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Growth of Cu_2O on TCO by the SILAR Method Acts as an Alternative Buffer Electrode Layer Towards the Development of Organic Solar Cells

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Abstract

In many types of optoelectronic devices, a layer of poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) is sandwiched between the indium-tin oxide (ITO) electrode and the active layer. Buffer PEDOT:PSS provides a well-defined work function, which is higher than that of ITO (4.2 eV); it smooths the ITO surface and so avoids shorts, and it also protects the active layer from ingress of indium or oxygen, leading to longer device lifetimes. The role of PEDOT:PSS is enormous. Unfortunately, PEDOT:PSS colloidal solution is strongly acidic, and the PSS is strongly hygroscopic, which has the potential to cause degradation in adjacent layers if water is not removed completely. To smooth the surface of ITO and to improve the durability of the device, a buffer layer of Cu_2O has been introduced, adopting the successive ionic layer adsorption and reaction (SILAR) method on ITO. A qualitative comparison has been performed between the Cu_2O buffer layer and the most familiar PEDOT:PSS buffer layer. The thorough characterization of both buffer materials is performed by XRD and UV-Vis transmittance spectroscopy. The surface morphology has been analyzed by atomic force microscopy (AFM). Finally, the influence of the buffer layer on energy conversion efficiency has been tested for the MEH-PPV/PCBM based organic solar cell by fabricating the device with the configuration ITO/buffer-material/MEH-PPV/PCBM/Aluminum. The improvement in open-circuit voltage and light conversion efficiency implied the potential applicability of Cu_2O over PEDOT:PSS as a buffer electrode layer.

Keywords: Successive ionic layer adsorption and reaction (SILAR) method, buffer layer, organic solar cell, growth of Cu_2O

INTRODUCTION

In many optoelectronic devices, layers of PEDOT:PSS are sandwiched between an indium-tin oxide (ITO) electrode and an active organic layer. The role of PEDOT:PSS is enormous. It provides a well-defined work function that is higher than that of ITO, smooths the rough ITO surface to avoid shorts, and protects the active layer from the ingress of indium or oxygen, leading to longer device lifetimes [1, 2]. Unfortunately, the special nature of this material has some disadvantages. The PEDOT:PSS colloidal solution is strongly acidic and has the potential to degrade adjacent layers if water is not completely removed. The morphology of the colloidal particles is gel-like, and they tend to swell or shrink considerably, depending on the humidity, owing to the strongly hygroscopic PSS [3]. During film formation under certain conditions, excess PSS from the colloidal solution appears to form a closed

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surface layer, which increases the work function but also considerably decreases the perpendicular conductivity. These effects considerably influence the final device performance and have therefore been the focus of extensive research in recent years. A wide range of approaches have been developed to address these problems. One of the most prominent methods is the introduction of secondary dopants in addition to PSS, for example, in the form of polyols or anionic surfactants, which change the film-forming properties, water uptake, or work function. Different types of chemical modifications include modifying the PEDOT:PSS colloidal solution with surfactants or organic solvents to suppress screening effects or improve wettability [4, 5]. Thermal treatments of PEDOT:PSS films have also been studied, although there is contradictory evidence as to whether these assist or reduce the formation of a PSS-enriched surface layer. Other reported results include the enhancement of PEDOT crystallinity, changes in surface roughness, diffusion of indium into PEDOT:PSS, or rupture of the ionic bonds between PEDOT and PSS due to annealing. Nevertheless, few studies have investigated the influence of annealing on the PEDOT:PSS complex itself, preheating effects on the colloidal solution, variation in film thickness, and their consequences for device operation [6].

PEDOT:PSS/ITO is a commonly used anode in OLEDs and organic solar cells. The work function of PEDOT:PSS is independent of the work function of the substrate; therefore, the use of PEDOT:PSS is promising for the reproducible fabrication of OLEDs with good performance. In organic solar cells, the PEDOT:PSS layer ensures an improved interface between the active layer and the electrode. The use of PEDOT:PSS also changed the shape of the EQE curve, which was attributed to the doping of the active layer by PSS. It was found that the use of PEDOT:PSS increased yield. The best efficiency reported to date for a small-molecule solar cell of 2.6% was obtained for a device structure using PEDOT:PSS [7]. However, it has also been reported that the use of PEDOT:PSS increases the series resistance of the solar cell, resulting in a lower fill factor. The contradiction between different studies on the influence of PEDOT:PSS on solar cell performance may be due to the fact that the device performance is dependent on the thickness of PEDOT:PSS, and that the properties of PEDOT:PSS are dependent on the thermal treatment. During film formation under certain conditions, excess PSS from the colloidal solution seems to form a closed surface layer, which increases the work function but also considerably decreases the perpendicular conductivity of the film. These effects considerably influence the final device performance and have therefore been the focus of extensive research in recent years [8–10].

In this experimental study, the influence of the buffer layer on the energy conversion efficiency of the MEH-PPV: PCBM bulk heterojunction solar cell was investigated and compared by employing a Cu_2O thin layer and PEDOT:PSS as buffer layers separately. Here, PEDOT:PSS and Cu_2O as buffer reagents were deposited on ITO-coated substrates individually by introducing two different techniques, (i) spin coating and (ii) sequential ionic layer adsorption and reaction, on which the main active MEH-PPV(donor):PCBM (acceptor) composite was spin-coated to fabricate bulk heterojunction solar cells. The material characterizations of the PEDOT:PSS and Cu_2O thin films on the ITO-coated substrate were performed using XRD and UV-Vis transmittance. The surface morphologies were recorded using an atomic force microscopy (AFM). The main bulk heterojunction was formed by incorporating PCBM, an organic acceptor, in conjunction with the MEH:PPV organic donor polymer. Finally, three bulk heterojunction solar cells with the following configurations were fabricated and characterized: ITO/MEH-PPV:PCBM/Al, ITO/PEDOT:PSS/MEH-PPV:PCBM/Al, and ITO/Cu_2O/MEH-PPV:PCBM/Al [11].

EXPERIMENTAL WORK

Reagents

The experimental reagents $CuSO_4$, $Na_2S_2O_3$, NaOH, and NH_4Cl of AR grade and acetone, ethanol, and 1,2-dichlorobenzene of GR grade were purchased from Merck and used without further purification. The transparent buffer material PEDOT:PSS was purchased from Sigma-Aldrich. To fabricate the main

active layer of the bulk heterojunction solar cells, MEH:PPV and $PC_{60}BM$ were purchased from Sigma-Aldrich. All chemicals were used without further purification [12].

Chemical Growth of Cu_2O Buffer Layer and its Characterization

ITO-coated glass was cleaned repeatedly with distilled water, acetone, and ethanol in an ultrasonic bath. It was then dried under vacuum at 70°C before preparing a thin buffer layer on the substrate. On the other hand, 100 mL of 0.1 M aqueous solution of $CuSO_4$ and 400 mL of 0.1 M aqueous solution of $Na_2S_2O_3$ were prepared separately. An aqueous solution of sodium thiosulfate was mixed with 100 mL of $CuSO_4$ solution dropwise until the mixture became colorless. In another beaker, 100 mL of a 0.1 M aqueous solution of NaOH was prepared separately. The pre-cleaned ITO-coated glass substrate was first immersed in a NaOH solution for 3–4 s, and then dipped into the previous mixture for 5 s. The chemical reaction scheme is as follows:

$$Cu^{2+} + SO_4^{2-} + Na^+ + S_2O_3^{2-}$$
$$\downarrow$$
$$Cu^+ + Na^+ + S_4O_6^{2-}$$
$$\downarrow$$
$$Cu_2O + S_2O_3^{2-} + Na^+ + H_2O$$

This process was performed twice, repeatedly and periodically, until a reddish-yellow layer was deposited on the ITO. The back side of the substrate was cleaned with an NH_4Cl solution. As a uniform transparent reddish-yellow layer appeared, the substrate was heated to 100°C in a vacuum oven. Thereafter, the thin active layer of the device was prepared. The substrate was characterized using XRD and AFM. To study transparency, the transmittance spectrum was recorded using a UV-Vis spectrophotometer [13].

Deposition of PEDOT:PSS Buffer Layer and Its Characterizations

The spin-coating technique was adopted for the preparation of the PEDOT:PSS thin layer. The pre-cleaned ITO-coated glass was washed repeatedly with distilled water, acetone, and alcohol and used as the substrate. PEDOT:PSS, purchased from Sigma-Aldrich, was spun onto the substrate at 600 rpm for 2 min and thereafter at 1400 rpm for the next 10 min using an SCU2007 spin-coating unit. As a thin uniform film of PEDOT:PSS was observed, it was transferred to the vacuum chamber and maintained at 100°C for drying. This substrate was characterized using XRD, AFM, and UV-Vis spectrophotometry.

ITO-Coated Substrate Without Buffer Layer and Characterizations

ITO-coated glass was washed with distilled water, acetone, and ethanol repeatedly using the ultrasonication technique. After the cleaning process was completed, the substrate was dried in a vacuum oven at a temperature of 100°C to prepare it for the fabrication of the active layer of the device.

The three ITO-coated substrates with the buffer layer Cu_2O, with the buffer layer PEDOT:PSS, and without any buffer layer were labeled as substrates S1, S2, and S3, respectively.

Fabrication of Solar Cells

Procured MEH-PPV (0.1 mg) was dissolved in 10 mL of 1,2-dichlorobenzene by ultrasonication. $PC_{60}BM$ solution was prepared by dissolving 0.1 mg in 10 mL of 1,2-dichlorobenzene under vigorous stirring for 10 min. As a ready active layer prior to the fabrication of the solar cell, MEH-PPV and $PC_{60}BM$ solutions were mixed at a wt. ratio of 2:1 and stirred for half an hour. Once the desired mixture was obtained, it was spun onto the pre-prepared substrates one after another on substrate (S1), substrate (S2), and substrate (S3) sequentially, at 600 rpm for 2 minutes and thereafter 1800 rpm for 10 consecutive minutes with the help of an SCU2007 spin coater. As the thin active layer was deposited onto the substrates, it was transferred to a vacuum oven for drying at 80°C. An aluminum electrode was used as a cathode on a thin film, which was deposited onto the active layer by the thermal evaporation technique through a shadow mask. To avoid the impact of ambient constraints and various

contaminations, the samples were prepared under identical conditions. A Keithley 2635B source meter was used to study the current–voltage (I–V) characteristics of the devices under dark and white light illumination (100 mW/cm^2). Aluminum electrodes were deposited using a 12A4D HHV vacuum coating unit. The device architecture is shown schematically in Figure 1.

Instrumentations

The structural characterization of the thin film grown on the ITO substrate was performed using a powder X-ray diffractometer (PXRD) (Bruker D8 Advance X-Ray Diffractometer). The optical properties of the film were characterized using a Lambda 365+ Perkin Elmer spectrophotometer in the 300–800 nm wavelength range. Surface Morphology was studied using a Bruker MultiMode 8 NanoScope V AFM under suitable conditions. Aluminum electrodes were deposited using a 12A4D HHV Vacuum coating unit. The current-voltage characteristics of the device were measured under dark and white light illumination using a Keithley 2635B source meter.

RESULTS AND DISCUSSIONS

Figure 2 shows the XRD patterns of the ITO substrate with Cu_2O as the buffer layer (blue), with PEDOT:PSS as the buffer layer (red), and without any buffer layer (black). The diffraction-responsible Bragg planes for ITO were determined with the JCPDS card no.: 71-2194 and Cu_2O by Card No.:05-0667. There were no external peaks for the PEDOT:PSS compound.

Figure 3 shows the AFM images of the buffer surfaces. From the images, it was noted that the thickness of the as deposited buffer layer was quite thin for Cu_2O (deposited by the SILAR method), whereas it was thick for spin-coated PEDOT:PSS.

Figure 4(a) shows the UV-Vis transmission spectra of the buffer materials. The transmittance spectra of the buffer thin layers, PEDOT:PSS and Cu_2O, were recorded by considering ITO as the reference using a Lambda 365+ Perkin Elmer spectrophotometer. The spectrum was recorded from 300 nm to 800 nm for both buffer materials. It was observed that the transmittance of the PEDOT:PSS buffer material was higher at higher photon energy spectra than that of the Cu_2O buffer materials. Within the visible wavelength range, the transmittance of Cu_2O was comparable to that of PEDOT:PSS. In the subject of selection mode to apply within solar cells, Cu_2O also had a potential impact in comparison to the PEDOT:PSS buffer material. As the absorption of the material is inversely related to transmittance, the Cu_2O buffer is more appropriate than PEDOT:PSS for fabricating photovoltaic devices. The absorption coefficient α was determined using Beer-Lambert's equation [14]:

$$\alpha = \frac{2.303}{t}\log\frac{1}{T} \quad (1)$$

where T is the transmittance and t is the thickness of the film. Figure 4(b) shows the variation of the absorption coefficient with respect to the incident energy.

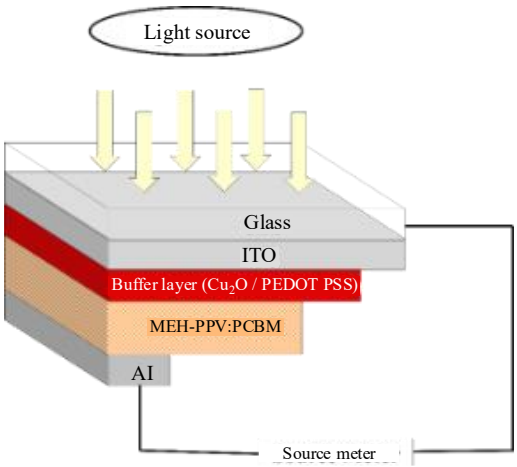

Figure 1. Schematic diagram of the fabricated solar cell.

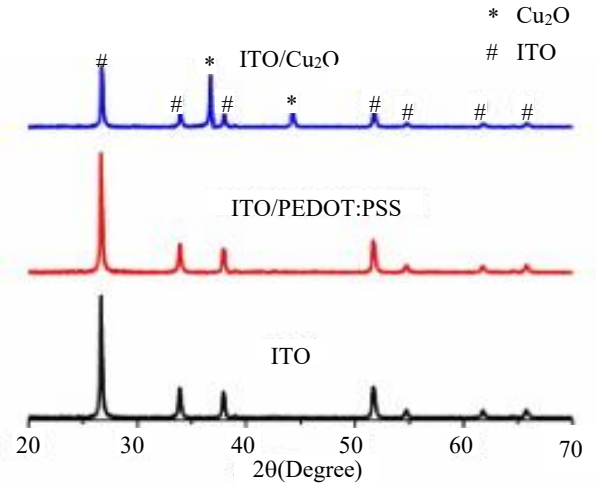

Figure 2. XRD pattern of ITO with and without buffer layers.

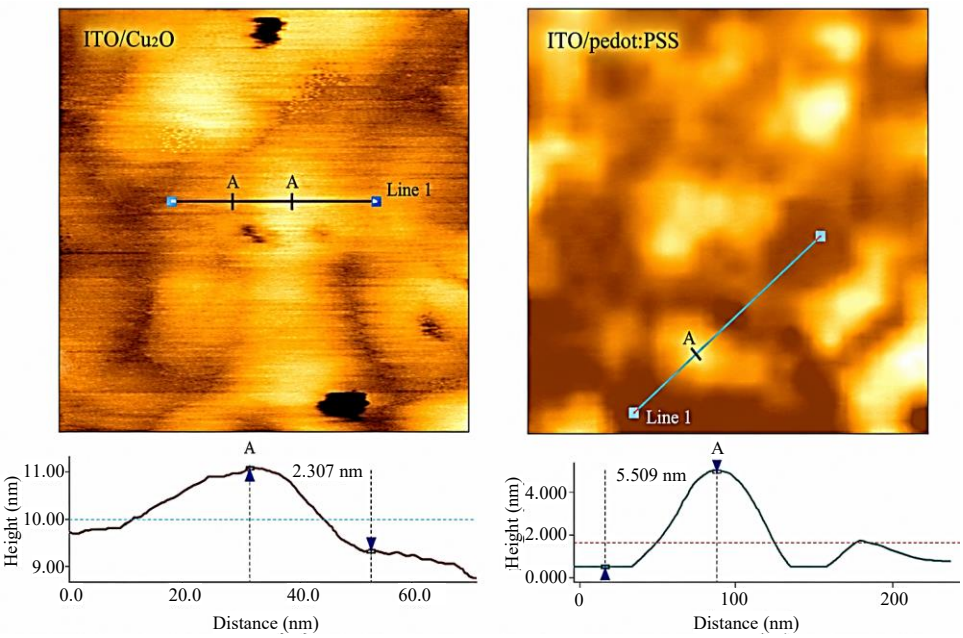

Figure 3. AFM images of as deposited buffer layers.

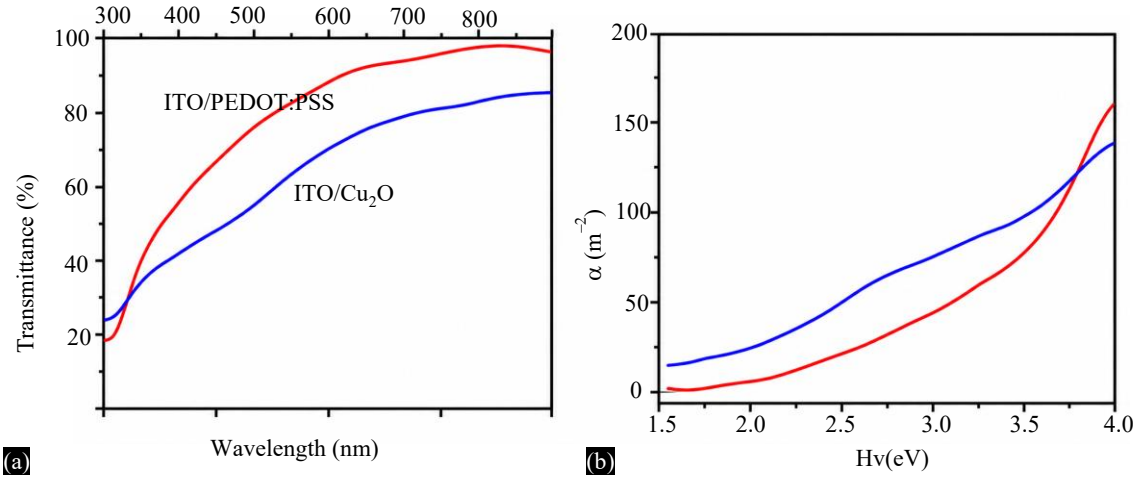

Figure 4. (a) Transmittance spectra; (b) α versus hν plot.

The J-V characteristics under solar irradiation (100 mW/cm^2) for three different compositions of devices on substrates S1, S2, and S3 are shown in Fig. 5(a). The open-circuit voltage (Voc) and short-circuit current density (Jsc) directly contribute to the power conversion efficiency (PCE) of the device.

The characteristic parameters were extracted from the curves using the following relation [15]:

$$FF = \frac{Jmax \times Vmax}{Jsc \times Voc} \quad (2)$$

$$PCE = \frac{Pmax}{P_{in}} = FF \frac{Voc\ Jsc}{P_{in}} \quad (3)$$

Figure 5 shows that after introducing PEDOT:PSS and Cu_2O as two distinct buffer layers on ITO, the open-circuit voltage of the device improved from 350 mV to 450 mV and to 490 mV sequentially. It is interesting to note that the short-circuit current was not improved at all for the device fabricated on the Cu_2O buffer, but it was improved from 19.32 mA/cm^2 to 20.09 mA/cm^2 for the device based on the PEDOT:PSS buffer. The Fill Factors (FF) measured for the devices from the characteristic J-V curves are 0.369, 0.503, and 0.504, respectively, which implies that after the incorporation of Cu_2O and PEDOT:PSS as buffer materials within the device, the FF is improved significantly. Notably, the value of the fill factor for both devices on substrate S1 compared to the device on substrate S2 does not reflect any change, even when the short-circuit current is reduced, after an increase in the open-circuit voltage. The measured values of J_{sc} and V_{OC} for all devices are listed in Table I. The carrier transport dynamics under light irradiance and the ambiguities within thin films, which occurred during the preparation or formation of junctions, were analyzed by estimating the effective series and shunt resistances from the characteristics of the J-V curve (Figure 5(a)). The measured R_s and R_{sh} values are listed in Table I. For an ideal solar cell, the theoretical value of the series resistance is zero, and the shunt resistance is infinite. The results show that after incorporating the buffer medium, the series resistance of the cell is reduced, whereas the shunt resistance is increased, which signifies better performance, an impact on PCE, and improved transport dynamics of excitons. The increase in the shunt resistance and decrease in the series resistance after introducing PEDOT:PSS is evident. Meanwhile, after introducing the inorganic Cu2O as a buffer layer, the series resistance of the cell is also reduced to 8.3 Ω cm^2, which is lower than that of the cell developed on bare ITO and higher than that of the cell fabricated on substrate S2.

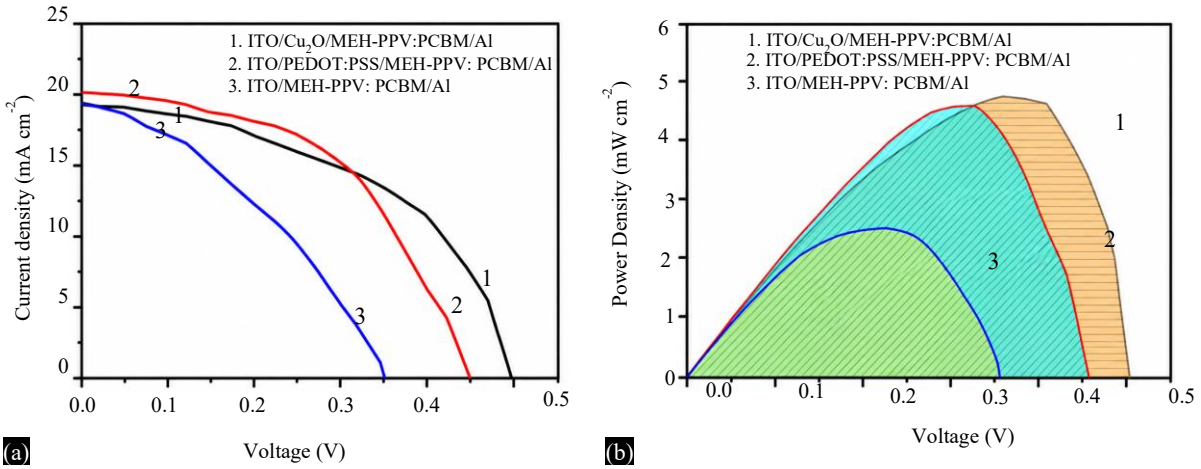

Figure 5. (a) J-V characteristics; (b) Power density against voltage plot.

Table 1. Characteristic parameters of solar cells.

Sample name	J_{SC} (mAcm^{-2})	V_{OC} (V)	P_{max} (mWcm^{-2})	(FF)	PCE (%)	R_s (Ω cm^2)	R_{sh} (Ωcm^2)
ITO/MEH-PPV:PCBM/Al	19.32	0.35	2.5	0.369	2.5	9.4	42
ITO/PEDOT:PSS/MEH-PPV: PCBM/Al	20.09	0.45	4.56	0.504	4.56	5.5	159
ITO/Cu_2O/MEH-PPV:PCBM/Al	19.12	0.49	4.72	0.503	4.72	8.3	140

In contrast, the improvement in the shunt resistance is comparably commensurate with the device made on S2. Figure 5(b) shows the variation in power density with bias voltage.

CONCLUSIONS

The present work of designing a new inorganic buffer layer is an approach toward finding transformative technology for the construction of MEH-PPV: PCBM organic solar cells. The impact of the buffer layer on the energy conversion efficiency, as well as on parasitic resistance with respect to exciton transport dynamics of the MEH-PPV: PCBM bulk heterojunction solar cell, was investigated and compared by employing Cu_2O and PEDOT:PSS thin layers as buffers on ITO. Regarding the electrode buffer layer, computing real-time data and analysis of results imply that the Cu_2O layer developed by the SILAR technique can be an inexpensive alternative to organic buffer materials and a potential buffer electrode towards the development of efficient organic solar cells.

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CRediT Authorship Contribution Statement

Asmita Patra: Conceptualization, formal analysis, data curation, writing—original draft, writing—review and editing. Suman Mahata: Data curation, formal analysis, writing—review and editing. Animesh Layek: Funding acquisition, conceptualization, formal analysis, writing—original draft, writing—review and editing.

Conflicts of Interest

Authors whose names are listed immediately below the title of this article certify that they have no affiliations with or involvement in any organization or entity with any financial interest (such as honoraria, educational grants, participation in speakers' bureaus, membership, employment, consultancies, stock ownership, or other equity interest, and expert testimony or patent-licensing arrangements) or non-financial interest (such as personal or professional relationships, affiliations, knowledge, or beliefs) in the subject matter or materials discussed in this manuscript.

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