Enhancing Light Outcoupling in Organic Light-Emitting Devices by Integration of Scattering Electrodes

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Limited outcoupling efficiencies are still a big problem in organic light-emitting devices (OLEDs). In this contribution, a simple and general approach for enhancing the outcoupling efficiency of OLEDs is presented: by using a three-step process, including electrospray deposition of a sacrificial material (step i), thin metal-layer deposition (step ii), and elution of the sacrificial material (step iii), scattering metal structures are fabricated that can be used in cavity OLEDs. The integration into green-emitting vacuum-processed OLEDs yields in an increased luminous efficiency and external quantum efficiency (EQE) by 30% and 27%, respectively.

1. Introduction

Although in organic light-emitting diodes (OLEDs) internal quantum efficiencies up to 100% can be achieved by intelligent harvesting of excitons,[1] the poor light extraction efficiency of less than 20–25% for common devices structures is still a problem that limits the overall performance.[2,3] There are various optical loss channels such as reabsorption of the electroluminescence (EL) in the layer stack,[2,4] surface plasmon polaritons (SPPs),[5–7] or total internal reflection.[14–16] The latter leads to waveguiding in the substrate (substrate modes) or the organic layers (organic modes). In bottom-emitting devices, external outcoupling features on the forward-facing side of the substrate can reduce total internal reflection at the glass/air interface and, therefore, are suitable to extract substrate modes.[8–13] However, if organic modes are to be extracted, the usage of internal outcoupling structures is necessary.[14–16]

One approach to extract light that is trapped in OLEDs due to waveguiding is the integration of periodic structures.[5,17–20] In general, the utilization of such periodic structures with length scales in the same order of magnitude as the optical wavelength has the disadvantage that it causes a wavelength- and angle-dependent outcoupling enhancement. Furthermore, the preparation of such structures requires rather elaborate fabrication techniques. By contrast, the application of nonperiodic, random structures[13–15,21–23] has the potential to enhance the light extraction efficiency without introducing spectral changes and directionality. In this context, several approaches have been reported in the literature, such as spontaneously formed buckles,[14] bioinspired moth’s eye nanostructures,[15] islands or network structures of active OLED layers,[23] or coevaporated scattering materials, among others.[22]

In this study, we describe a new concept for the generation of random internal scattering structures to extract organic modes in OLEDs. By using a three-step process consisting of (i) simple and fast deposition of a sacrificial material by electrospray deposition (ESD)[24–31] onto commonly used ITO (indium tin oxide) substrates, followed by (ii) deposition of a thin silver layer, and (iii) elution of the sacrificial material, a semitransparent scattering electrode is generated. We demonstrate its successful integration into vacuum-processed, bottom-emitting cavity OLEDs, featuring an external quantum efficiency (EQE) increased by ≈30% compared with the flat reference.

2. Results and Discussion

To be of general use, the scattering electrode should be semitransparent, compatible with solution and vacuum-processing, and scalable for large-area processing which is possible using ESD as deposition method by using multinozzle spray setups. In addition, processing should be done prior to the device fabrication and fully independent of successive fabrication steps. In general, ESD is especially well suited as deposition method for our purpose because it is a very simple method that allows fast deposition of a wide range of materials onto conducting substrates (see “Conclusion” section). Furthermore, it allows morphology control of the deposited material film by simply changing the deposition parameters. In the past, we have shown that sprayed active hole-transport materials can be used in OLEDs without deteriorating OLED characteristics.[31]
2.1. Reference OLED Fabrication

For this study, we selected a bottom-emitting cavity OLED because spectral changes (width, intensity, angular dependence) caused by the scattering structure are more pronounced and better to observe and interpret in cavity OLEDs than in the standard devices.

For reference, we fabricated a flat cavity OLED (referred to as REF from now on) with a semitransparent reflective bottom electrode (silver, 30 nm), a thin emission layer (10 nm), and a fully reflective top electrode (Ca/Ag). The layer thickness for the hole-transport layer (HTL) to the electron-transport layer (ETL) was optimized for maximum EQE and the 2nd emission maximum for HTL to avoid pinholes and shorts by using the optical simulation software SETFOS. Please note that for the HTL the thicknesses of the doped and nondoped layers were combined. Simulations were performed assuming (i) an IQE of 1, (ii) a Delta-Dirac distributed emission profile in the middle of the emitting layer, and (iii) an emitter distribution that is isotropic. In the following, these are referred to as “standard simulation parameters.”

According to the simulation (see Figure S1, Supporting Information), the optimized OLED stack would be: ITO (150 nm)//Ag (30 nm)//MoO3 (10 nm)//STTB: F6TCNNQ (100:1; 170 nm)//STTB (10 nm)//TMM004:TEG (S:1; 10 nm)//TMM004 (10 nm)//TPBI (60 nm)//Ca (8 nm)//Ag (150 nm).

The simulated mode distribution in this optimized flat reference OLED yields 1) air mode (outcoupled intensity) 17.1%, 2) substrate mode (SM) 6.3%, 3) organic mode (OM) 14.6%, 4) surface plasmon polariton (SPP) 35.8%, and finally 5) absorption losses 26.2% (see also Table S1, Supporting Information, stack A). Due to the cavity structure, the absorption loss is as high as expected having a semitransparent Ag layer. The combined loss due to waveguiding (SM + OM + SPP) amounts to 56.7%, giving a lot of room for improvement by introduction of random scattering structures.\[2,5,14,32\]

The luminance–current–voltage (LIv) and the emission characteristics of the REF OLED are shown in Figure S2 and S3, Supporting Information. All important electro-optical data are shown in Table 1. The EQE values of all devices, even the reference device, are unexpectedly low for phosphorescent green cavity OLEDs. We believe that this might be due to the two-step evaporation of the bottom Ag layer, which requires removing the substrate from the high-vacuum evaporation chamber into a N2-flooded glove box. This may have resulted in contamination and, as a result, contact issues at the hole-injecting side of the device. The two-step evaporation was done to treat the reference devices the same way as the devices with scattering feature (see later). Our assumption that the low EQE is caused by a poor hole contact is supported by comparing the stack used within this study (see earlier) with a very similar stack recently published by our group with the main difference being at the hole side: glass//ITO//PEDOT:PSS (35 nm)//QUPD (× nm)//STTB (130 nm)//NPB (10 nm)//TMM004 + TEG (12 nm)//TMM004 (10 nm)//TPBI (50 nm)//Ca/Ag. With this stack we could reach EQEs of 7–8% despite being a noncavity OLED.\[31\]

As knowledge of the true (experimental) layer thicknesses is important because of their strong impact on the device performance, we verified the layer thicknesses by comparing the experimental emission spectra as well as the angular distribution with the expectations of the optical model. Slight differences were observed. Fitting simultaneously emission spectra and angular distribution using STTB, TPBI, and semitransparent Ag layer thicknesses as fitting parameters (see Figure S4, Supporting Information) suggests that the real thicknesses in the REF device are: Ag (31 nm)//MoO3 (10 nm)//STTB + F6TCNNQ (179 nm)//STTB (10 nm)//TMM004 + TEG (10 nm)//TMM004 (10 nm)//TPBI (57 nm)//Ca (8 nm)//Ag (150 nm). We refer to this stack as stack B. The differences between intended and real thicknesses are in the expected range of error caused by the evaporation processes. As all devices were fabricated in one process run and the samples were arranged at the same angular positions with respect to the middle of a rotating sample holder to ensure uniform films for all devices, we are certain that thicknesses of all devices are similar and a performance comparison between the different cavity devices is valid.

Using the suggested real thicknesses (stack B) within the simulation, it can be shown that the mode distribution is only slightly changed (<1%) compared with the optimized stack A thicknesses (see Table S1, Supporting Information).

2.2. Fabrication of the Scattering Electrode-Step [i]: Electrospray Deposition

In the following, we describe the fabrication of the scattering electrode. The first crucial step is the electrospray of the

| Table 1. Optoelectronic characteristics of the investigated OLED devices. The following parameters are listed: spray time (t_{spr}), current density (j), brightness (L), and luminous efficiency (η) in forward direction; EQE; quality factor of the cavity (Q = δν/δν_0, with δν being the resonance frequency and δν_0 being the full width at half maximum (FWHM)) at different emission angles; and the optimal outcoupling angle (α_{max}). |
|---|---|---|---|---|---|---|
| t_{spr} [s] | j [mA cm^{-2}] at 5.4 V | L [cd m^{-2}] at 5.4 V | η [cd A^{-1}] at 5.4 V | EQE [%] at 20 mA cm^{-2} | Q | α_{max} [''] |
| 0 (REF) | 9.8 | 635 | 6.5 | 1.26 | 27.4 | 27.0 | 21.4 | 30 |
| 20 | 12.5 | 1025 | 8.2 | 1.60 | 21.8 | 22.2 | 15.6 | 20 |
| 40 | 14.4 | 1229 | 8.5 | 1.49 | 21.4 | 22.1 | 15.3 | 18 |
| 60 | 15.5 | 1222 | 7.9 | 1.37 | 22.7 | 22.7 | 16.0 | 18 |
sacrificial material (step 1). Here, we have to find a suitable material that 1) can be deposited by ESD, 2) allows for the generation of size-dispersive scattering particles (x-, y-, and z-dimensions), and 3) has no negative impact on the device performance due to its too high feature size in z-dimension, which may lead to shorts or other topology-related defects. We expect that the height (z-dimension) should not exceed 100 nm and “sharp” features should be avoided.

After screening different materials systems, we found that a blend consisting of poly(3-hexylthiophene-2,5-diyl) and phenyl-C61-butyric acid methyl ester (P3HT:PCBM) dissolved in o-dichlorobenzene (ODCB) can meet the aforementioned requirements. To simulate the conditions in the actual OLED stack (see earlier), we deposited 30 nm of silver (Ag) on the sprayed material before further investigation. Scanning electron microscopy (SEM) (Figure S5, Supporting Information) and atomic force microscopy (AFM) topography measurements (Figure 1A,B) indicate that during the ESD of the P3HT:PCBM blend two kinds of particles are simultaneously deposited onto the ITO surface: "droplets" with a diameter between 2–15 μm (height, z-dimension: 10–20 nm) and "nanoparticles" with a diameter ranging from 0.1 to 1.5 μm (height, z-dimension: 15–60 nm). Thus, a bimodal size distribution with a large width, respectively, is obtained, i.e., clearly nonperiodic and well suited for scattering visible light. Importantly, the z-dimension of both features is in the acceptable range of 10–70 nm.

The average surface roughness of the electrode surface and, thus, its scattering properties can be conveniently controlled by varying the ESD time (t_{ESD}) at a given concentration of the spray solution, the pump rate, and a given electric field. For short t_{ESD}, formation of individual nanoparticles or droplets is observed. At longer times, complex nanostructures consisting of overlapping droplets and nanoparticles are formed (see AFM: Figure 1B or SEM: Figure S5, Supporting Information).

We used AFM measurements on the Ag capped nanostructures to determine the average surface roughness (R_a) as a function of the spray time. As can be seen in Figure 1C, R_a first increases linearly and finally reaches ≈7 nm for a spray time of 60 s. For t_{ESD} > 60 s, there is no further increase in the average surface roughness; this is expected because, around this time, the surface is fully covered with particles, and longer spray times only increase the deposited layer thickness but not its surface roughness. Therefore, the variation of spray time allows preparing scattering silver layers with a variable average surface roughness between 3 and 7 nm.

Measurements of the diffuse reflection for samples that were prepared using different t_{ESD} show that there is an exponential correlation between surface roughness and diffuse reflection (Figure 1D and Figure S6, Supporting Information). Ultimately, diffuse reflection as high as 17% is achieved at the OLED emitter wavelength (550 nm) for t_{ESD} ≥ 60 s. This time may seem long, but can be shortened by increasing the concentration of the spray solution (here 2.5 mg mL⁻¹) or by increasing the pump rate (here 0.5 μL min⁻¹).

2.3. Fabrication of the Scattering Electrode-Step (ii/iii): Metal Evaporation/Elution of Sacrificial Material

As next step, the sacrificial material (here P3HT:PCBM) used to induce the scattering structure to the metal should be removed before fabricating the OLED stack atop to avoid absorption of emitted light within this layer. This can, in principle, be achieved by washing out the material from underneath the silver layer.

Figure 1. Characterization of scattering electrodes: ITO/sacrificial layer/Ag (30 nm). AFM topography for t_{ESD} of A) 10 s and B) 90 s (area = 80 × 80 μm²). C) Average surface roughness (R_a) determined by AFM as function of t_{ESD}. Red lines are guides to the eye. D) Diffuse reflection at 550 nm as a function of average surface roughness (R_a, data from (C)) for varying t_{ESD} (20, 30, 50, and 60 s). The red line is a guide to the eye.
However, in the case of 30 nm thick silver layers, as used in our cavity OLEDs, rinsing with any solvent has no significant effect on the topography of the metal film because the metal film forms a perfect protection layer. Thus, we modified the aforementioned procedure by first depositing only a very thin silver layer (≤8 nm, step ii); due to dewetting such layer features lots of pinholes which allow penetration of a solvent (here ODCB) and elution of the underlying organics (step iii). The successful removal of the sacrificial material was confirmed by SEM (see Figure 2). Before rinsing, P3HT:PCBM is clearly visible sandwiched by the ITO and the silver layers; by contrast, after rinsing the P3HT:PCBM is missing, and the previous droplet structures have collapsed, leaving behind a buckled metal structure characterized by metal “rings.” AFM measurements (see Figure S7, Supporting Information, bottom) showed the edges of the former droplets to be about 10–30 nm high. Furthermore, the average surface roughness remained in a similar range as prior to removal (3.8 nm after washing compared with 4.1 nm before). As evident from the SEM images, the silver layer is directly connected to the ITO. Therefore, we expect that such a scattering bottom electrode is very well suited for OLED application.

### 2.4. OLED Fabrication with Scattering Electrode

The following part addresses the integration of the novel scattering electrodes into bottom-emitting cavity OLEDs and the comparison to the REF device based on normal, flat electrodes. The complete fabrication process of the OLEDs with scattering electrodes is schematically shown in Figure S8, Supporting Information. We fabricated a series of devices with scattering electrodes varying the increasing spray time $t_{\text{ESD}}$ (20, 40, and 60 s).

In this context, we emphasize that AFM measurements on completed OLED stacks show that the topography of the bottom contact layer is replicated through all subsequent layers and ultimately also observed on the top electrode (see Figure S9, Supporting Information). This is indirect proof that similar layer thicknesses have been achieved everywhere, which is not only expected to be beneficial in terms of preventing leakage currents and shorts, but it also means that all internal interfaces may contribute to light scattering (depending on the difference of the respective refractive indices). Therefore, we expect that the extraction of organic and SPP modes is even more efficient compared with the case that only the bottom electrode would be scattering.

The $LIV$ characteristics of all devices are shown in Table 1 and Figure S2, Supporting Information. The incorporation of scattering electrodes into the cavity OLED stack impacts the $JV$ characteristics. A slight increase in current density compared with the REF is observed, which might originate from the relatively rough surface of the electrodes, specifically from locally increased electric fields and a resulting higher injection rate. It was also reported for comparable OLED stacks with scattering electrodes.$^{[14,17,18]}$

*Figure 2. Removal of sacrificial layer (step iii). Top: illustration of experimental procedure; bottom: SEM images (top row: tilted plane view; bottom row: cross section) of scattering electrodes A) before and B) after rinsing with ODCB. The samples were prepared using a spray time of 30 s. The Ag thickness is 8 nm.*
More importantly, the introduction of our scattering electrodes results in a significant increase in device efficiency. At a spray time of \(\approx 40\) s, an maximum increase in luminous efficiency up to 30\% compared with the non-scattering reference was achieved (6.5 \(\text{cd} \text{m}^{-2}\) \(\rightarrow\) 8.5 \(\text{cd} \text{m}^{-2}\)). The maximum increase of EQE by 27\% was achieved for a spray time of 20 s (1.26 \(\rightarrow\) 1.6\%), whereas 40 s lead to an increase in EQE of only 18\% (see Table 1). As expected, the introduction of the scattering layer leads to spectral broadening (see Figure S3, Supporting Information, left) and, thus, a reduction of the quality factor (Q) of the cavity (see Table 1).\(^{[13]}\) In our case, the Q factor in forward direction is reduced from 27.4 for the planar reference to \(\approx 22\) for the devices with scattering electrodes. It follows also the trend that cavity OLEDs with scattering structures have a less pronounced spectral shift over the emission angle (see also Figure S3, Supporting Information, left).\(^{[14–16]}\) The planar reference shows a \(\Delta \lambda_{\text{max,0}} - 40 = 25\) nm, whereas the devices with scattering electrodes have a \(\Delta \lambda_{\text{max,0}} - 40 = 20\) nm. Angle-dependent measurements of the luminance show an enhancement without promoting specific angles (see Figure S3, Supporting Information, right), as expected using random (non-periodic) scattering structures with a wide size distribution. The angle of emission intensity maximum \(\theta_{\text{max}}\) is shifted slightly toward the forward emission (see Table 1) together with a slight variation in the overall shape.

It has to be mentioned that small changes in Ag thickness also strongly influence the cavity strength, and thus (i) the Q factor, (ii) the spectral shift over angle, and (iii) the extraction efficiency. Using the Ag thickness as the only fitting parameter in our flat OLED allows to simulate similar changes in spectral shift and Q factor as measured within our experiments by introducing the scattering electrodes. To reach similar effects in spectral shift and Q factor, the Ag thickness has to be reduced only from 31 to 28 nm (stack C). However, the simulated maximum enhancement of extraction efficiency by this assumed reduction of the Ag thickness is only around 10\% (cf. Table S1, Supporting Information, stack B and C) and cannot explain the 27\% enhancement reached within our experiments. Thus, here is no doubt that the increased outcoupling is indeed caused by scattering of organic and SPP modes.

3. Conclusion

This study presents a new approach toward improved light outcoupling in OLEDs by integration of a scattering bottom electrode achieved in a three-step process via (i) ESD of a sacrificial material, (ii) thin metal-layer deposition, and (iii) elution of the sacrificial material. In green-emitting vacuum-processed cavity OLEDs, the luminous efficiency and EQE could be increased by 30\% and 27\%, respectively. These values could be reached because in the devices all layers from the top to the bottom contact feature the rough (and thus scattering) structure, which is very beneficial for extraction of OM and SPPs.

This new approach is also suitable for top-emitting cavity OLEDs. In this case, as we are independent of the transparency of the sprayed material, the process could be additionally simplified because the rinsing step would be obsolete. In the case of the bottom-emitting OLEDs as used in this study, the rinsing step would be obsolete as well and the process straightforward, if the sacrificial material would not be absorbing in the wavelength region of the OLEDs emission, but having a high enough conductivity.

In this study, the costly material P3HT:PCBM was used as sacrificial material because it turned out to be ideally suited for our purpose after having screened the typical organic electronics (OE) materials that we use usually in our lab. However, as the material is washed out in step (iii), one could use a wide range of other materials that can be processed by ESD (the spray solution has to have a conductivity in the range of \(10^{-4} - 10^{-2}\) \(\text{S} \text{m}^{-1}\), and the solution needs to have electrochemically chargeable components).

Finally, the approach might be beneficial for the fabrication of organic solar cells as well, improving the light coupling into the absorption layer.

4. Experimental Section

Sample Preparation: The sample preparation is schematically shown in Figure S8, Supporting Information.

(Step 0) ITO-coated glass sheets with a sheet resistance of \(\approx 15\ \Omega\ \text{m}^{-2}\) were commercially obtained from Thin Film Devices. The ITO sheets were cut into \(1\ \text{in}^2\) sized substrates and structured via reactive etching. Subsequently, the substrates were carefully cleaned with different solvents in an ultrasonic bath, washed with deionized water, and finally dried in a nitrogen flow.

(Step 1) To reduce the effective height of the edge between substrate and the ITO electrode and to prevent shorts at the edges upon operation, a photoresist (\(N,N'-\text{bis}[4-(6-[(3-ethyloxetan-3-yl)methoxy]-hexyloxy)-phenyl]N,N'-\text{bis}(4-methoxyphenyl)biphenyl-4,4'-diamine + 2 wt\% 4-octylxyloxyphenyldioonium hexafluorantimonate)\(^{[13]}\) was deposited on the substrate via spin coating. The resist was selectively cross-linked by means of photolithography using a Süss Microtech maskaligner. After removing non-cross-linked residues, the substrates could be used for ESD.

(Step i) To generate the scattering nanostructures by ESD, a spray solution consisting of a P3HT:PCBM mixture in \(\text{ortho-dichlorobenzene} (c = 2.5 \text{ mg mL}^{-1})\) was pumped using a PicoPlus syringe pump (Harvard Apparatus) with a constant flow rate through a polycrystalline-coated fused silica capillary to the spray capillary (Unimed, C26 capillary). The spray capillary was connected to the fused silica capillary using PEEK connectors (MicroTight). The spray capillary was connected to a HCP 1.25M-12500 high-voltage power supply (FuG). To stabilize the spray mist, a corona ring was installed in plane with the capillary tip. For the generation of nanoparticles, 2 and 5 kV were applied. Pump rates 0.2–2 and 2 \(\mu\text{L min}^{-1}\) were used. The distance between the capillary tip and the substrate was 7 cm.

(Step ii) Next, 8 nm silver were deposited using a K. J. Lesker spectros evaporator. The base pressure during thermal evaporation was \(2 \times 10^{-6}\) mbar.

(Step iii) Subsequently, the coated substrates were submerged in ODCB for 2 h.

(Step 2) After removing ODCB residues in vacuum at 25 °C for 24 h, a second evaporation step was conducted to obtain a silver layer with a thickness of 30 nm in total.

(Step 3) The vacuum-processed OLEDs were fabricated by sequential thermal evaporation of the respective small-molecule materials in a K. J. Lesker spectros evaporator. The base pressure during thermal evaporation was \(2 \times 10^{-7}\) bar. Additional organic materials that are not explained in the text were \(F4TCNQ (2,2'-\text{perfluoronaphthalene}-2,6-diyldiene)dimalononitrile)\), \(TMM004 (\text{bis}(9,9'-\text{spirobi} \text{fluorene}-2,2'-\text{yl})\) phenylphosphanoxide), and \(\text{TEG} \ (\text{iridium}^{(III)}(2-(2',5'\text{-dimethylphenyl}-3-yl)pyridine)).\)
LIV Characteristics: Measurements of the LIV characteristics were performed with a Keithley 2004 source meter. The luminance values were measured simultaneously with a calibrated photodiode. The applied bias voltage was varied in steps of 0.2 V. The photodiode was calibrated with a luminance meter (Minolta CS-100 chroma meter).

EL Spectra in Forward Direction: EL spectra were recorded using an OCEAN OPTICS SD2000 dual channel fiber optic spectrometer, calibrated with an OCEAN OPTICS LS-1-CAL light source. Automatic measurements recorded the EL spectra within the desired voltage range in 0.2 V steps. Due to the relatively low sensitivity, at least a brightness of 100 cd m\(^{-2}\) is required to obtain spectra with acceptable signal to noise ratios.

Angle-Dependent Light Distribution: The angle-dependent light distribution was measured using a Hamamatsu C9920-11 brightness light distribution characteristics measurement system. The devices were placed in a sample holder on a rotating stage. The OLEDs were operated using a Keithley 2004 source meter, and the emitted light was measured with a CCD-spectrometer (C10027 Photonic Multichannel Analyzer, Hamamatsu) after passing through a dedicated condensing lens and a fiber. The emission angle was varied in steps of 2°.

EQE: The EQE values were calculated based on the angle-dependent light distribution, and the EL spectra were recorded under different emission angles. The absolute brightness value at 0° was obtained from the LIV measurement.

Optical Simulations: Optical simulations were performed using SETFOS (Fluxim) 4.6.7. The n and k values were provided by Fluxim or determined using ellipsometry.

Measurement of Diffuse Reflectance: The diffuse reflectance was measured using a LAMBDA 1050 UV/vis/NIR pectrophotometer (Perkin Elmer) with integrated integrating sphere. The sample was placed in the back of the sphere and angled at 8°. For measurement of the total reflectance, the incident light is reflected through sample and collected by the sphere. The measurement of the diffuse reflectance requires that the specular component is excluded. This can be achieved by allowing the specular component to exit the sphere through the open specular port.

AFM: The AFM images of the scattering electrodes were measured using an SPM SmartsPM7TM-1000 (AIST-NT) in a nitrogen atmosphere. All AFM measurements were performed in noncontact mode.

SEM: SEM images were taken using a Zeiss Supra 40 VP field emission scanning electron microscope. Cross sections were prepared using a Jeoult SM-09010 argon ion cross section polisher.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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