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Mixed Monolayers Prepared from Diynoic Acids and Amphiphilically Modified Nucleobases: I. Phase States at the Air–Water Interface and in Langmuir–Blodgett Films

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Abstract—This paper reports the preparation and properties of mixed monolayers of diynoic acids and amphiphilically modified nucleobases. Synthetic nucleobases in the monolayers retained the ability to bind complementary nucleotides from the aqueous subphase. Photopolymerization of diynoic acid resulted in fixing the monolayer structure. Such monolayers are a new type of models for studying molecular recognition on surfaces of biological membranes. Procedures were developed for transferring the monolayers onto solid substrates and for photopolymerizing the diynoic acid–nucleobase derivative mixtures. The deposited films were characterized using atomic force microscopy. The concentration ranges of diynoic acid corresponding to the formation of homogeneous (true) mixtures or domain-like structures were determined from the compression isotherms of mixed monolayers. The same concentration ranges were obtained from examining atomic force microscopy images of the deposited films.

Key words: molecular recognition, nucleobases, photopolymerization, monolayer, Langmuir–Blodgett film, atomic force microscopy

INTRODUCTION

Molecular recognition (specific interaction between molecules based on the complementarity of their surfaces), which is well known in chemistry, is key to understanding a multitude of functionally important biological processes and to developing new materials with preset functional properties. One example of molecular recognition is spontaneous and highly specific interaction between complementary nucleobases (base pairing), which underlies accumulation, storage, replication, and evolutionary modification of genetic information. Given that replication and other processes involving DNA take place in cells, it is of interest to study the mutual recognition of nucleobases in conditions simulating cell systems. Their main characteristic is the presence of membranes. Moreover, bacterial DNA synthase is known to be incorporated into the cell membrane. Therefore, the goal of this series of articles was to study the interaction of nucleobases in monolayers, which can be viewed as a model of membrane systems [1–4]. On the other hand, organized monolayers produced using the Langmuir–Blodgett technology offer a unique opportunity for studying intermolecular interactions [5–7] and, hence, molecular recognition [8–10]. For example, a lipid monolayer bearing nucleobases on its surface is capable of hydrogen bonding-based molecular recognition and resembles in this respect biological molecular systems [11, 12]. Amphiphilic derivatives of nucleobases (nucleolipids) have been synthesized just recently [11–13], although the first attempts date back to 1985. Monolayers formed by these lipid molecules carrying nucleobases as hydrophilic "heads" can bind complementary bases from the aqueous subphase [11–14].

Abbreviations: HDA, heneicosa-10,12-diynoic acid; EA, erucyl adenine.