Interaction of a Phospholipid and a Coagulating Protein: Potential Candidate for Bioelectronic Applications

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1. INTRODUCTION

Biomembranes are the outer layer of cells and are mainly composed of phospholipids, glycolipids, sphingolipids, sterols, proteins, etc. Essentially, this system behaves like a two-dimensional fluid at the submicrometer dimension. A biomembrane may be considered as the 2D colloidal system with different novel physical properties like elastic properties, which are essential for various biological functions. They are multicomponent systems. The development of the physical basis of self-organization within multicomponent systems is a real challenge. It is possible to mimic an artificial model membrane with one or more biomembrane components to study the specific membrane functionality at the molecular level. Accordingly, interest on the study of the membrane structure and dynamics as well as interactions between membrane components is gaining to a large extent.

The Langmuir–Blodgett (LB) method is one of the best techniques to mimic and study an artificial biomembrane. This technique allows one to investigate the thermodynamic behavior and the biophysical and biochemical processes within the membrane, interaction between various membrane components, and other suitable molecules like drugs, antibiotics, proteins, lipids, polymers, surfactants, and various other biomacromolecules. The main advantage of the LB method is that one can have molecular level control during membrane formation. Using the LB method, one can prepare bilayer assemblies with various lipid compositions and structural arrangements. This key feature of the LB technique allows preparation of asymmetric lipid assemblies.

With the appropriate choice of lipid composition and thermodynamic conditions (i.e., temperature, pH, film deposition pressure, etc.), a great deal of control can be achieved over the lateral organization in membrane assemblies using this technique. In the present study, we investigated the interaction between a phospholipid 1,2-dioleoyl-sn-glycero-3-phosphocholine (DOPC) and a coagulating protein protamine sulfate (PS) using the Langmuir–Blodgett (LB) technique. The π−A isotherm, π−t characteristics, and analysis of isotherm curves suggested that PS strongly interacted with DOPC, affecting the fluidity of the DOPC layer. Electrical characterization indicates that PS as well as the PS−DOPC film showed resistive switching behavior suitable for write once read many (WORM) memory application. Trap-controlled space charge-limited conduction (SCLC) was the key mechanism behind such observed switching. The presence of DOPC affected the SCLC process, leading to lowering of threshold voltage ($V_{TH}$), which is advantageous in terms of lower power consumption.

ABSTRACT: In the present communication, we have investigated the interaction between a biomembrane component 1,2-dioleoyl-sn-glycero-3-phosphocholine (DOPC) and a coagulating protein protamine sulfate (PS) using the Langmuir–Blodgett (LB) technique. The π−A isotherm, π−t characteristics, and analysis of isotherm curves suggested that PS strongly interacted with DOPC, affecting the fluidity of the DOPC layer. Electrical characterization indicates that PS as well as the PS−DOPC film showed resistive switching behavior suitable for Write Once Read Many (WORM) memory application. Trap-controlled space charge-limited conduction (SCLC) was the key mechanism behind such observed switching. The presence of DOPC affected the SCLC process, leading to lowering of threshold voltage ($V_{TH}$), which is advantageous in terms of lower power consumption.

Received: December 31, 2021
Accepted: April 15, 2022
Published: May 17, 2022

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addition, such devices are biodegradable and may be the alternative for present Si-based electronics with almost no hazardous e-waste. In addition, biomaterials are lightweight, compatible with a flexible substrate, cheap, and widely available. Accordingly, we have investigated the resistive switching (RS) behavior using PS and PS–DOPC as the active layer of such devices. RS memory has been considered as the potential candidate for future memory technology. Memory has added advantages like reliability, high density, scalability, fast switching, and low power consumption. In addition, a variety of materials, viz., organic, inorganic, polymer, biomaterials, etc., can be used to design RS memory device. Biodegradable RS memory is assumed as the alternative to the present semiconductor memory with a sustainable solution toward e-waste management. Of late, a number of biomaterials, viz., proteins, plant extracts, polysaccharides, chitosan, etc., have been exploited to design RS memory. In the present case, it has been observed that PS can be used to design biocompatible resistive memory. Interestingly, the PS–DOPC system enhances the memory performance in terms of lower power consumption.

2. EXPERIMENTAL SECTION

2.1. Materials. DOPC (purity, >99%) was purchased from Sigma Chemical Company and used as received. Proteamine sulfate (PS) from herring (grade III) was also purchased from Sigma-Aldrich Chemical Co. (CAS number: 9007-31-2) and used as received. Chloroform (99.9%; SRL, India) was used as a solvent for the preparation of DOPC solution. A working solution of PS was prepared by dissolving it into distilled water. The key components of acetate buffer, acetic acid (glacial 100% HR), and sodium acetate were purchased from Merck Limited.

2.2. Surface Pressure vs Area per Molecule Isotherm. Surface pressure versus area per molecule (π–A) isotherms were obtained with a commercially available LB film deposition instrument (Apex 2000C, Apex Instruments Co., India). The area of the Langmuir trough is 472.5 cm² having a length of 31.5 cm and a breadth of 15 cm. The concentrations of the working solutions for PS and DOPC were 0.5 mg/mL. The π–A isotherms of DOPC in the presence and absence of PS were recorded. For isotherm measurement in the presence of PS, different amounts of PS were premixed in the subphase. To do that, 25 μL of chloroform solution of DOPC was spread on the aqueous subphase (pure water) of the LB trough in the presence and absence of PS using a microsyringe. However, to check the effect of pH, acetate buffer (ionic strength, 0.05 M) was used. After waiting for sufficient time to evaporate the solvent, the barrier of the LB trough was compressed slowly at a rate of 5 mm/min to study the isotherm characteristics. The surface pressure (π) versus average area available for one molecule (A) was measured by a Wilhelmy plate arrangement. Data for surface pressure–area per molecule isotherms were obtained by a computer interfaced with the LB instrument. Before each isotherm measurement, the trough and barrier were cleaned with chloroform and then rinsed with distilled water. Each isotherm was repeated a number of times, and each isotherm curve presented here is an average of three independent measurements with a fluctuation of ±0.01 nm². All the experiments were performed at room temperature (25 °C).

2.3. Reaction Kinetic Study. To study the interaction between DOPC and PS, DOPC solutions were spread onto the LB trough (volume, 360 mL) containing different amounts of PS. After waiting for various amounts of time for interaction to occur, the π–A isotherms were recorded. Also, in certain cases, the barrier was kept fixed and changes in surface pressure with time (π–t curve) were recorded, where the surface pressure is measured by the Wilhelmy plate arrangement. This experiment was performed at room temperature (25 °C). The resistivity of water was 18.2 MΩ·cm and pH was 6.8. The π–t curve presented here is an average of two independent measurements with a fluctuation of ±0.2 mN/m.

2.4. Resistive Switching Device. Resistive switching devices were designed by depositing PS, DOPC, and PS–DOPC (molar ratio, 10:1) mixture onto an ITO-coated glass substrate using the drop-casting method. After allowing sufficient time to evaporate the solvent, the deposited films were kept in vacuum for 24 h. Gold (Au) electrodes were deposited onto these films using vacuum deposition. In the designed device, either PS or PS–DOPC acted as the active layer, Au as the top electrode, and ITO as the bottom electrode. A source meter (Keithley 2401) and a homemade probe station were used to characterize the device. The I–V curve presented here is an average of five independent measurements having a standard deviation of less than 10% in the threshold voltage.

3. RESULTS AND DISCUSSION

3.1. Surface Pressure vs Time Characteristics. Measurement of change in surface pressure as a function of time using the LB technique allows one to have an idea about the surface activities within the Langmuir film. This process has been extended to study the interactions between phospholipids and other biomembrane components with suitable materials like drugs, nanoparticles, enzymes, DNA, and cholesterol. In the present case, also the LB technique has been employed to study the interaction between floating DOPC films and water-soluble coagulating protein PS. Here, we recorded the variation of surface pressure for DOPC Langmuir films in the absence and presence of PS in the subphase as a function of time. To do that, we spread DOPC in the subphase and the barrier was kept fixed when 5 mN/m surface pressure was attained. At around 5 mN/m surface pressure, the monolayer attains the liquid-expanded phase. Here, we have chosen 5 mN/m surface pressure (onset of the liquid-expanded phase) so that the PS molecules can penetrate into the lipid layer or, when PS molecules interacted with DOPC, a reorientation of the molecules may be possible. Considering the volume of the trough where PS was premixed and the surface area of the trough where DOPC was spread, the molecular volume/area of interaction for PS–lipid was 77.1.

It has been observed that in the absence of PS, no significant change in the surface pressure of the DOPC monolayer is observed even after 6 h as shown in Figure 1. This indicates that DOPC forms a stable film at the air–water interface. However, in the presence of PS in the subphase, the surface pressure is increased close to 8 mN/m within a span of 6 h even when the barrier is kept fixed. It has been observed that, initially, the surface pressure increases at a higher rate. However, after passage of time, the rate of increase in surface pressure decreases. With time greater than 5 h, the curve has become almost flat with respect to the time axis. This indicates the completion of reaction/interaction between floating DOPC and PS in the subphase.

It may be mentioned here that the increase in surface pressure in the π–t curve indicates the penetration as well as interaction of PS molecules within the lipid layer. A similar increase in surface pressure was also observed when the enzyme penetrated...
through the DPPC floating layer. Our later studies revealed that in the case of pure DOPC isotherm, the mean molecular area or limiting molecular area is 1.008 nm² (Table 2). In the presence of PS, the area per molecule increases up to 2.75 nm² (Table 2). This also clearly indicates the interaction between PS and DOPC molecules. However, considering the large size of PS (molecular structure given in the Supporting Information, Figure S2, MW = 4–5 kDa) and the increase in the area of isotherm in the presence of PS, it can be concluded that partial penetration of the PS molecule occurred. Also considering the overall ionic charge of PS as +21 and the molecular volume/area of interaction for the PS-lipid ratio of 77:1 used in this experiment, it is assumed that all the lipids interacted with PS in the system when completion of reaction occurred.

At this point, almost all floating DOPC molecules are attached to PS and no free DOPC molecules exist to further incorporate PS molecules. Accordingly, the PS–DOPC complex film is formed at the air–water interface. This clearly demonstrates the interaction and incorporation of PS onto the DOPC monolayer.

### 3.2. Pressure–Area Isotherm

To have an idea about the interactions between PS and DOPC, we have investigated the Langmuir film behavior of DOPC in the presence of PS in the subphase at various concentrations and varying waiting times. Corresponding pressure–area isotherms are shown in Figure 2a,b.

The isotherm of DOPC in pure water starts rising with a lift-off area of 1.32 nm² and shows a steep rise before the collapse pressure is reached at around 41.8 mN/m. It has been observed that the DOPC monolayer showed the characteristics of an expanded-like feature until the collapse pressure is reached. No such distinct phase change and/or plateau in the isotherm were observed. This may be due to the fact that nonsaturated hydrogen chains of DOPC molecules make the Van der Waals cohesive interactions between the hydrophobic chains weaker. This may lower the efficiency of molecular packing and also prohibits the formation of a typical condensed phase. The values as well as the shape and nature of the DOPC isotherm are similar to the previously reported results. However, it is interesting to note that the DOPC isotherm recorded in the presence of PS in the subphase is shifted to a larger area per molecule. This suggests that the floating monolayer gets expanded due to interaction of PS and the DOPC molecules. The enlargements in molecular areas of the isotherms indicate that there exists strong interaction between cationic PS and zwitterionic lipid DOPC in the floating layer. There may be different kinds of interactions between protein and lipid, such as electrostatic, hydrophobic, hydrogen bonding, etc. It is important to check the surface activity of PS alone during Langmuir film formation. To do that, PS solution was spread onto the subphase and the barrier was compressed, but no significant rise in surface pressure was observed, indicating that PS possesses almost no surface activity. This suggests that water-soluble PS does not remain on the surface but rather mixed into the subphase (water) of the Langmuir trough. However, the expansion of the area of the π–A isotherm of DOPC in the presence of PS demonstrates the monolayer-disturbing action of PS. To have an idea about the interaction, we have mixed a fixed amount of PS (0.5 mg/mL, 1 mL) within the subphase in the Langmuir trough. After that, DOPC solution (0.5 mg/mL, 25 μL) was spread onto the subphase. The PS:DOPC molar ratio was 229.9. Then, the compression isotherms were recorded with varying waiting times, viz., 30, 60, 90, and 120 min (Figure 2a).

Interestingly, it has been observed that with the increase in waiting time, the isotherms of DOPC shifted toward a larger area per molecule up to 120 min. However, at a waiting time higher than 90 min, no further significant increase in area per molecule occurred. DOPC used in this study is zwitterionic and PS is cationic in nature. When DOPC solution was spread onto the PS subphase, the PS molecule came in contact with the floating DOPC layer and attached it with the DOPC molecules through electrostatic interaction. The size of the PS–DOPC complex is large compared to pure DOPC molecule. So, the areas per molecule tend to shift toward the larger area. This interaction is a time-dependent process as observed from the surface pressure vs time characteristics (Figure 1). Numbers of PS molecules that come in contact with floating DOPC molecules increase with time. Accordingly, the compression isotherm shifted to a larger area with higher waiting time (Figure 2a). The isotherm with a waiting time of 120 min possessed the maximum shift toward a larger area. A plot of mean molecular area as a function of waiting time (inset of Figure 2a) also indicated that, initially, the area of isotherm linearly increases

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**Table 1. Monolayer Characteristics Extracted from the π–A Isotherm Curves (Figure 2a) of DOPC Measured with Varying Waiting Times and the Absence of PS**

|        | C1 (mN/m) | C2 (mN/m) | lift-off area (nm²) | mean molecular area (nm²) | collapse Pressure (mN/m) |
|--------|-----------|-----------|---------------------|---------------------------|--------------------------|
| DOPC in pure water | 22 | 15.36 | 1.324 | 1.008 | 41.8 |
| time: 30 min | 22.79 | 19.25 | 2.286 | 1.604 | 42 |
| time: 60 min | 21.11 | 18.47 | 2.86 | 2.173 | 42 |
| time: 90 min | 21.98 | 18.97 | 3.15 | 2.5 | 42 |
| time: 120 min | 21.46 | 15.38 | 3.295 | 2.634 | 42 |
with waiting time and, near 120 min, it becomes almost flat. This indicates that within 120 min, most of DOPC in the floating layer interacted with PS and almost no further DOPC molecules exist in the floating layer to incorporate further PS molecules. Accordingly, no further significant increase in the area of the floating layer occurred with a waiting time higher than 120 min. As a whole, this observation suggested that within 120 min, after spreading of DOPC onto the PS subphase, the interaction between DOPC and PS almost completed.

To have further insight about this interaction, DOPC in the lipid layer, and PS in the subphase, we have recorded DOPC isotherms spread onto the PS subphase with varying amounts of PS, viz., 0.5, 1, 1.5, 1.75, and 2 mL mixed in the subphase, resulting in final PS concentrations in the LB trough of $6.9 \times 10^{-3}$, $1.3 \times 10^{-3}$, $2.1 \times 10^{-3}$, $2.4 \times 10^{-3}$, and $2.7 \times 10^{-3}$ mg/mL, respectively. For all the cases, the DOPC amount was kept fixed at 25 µL, resulting in PS:DOPC molar ratios of 114.9, 229.9, 344.8, 402.3, and 459.8. The waiting time after spreading was 15 min. Corresponding isotherm curves (Figure 2b) suggested that with the increase in PS amount, the isotherm shifted toward a larger area of up to 1.75 mL of PS in the subphase. After that, no further significant shift in the area of the isotherm curve was observed. This indicates that higher numbers of PS molecules are attached to floating DOPC molecules with the increase in PS amount in the subphase. However, when the PS amount in the subphase increased beyond 1.75 mL, no further shift of isotherm curves occurred. A plot of mean molecular area as a function of PS:DOPC molar ratio (inset of Figure 2b) also indicated that, initially, the area of isotherm linearly increases with PS concentration and, at the PS:DOPC molar ratio of 402.3, it becomes almost flat. This indicates the completion of the interaction. In the PS–DOPC mixed system, the relative molar ratio of PS:DOPC was varied from 114.9 to 459.8. The overall ionic charge of PS is $\pm 21.59$. Considering the large amount of PS present and higher charge of PS, it is assumed that all the lipids interacted with PS in the system when no further rise in the area of lipid isotherm occurred, i.e., after completion of the interaction.

However, a close look at Figure 2a,b reveals that at the final equilibrium state, area per molecule values are different for the two cases. This may be due to different approaches and conditions to record the isotherms presented in two figures. But the insets of both figures confirmed that, initially, the area per lipid increases with an increase in waiting time or PS concentration present in the subphase until completion of interactions.

Also, a comparison between Figures 1 and 2a shows the difference in time to reach the final equilibrium stable state corresponding to completion of interaction. During π–t measurement, the barrier, i.e., the area of the trough, was kept fixed corresponding to the surface pressure of 5 mN/m. Here, a smaller trough area was available for floating molecules. On the other hand, during the isotherm measurement for different waiting times after spreading, the area of the trough was at maximum, i.e., the barrier was kept at the maximum expanded condition. During PS–DOPC interaction, PS molecules came onto the surface, reorientation of the DOPC molecules occurred, and finally, an equilibrium orientation was reached, corresponding to completion of reaction. The observed difference may be due to different geometry of the available area in the trough for two different approaches. Since, during isotherm measurement, the available area was at maximum, reorientation was also easily possible at a faster rate.

To have an idea about the effect of pH on the PS–DOPC interaction, compression isotherms of DOPC on the subphase containing PS at different pH values, viz., 3.5, 4.75, 6.0, 6.8, and 7.3, have also been recorded. Corresponding isotherm curves are shown in Figure 3. From the figure, it has been observed that at lower surface pressure, all the isotherms at different pH values are almost similar. However, at the higher pressure region, the nature of isotherm curves differs. With the decrease in pH, the collapse pressure decreases. This suggests that the pH of the subphase affected the PS–DOPC interaction to a certain extent. Also, it has been observed that with increasing pH, the mean molecular area decreases (inset of Figure 3). This suggests more ordered and compact film formation at higher pH. This may be...
due to the reorientation of DOPC molecules at higher pH in the presence of PS.

3.3. Analysis of the Pressure–Area Isotherm. To have further insight about the floating DOPC layer in the presence of PS, different parameters like compressibility, lift-off area, mean molecular area, collapse pressure, etc., were extracted from the isotherm curves of Figures 2a,b and 3 following a standard procedure. Corresponding values are listed in Tables 1–3, respectively. The compressibility of the Langmuir film in two dimensions as a function of surface pressure can be calculated from the π–A isotherm using the following standard thermodynamic relation,

\[ C = \frac{1}{\pi} \left( \frac{\pi_2 - \pi_1}{a_2 - a_1} \right) \]

where \( a_1 \) and \( a_2 \) are the areas per molecule at surface pressures \( \pi_1 \) and \( \pi_2 \), respectively. Here, two compressibilities were calculated, one at a lower surface pressure range (\( \pi_1 = 5 \text{ mN/m} \) and \( \pi_2 = 15 \text{ mN/m} \)) and the other at a higher surface pressure range (\( \pi_1 = 25 \text{ mN/m} \) and \( \pi_2 = 35 \text{ mN/m} \)).

On the other hand, the mean or limiting molecular area (\( A_{\text{lim}} \)) was estimated by extrapolating the steep linear part of the isotherm to zero surface pressure in the area per molecule axis. The idea about compressibility is very important in the case of lipid films. An increase in compressibility reflects the lowering of the order of monolayer structure and hence an increase in fluidity of the lipid layer. At the same time, the \( A_{\text{lim}} \) value gives the idea about the packing as well as the distance between the adjacent molecules in the Langmuir films. In the present case, for all the DOPC isotherms measured in the presence of PS in the subphase, the \( A_{\text{lim}} \) values increase with an increase in waiting time as well as an increase in the amount of PS present in the subphase. This indicates the expansion of floating lipid films in the presence of PS. However, this expansion gets stabilized after a waiting time of 120 min or when the amount of PS is 1.75 mL in the subphase under the present experimental condition. On the other hand, we have calculated compressibility for all the isotherms of both lower and higher surface pressure regions. Calculated values of compressibility revealed that for all the cases at the lower surface pressure region, the lipid layer showed higher compressibility. This indicates that at lower surface pressure, the lipid films remained in expanded form with higher fluidity. A decrease in compressibility value at the higher surface pressure region indicates the increase in molecular order and formation of a high-density lipid layer at higher pressure. This is also indicative of lowering of the film fluidity. The analysis of compressibility values also indicates that lipid molecules attain maximum ordered and compact organization within the Langmuir films in the presence of PS for a waiting time of 120 min and when the PS amount present in the subphase is 1.75 mL. We have also drawn the compression modulus vs surface pressure (\( C_{\text{s}}^{-1} \)) curves from the isotherm characteristics using the formula,

\[ C_{\text{s}}^{-1} = -A \frac{d\pi}{dA} \]

\( C_{\text{s}}^{-1} \) is proportional to the first derivative of the surface pressure (\( \pi \)) with respect to the mean molecular area (\( A \)). Corresponding \( C_{\text{s}}^{-1} \) vs \( \pi \) plots are shown in Figure S1 of the Supporting Information. \( C_{\text{s}}^{-1} \) values indicate the details of phase behaviors associated with Langmuir films. Davies et al. systematically described the use of \( C_{\text{s}}^{-1} \) to characterize the physical state of lipid Langmuir films. It has been reported that for the lipid monolayer, the gaseous phase is characterized by \( C_{\text{s}}^{-1} \) values of less than 12.5 \text{ mN/m}^2, the liquid-expanded (LE) phase is characterized by \( C_{\text{s}}^{-1} \) values ranging between 12.5 and 50 \text{ mN/m}^2, and the liquid phase is characterized by \( C_{\text{s}}^{-1} \) values ranging between 50 and 100 \text{ mN/m}^2, whereas the liquid-condensed (LC) phase is characterized by \( C_{\text{s}}^{-1} \) values ranging between 100 and 250 \text{ mN/m}^2. In general, the minima observed in the \( C_{\text{s}}^{-1} \) curves indicate the phase transition point in a lipid monolayer film. On the other hand, the maxima (Figure S1c,d) observed in the \( C_{\text{s}}^{-1} \) curve correspond to the most compressed states of the lipid monolayer films. A close look into Figure S1 revealed that all the monolayers are in the typical liquid-expanded (LE) state in accordance with the Davies and Rideal criteria. Plots of the maximum values of \( C_{\text{s}}^{-1} \) as a function of time and PS:DOPC molar ratio (Figure S1c,d)

**Table 2. Monolayer Characteristics Extracted from the π–A Isotherm Curves (Figure 2b) of DOPC Measured with Varying Amounts of PS in the Water Subphase**

| Sample Description | C1 (mN/m) | C2 (mN/m) | Lift-off area (nm²) | Mean molecular area (nm²) | Collapse pressure (mN/m) |
|--------------------|-----------|-----------|---------------------|--------------------------|--------------------------|
| DOPC in pure water | 22        | 15.36     | 1.324               | 1.008                    | 41.8                     |
| PS:DOPC: 114.9     | 21.45     | 15.295    | 1.529               | 1.151                    | 42                       |
| PS:DOPC: 229.9     | 20.77     | 16.87     | 2.003               | 1.63                     | 42                       |
| PS:DOPC: 344.8     | 20.87     | 17.05     | 3.06                | 2.145                    | 42.5                     |
| PS:DOPC: 402.3     | 20.32     | 13.22     | 3.7                 | 2.741                    | 42                       |
| PS:DOPC: 459.8     | 23.42     | 16.324    | 3.6                 | 2.75                     | 42                       |
also clearly indicate that for all the monolayers, \( C_{m}^{-1} \) values lie well within the range corresponding to the LE phase. \(^{57}\) However, slight variations in the maximum values of \( C_{m}^{-1} \) indicate that the presence of PS affected the compressibility and hence the fluidity of the lipid layer to a certain extent.

It is well known that coagulating proteins tend to segregate the lipid films into lipid microdomains, representing a “hotspot” during the blood coagulation process. \(^{75}\) In the present case, the PS used is a coagulating protein. \(^{76,77}\) Therefore, in the presence of PS, there is a high probability of formation of microdomains or partition within the DOPC layer, leading to a slight change in the fluidity of the lipid layer in the presence of PS.

Different parameters extracted from the isotherm curves measured at different pH values are listed in Table 3. A close look into Table 3 showed that compressibility values systematically decrease with an increase in pH. At the same time, the mean molecular area also decreases at higher pH values. This suggests that at higher pH, PS–DOPC forms a compact film at the air–water interface. The collapse pressure of the PS–DOPC film increases at higher pH, indicating increases in stability of the mixed film.

It is worth noting that DOPC is a zwitterionic lipid with a positively charged choline group and a negatively charged phosphate acid group. \(^{78,79}\) Polar head groups of DOPC can reorient depending on the ionic strength. \(^{80,81}\) At low ionic strength, the choline groups are located below the phosphate groups, whereas at higher ionic strength, the situation is reversed. \(^{81}\) Therefore, in the present case, also under the investigated range of pH 3.5 to 7.3, changes in ionic strength of DOPC head groups occur. This internally affects the orientation of the DOPC head in the presence of PS. As a whole, the change in the ionic nature causes reorientation of DOPC molecules in the floating layer. This is reflected as the change in the shape of isotherm curves measured at different pH values. On the other hand, it has also been shown that PS binding with the cell wall increases with an increase in pH. \(^{82}\) Therefore, in the present case, binding of coagulating PS with the DOPC layer may also increase at higher pH. Accordingly, the PS–DOPC mixed film becomes compact and more stable at higher pH.

### 3.4. Resistive Switching Behavior

The schematic of the switching device structure is shown in Figure 4. Two devices have been prepared with device structures Au/PS/ITO (device-1) and Au/PS–DOPC/ITO (device-2). \( I–V \) characteristics for both the devices are shown in Figure 5. For both the devices, Au is used as the top electrode, whereas ITO is used as the bottom electrode. Initially, both the devices are in low conducting state, i.e., in high resistance state (HRS). In the case of device-1, a forward bias (0 \( \rightarrow \) +2 V) is applied; the device maintains its HRS until an applied voltage \( V \) of \(<1.30 \text{ V})\). When the scanning voltage reaches an applied voltage \( V \) of 1.30 V, the device abruptly switches from low conducting to high conducting state, i.e., low resistance state (LRS). The corresponding voltage is known as the typical threshold voltage \( (V_{th})\) for the device. At this voltage \( (V_{th} = 1.30 \text{ V})\), the device switches from HRS to LRS, i.e., OFF to ON state. Interestingly, once the device switches to its LRS/high conducting state (ON state), it retains the ON state even when the bias/scanning direction is reversed. Also, the device preserves/retains its ON state even when the electrical power is switched off. So, the observed switching between two resistance states \( (\text{HRS} \rightarrow \text{LRS})\) is irreversible and nonvolatile in nature. \(^{83,84}\) Such behavior is referred to as the Write Once Read Many (WORM) phenomenon. The ON/OFF ratio, i.e., the memory window, was found to be \(\sim 4.57 \times 10^2\). The memory window characterizes the distinction between the two states, i.e., ON and OFF states of the memory devices. In the case of resistive switching-based memory devices, this can be estimated by measuring the ratio of ON state current and OFF state current at a particular read voltage. This can also be calculated as \( R_{\text{OFF}}/R_{\text{ON}} \). A higher memory window is advantageous. In the present case, an observed memory window of 1.11 \(\times 10^2\) is well within the suitable limit for potential memory application point of view. \(^{85}\)

Device-2 also showed similar memory characteristics. However, in the case of device-2, while scanning from 0 \( \rightarrow +2 \text{ V})\), the device switches to its ON state (HRS \( \rightarrow \text{LRS})\) at 0.6 V. Here, the device switches from OFF to ON state at a much lower threshold voltage compared to that in the case of device-1.

To have insight into the mechanism of such observed switching, we have analyzed the \( I–V \) curves for both the devices in a double-logarithmic plot. \(^{33}\) Corresponding plots are shown in Figure 6a,b. Results revealed that both the devices follow Ohm’s law at LRS as confirmed by the linear fitting with slopes of 1.06 and 1.09, respectively, for device-1 and device-2. \(^{46}\) Here, the linear relationship between current and voltage is well maintained. This suggested that in LRS, a conductive channel is formed in between the two electrodes across the active layer. \(^{27}\) On the other hand, in the case of HRS, i.e., the OFF state, both the devices follow Ohm’s law at the lower voltage region as observed from the slopes of 1.14 and 0.968 of the fitted curves (Figure 6a,b).

However, at the later stages, both the devices follow larger slopes of 1.94 and 2.46 for device-1 and device-2, respectively. This indicated that at this stage, the current across the device is

### Table 3. Monolayer Characteristics Extracted from the \( \pi–A \) Isotherm Curves (Figure 3) of DOPC and PS with Different pH Values in the Acetate Buffer Subphase

| pH   | \( C_{1} \) (mN/m) | \( C_{2} \) (mN/m) | Mean Molecular Area (nm²) | Collapse Pressure (mN/m) |
|------|-------------------|-------------------|--------------------------|--------------------------|
| 7.3  | 21.688            | 19.974            | 2.68                     | 46.01                    |
| ambient pH 6.8 | 21.94            | 20.40             | 2.74                     | 42.5                     |
| 6.0  | 25.521            | 20.198            | 2.86                     | 40.73                    |
| 4.75 | 25.644            | 21.431            | 3.02                     | 38.9                     |
| 3.5  | 25.832            | 29.79             | 3.43                     | 32.51                    |
due to the accumulation of charges, i.e., trap-controlled space charge-limited conduction (SCLC) mechanism. This happens mainly due to the presence of traps across the active layer. In the present case, the traps may be created in the functional layer of the device during PS or PS−DOPC layer deposition to form switching devices. Also, the chemical composition or functional groups present in the active layer material may induce such trap formation. Here, the presence of a carboxyl group and amino linkage in the PS molecule may contribute toward trap formation. In the case of device-2, switching occurred at a lower threshold voltage ($V_{th} = 0.6$ V) compared to device-1 ($V_{th} = 1.30$ V). This may be due to variation of depth and shape of trap centers in the presence of DOPC for device-2 as observed from the increase in slope (from 1.94 to 2.46). Accordingly, in the case of device-2, switching was observed at lower threshold voltage. It is relevant to mention in this context that Zhao et al. observed decreases in threshold voltage corresponding to the increase in the slope of linear fitting of the $I$−$V$ curve. The fluidity of the lipid layer was affected by the presence of PS. The fluidity decreases with an increase in surface pressure as well as an increase in PS amount and waiting time. $I$−$V$ characterization indicated that PS and PS−DOPC can be used as the active layer of RS devices suitable for WORM memory applications. It has been observed that the presence of DOPC affected the charge conduction within the active layer of the

4. CONCLUSIONS

In conclusion, we have investigated the interaction process in between the biomembrane component DOPC layer and a coagulating protein protamine sulfate (PS). The pressure−area isotherm as well as $π$−$t$ characteristics indicated that PS molecules strongly interact with DOPC in the Langmuir films. The fluidity of the lipid layer was affected by the presence of PS. The fluidity decreases with an increase in surface pressure as well as an increase in PS amount and waiting time. $I$−$V$ characterization indicated that PS and PS−DOPC can be used as the active layer of RS devices suitable for WORM memory applications. It has been observed that the presence of DOPC affected the charge conduction within the active layer of the
device, leading to the lowering of threshold voltage. This is advantageous in terms of power consumption requirements.

### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge at [https://pubs.acs.org/doi/10.1021/acsomega.1c07395](https://pubs.acs.org/doi/10.1021/acsomega.1c07395).

Compression modulus vs surface pressure ($C_{\text{m}}$) and variation of the maximum value of $C_{\text{m}}$ ($C_{\text{m}}$) with waiting time and PS:DOPC molar ratio graphs of DOPC in the absence and presence of PS in the subphase (Figure S1); molecular structure of PS (Figure S2) (PDF).

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#### Notes

The authors declare no competing financial interest.

### ACKNOWLEDGMENTS

S.A.H. is grateful to DST for funding to carry out the work (ref. CRG/2021/004073). The authors are also grateful to UGC, Govt. of India, for financial support to carry out this research work through financial assistance under the UGC-SAP program 2016. The authors also acknowledge the fund by the Deanship of Scientific Research at Jouf University to carry out the research. The authors would like to thank Md. Jashim Uddin, Human Physiology Department, Tripura University, for fruitful discussion at different stages of the work.

### ABBREVIATIONS

DOPC, 1,2-dioleoyl-sn-glycero-3-phosphocholine; PS, protamine sulfate; RS, resistive switching; WORM, Write Once Read Many

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