhere[6] except in the gas state at surface pressures below 3 mNm^{-1}. The surface pressures have been measured with a precision of 0.05 mNm^{-1} in this state for varying values of the molecular area. Figure 3a and Figure 4a represent curves that have been reproduced at least four times. The results have been analysed in terms of $\pi A - \pi$ coordinates. As shown in Figure 3b and Figure 4b, these curves are linear, therefore the equation $\pi(A-A_0)=n^{-1}$ kT similar to that of ideal gas state can be applied.[8] In this

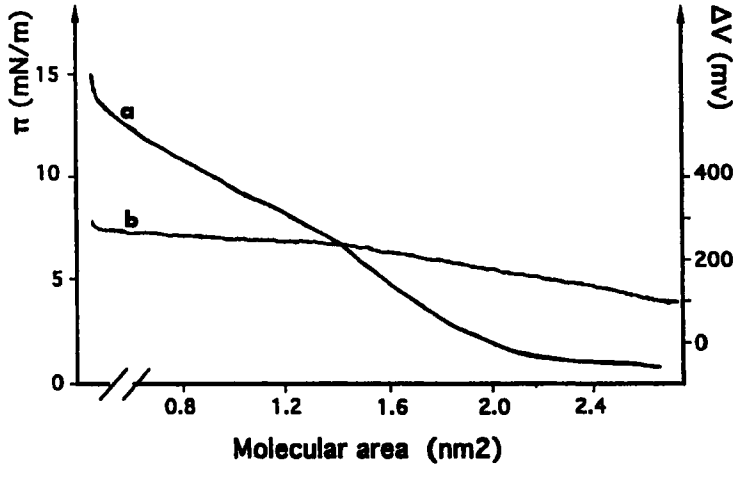

FIGURE 1 Compression isotherm of MON-Na on water (a), and the electric potential jump (b).

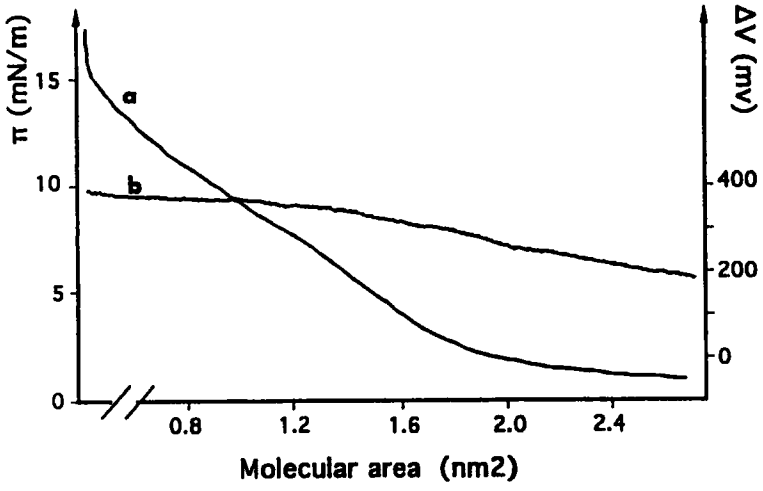

FIGURE 2 Compression isotherm of LAS-Na on water (a), and the electric potential jump (b).

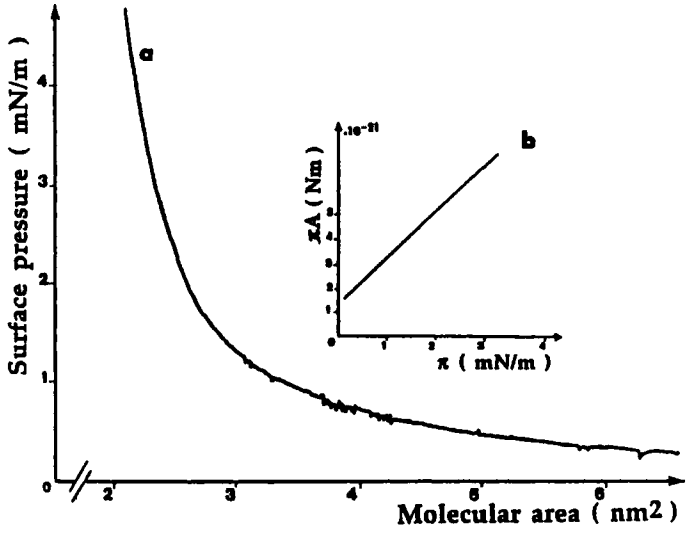

FIGURE 3 Compression isotherm of MON-Na in the gas state.

equation A_0 is the effective area occupied by the molecules on the surface and appears as a cosurface correction and n^{-1} takes into account the cohesion forces. [9,10] The slope of the linear part of the isotherm in $\pi A - \pi$ coordinates corresponds to A_0. When we extrapolate to $\pi = 0$, the limit value of the energy on the vertical axis divided by kT is equal to the number of molecules in the case of a perfect gas.[9]

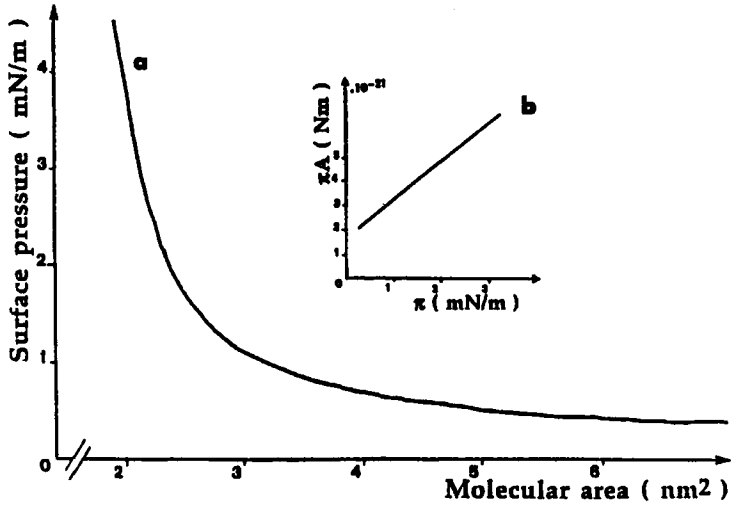

FIGURE 4 Compression isotherm of LAS-Na in the gas state.

In our case we have obtained the value $1.4\ 10^{-21}$ Nm for MON-Na and $1.6\ 10^{-21}$ Nm for LAS-Na. Since $kT = 4.06\ 10^{-21}$ Nm, we have $n = 2.9$ for MON-Na and $n = 2.5$ for LAS-Na. These results can be interpreted as associations in the monolayers of about 3 molecules in the case of MON-Na; whereas two kinds of associations of two or three molecules are equally probable in the case of LAS-Na.

The electric potential jump ΔV does not present any abrupt variations thus the monolayer can be considered as homogeneous. The values of ΔV (near collapse) are 270 mV for MON-Na and 360 mV for LAS-Na. Moreover they did not change when 10^{-3} M KCl was added to the subphase. This means that the value of the jump depends only on the dipole structure of the antibiotic molecules and that the molecules have no charge (when using a subphase of pH = 6).

The present results have shown that among the various configurations adopted by lasalocid and monensin, aggregates seem to be formed under certain conditions as here in the gaseous state of Langmuir monolayers. The association phenomena appear as low but positive values of the surface pressure up to large molecular areas. In this respect, both antibiotic molecules behave in a similar way.

Acknowledgement

This work was partially supported by DGA (DRET) France.

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