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A Simulation-Based Investigation of Cu_2ZnSnS_4 Solar Cells with Graphene as a Window Layer

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Abstract: *Graphene exhibits great mechanical, electrical, and optical properties, making it an imperative material to use as a window layer in a thin-film solar cell. The efficiency and fill factor of the Cu_2ZnSnS_4 (CZTS) solar cell are increased owing to the incorporation of graphene. Numerical modeling and Solar Cell Capacitance Simulator (SCAPS)-1D simulation have been explored on an efficient CZTS based thin-film solar cell via simulating the proposed structure (Graphene/ZnO/CZTS/Ni) to obtain a nature-friendly solar cell. Besides, the effects of the absorber layer's properties on cell performance parameters have been analyzed. In the proposed cell, the number of layers has been optimized using highly transparent graphene to reach enough sunlight intensity to the absorber layer and back contact. This paper also revealed the effect of temperature and found higher stability in the proposed cell. The optimized cell attributes V_{oc}=0.85 V and J_{sc}=23.815 mAcm^{-2}, FF=84.68%, η=17.14%.*

Keywords: CZTS solar cell; Graphene; SCAPS simulation; Thermal Stability; Window layer.

1. INTRODUCTION

Energy is the source of our existence. However, people are experiencing a crisis of energy owing to the accretion of global energy demand. To combat the energy crisis, researchers have suggested shifting from non-renewable energy to renewable energy. Among all the renewable energy sources, solar energy is a growing source of energy for the generation of electrical power. In the field of energy harvesting, solar energy can be the most efficient, reliable, and abundant source, which deals as a substitute energy resource for ever-growing power demand [1,2]. Moreover, solar energy is clean, cost-effective, environment-friendly, and sustainable energy resource.

A study has reported that in comparison with all other energy sources, the demand for renewable energy grows by ~1% from 2019 [3]. Solar PV is the fastest-growing of all renewable energy sources in 2020. Though silicon-based photovoltaic cells are proposed to be first-generation solar cells with 25% power conversion efficiency [4], the efficiency degrades gradually at a certain rate than thin-film solar cells. Also, it has a low absorption cross-section and high manufacturing and installation cost. Cadmium Telluride (CdTe), quaternary semiconductor material $Cu_2In_xGa_{1-x}Se_2$ (CIGS), and Cu_2ZnSnS_4 (CZTS) have been regarded as polycrystalline second-generation thin-film solar cells (TFSC). CdTe and CIGS-based solar cells have exhibited 20% to 22% conversion efficiency; however, they contain rare material indium (In) and tellurium (Te), and toxic material cadmium (Cd) [5].

Alternatively, CZTS solar cells possess promising features such as abundant in nature and cost-effective, containing non-toxic zinc (Zn) and tin (Sn). Thin-film of the kesterite compound CZTS absorber layer has excellent optical properties, tunable bandgap energy of 1.4 to 1.5 eV, and a higher absorption coefficient (>10^4 cm^{-1}) for which it can be considered a potential material to use as an absorber layer [6]. Afterward, the investigation has been done by adding different layers on CZTS thin-film solar cell to explore its potentiality. Scientists of UNSW have reported that the efficiency of CZTS solar cells is 10% [6]. Shockley et al. have reported the limiting value of conversion efficiency of CZTS solar cells is 32.2% [4], indicating an excellent opportunity to increase the efficiency of CZTS solar cells.

At the front end, solar cells generally use a transparent window layer to obtain higher conversion efficiency and serve as a metal contact that allows light to pass so that higher transmittance is to be achieved. CZTS solar cell with i-ZnO/MoS_2/CZTS/Mo and FTO/i-ZnO/CdS/CZTS/SnS/Mo/SLG structure displayed efficiency of 17.03% and 15.86%, respectively [7]. In the first structure, i-ZnO is used as a window layer, whereas Fluorine doped Tin Oxide (FTO) for the second structure. The efficiency of the second structure has improved to 16.34% with the Indium doped Tin Oxide (ITO) window layer and 16.27% for Aluminum doped zinc oxide (AZO) [8]. However, solar cells with an FTO window layer cause a rise in the leakage current density due to their deformation [9]. On the other hand, Indium tin oxide (ITO) is the most widely used as a window layer; however, it comprises the rare material indium (In) and associates with costly fabrication processes [10]. Therefore, a new material, graphene nanoplatelets (GnP), has been explored attributes superb mechanical, electrical, and optical properties. These features make it an important consideration as a window layer in the coming years to enhance the stability and efficiency of thin-film solar cells.

Graphene is a two-dimensional material with a honeycomb structure having high conductivity and high intrinsic carrier mobility (20000 cm^{2}V^{-1}s^{-1}) and tunable work function (≥5 eV) [11,12]. Also, graphene sheets display a flexible atomic layered structure and are non-brittle, indicating high durability [13]. It is abundant in nature which made it a relatively inexpensive material. The bandgap of GnP is nearly zero signifying a lightweight material [14]. Besides, graphene has a low light absorption coefficient of 2.3%, reflecting it as a transparent material [13]. Studies have shown that doped graphene with B or Cu can change one absorbed photon of a few electrons, increasing the overall performance of solar panels [15].

In this work, graphene is used as a window layer in CZTS solar cells. A thorough investigation of the numerical parameters of graphene has been performed to bring out an efficient, environmentally friendly solar cell. Solar device simulator SCAPS (1-D) is used to execute the numerical simulation of CZTS based solar cells by varying different layer properties. The following subsections describe the working procedures, simulation environment, and relevant results. The proposed structure with graphene as window layer displayed a significant improvement in the solar cell parameters (V_{oc} = 0.85 V, J_{sc} = 23.815 $mAcm^{-2}$, FF = 84.68%, and η = 17.14%).

2. METHODOLOGY

2.1 Device modeling

Solar radiation has different photon energies, and these energies can be converted into electrical energy via integrating layers of different bandgap materials. Material's bandgap should be significant to absorb the photons of higher energy. That's why a higher bandgap layer is used at the front. Usually, two types (heterojunction and multi-junction) of solar cell structures are widely implemented. In this work, the proposed structure is based on a heterojunction solar cell, as shown in Fig. 3.

2.2 Window layer

The transparent conducting oxide (TCO) layer is used as a window layer in optoelectronics and photovoltaic devices made of doped metal oxide. They possess greater than 80% transparency, higher than 10^3 Scm^{-1} of conductivity, and the least carrier concentration of 10^{20} cm^{-3} [16]. Here, graphene is selected as a window layer since it has attractive optical and electrical properties, high conductivity, and transparency (>90%) [17]. Moreover, it provides higher thermal stability than other conventional conducting oxide layers. However, studies have shown that with the increase of thickness of graphene, transparency of graphene window layer decreases [17]. Also, the bandgap of multilayer graphene can be tuned to a value greater than 0.2 eV [18–20].

2.3 Buffer layer

The buffer layer (single or double) is employed to create a PN junction with the absorber layer. A more comprehensive bandgap layer should be used as a buffer layer to ensure maximum light transmission, minimum absorption loss, and minimum recombination loss, i.e., transporting the maximum amount of photogenerated carriers to the outer circuit. It also has an optimum thickness to ensure low series resistance. This buffer layer plays a crucial role in enhancing the open-circuit voltage (V_{oc}) in the solar cells [21,22]. In this work, zinc oxide (ZnO) is chosen as a buffer layer material. An n-type semiconductor with a wide direct bandgap of 3.3 eV improves the solar cell performance by reducing absorption loss [23]. Also, ZnO is eco-friendly and highly abundant. In addition, the CZTS-ZnO interface has excellent lattice matching, which is another crucial parameter for an efficient solar cell [22].

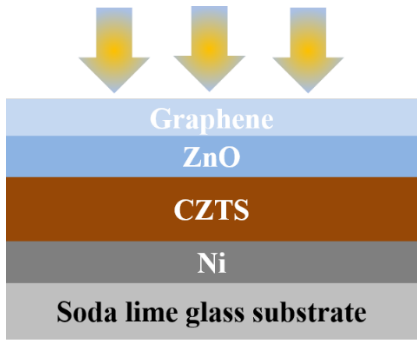

Fig. 3. The proposed structure.

2.4 Absorber layer

An absorber layer is another important and integrated part of a solar cell, which absorbs sunlight or artificial light energy. Most of this light energy is in the visible region of the electromagnetic radiation, and an efficient absorber layer should absorb these radiations at those wavelengths. In the proposed structure, Cu_2ZnSnS_4 (CZTS) thin film is considered as an absorber layer because it exhibits good physical and electrical properties (bandgap around 1.4 to 1.6 eV) and an attractive absorption coefficient (>10^4 cm^{-1}) [24]. Moreover, they are nature-friendly, earth-abundant, and exclusively offer an efficiency of more than 20% [9].

2.5 Back contact

The back contact has a significant role in increasing solar cell efficiency as well as performance parameters. Nickel (Ni) as a back contact shows a high performance than others. It has a work function of 5.15 eV [25], which is defined as the energy required to extract electrons from the metal surface. A stable ohmic contact can reduce the back-contact interface recombination, which significantly improves the device's performance. CZTS has an electron affinity of 4.5 and a bandgap of 1.5 eV [26]. Thus, a higher work function metal is obligatory for a static ohmic contact, and Ni does provide the same.

2.6 Soda-lime glass (SLG) substrate

Glass substrate constituents are also essential parts of a thin-film solar cell. Diffusion of Na from SLG substrate to the grain boundaries of the absorber layer prevents the electron-hole recombination at the grain boundaries. That's why alkali metal oxides (Na_2O, K_2O) have been characterized and synthesized experimentally [27]. This SLG substrate is extensively used for thin film deposition, which is smooth and provides mechanical support to the thin-film solar cell. It is chemically stable and comparatively low in price, making it highly workable in solar cell research [28].

2.7 Simulation

SCAPS (a Solar Cell Capacitance Simulator) is a one-dimensional solar cell simulator that allows the simulation of up to seven layers, developed by Burgleman et al. [29]. SCAPS-1D provides fast simulation and batch calculations. Numerical modeling and simulation are mandatory before the fabrication process to ascertain the performance and stability of the proposed cell. The performance of the cell is defined by the layers' simulation parameters. So, by varying different parameters of a layer, such as the thickness and doping density of the absorber layer, the cell structure is analyzed. Moreover, the effect of temperature is investigated to evaluate the cell's durability and thermal stability. The proposed structure encompasses three layers which are simulated via the SCAPS3309 tools. The electrical and optical parameters of the materials are shown in Table 1, which are obtained from the works of literature [9,13–15,17,30–33] for reasonable estimation.

Table 1. Parameters used for different layers

Parameters	GnP	ITO	ZnO	CZTS
Thickness (nm)	2	60	100	2000
Energy bandgap (eV)	1.80	3.50	3.35	1.52
Electron affinity (eV)	3.92	4.00	4.35	4.50
Relative permittivity, ε_r	10	10	10	10
Density of states in CB, ($\times10^{18}$ *cm*$^{-3}$*)*	1000	2.2	2.0	2.2
Density of states in VB, ($\times10^{19}$ *cm*$^{-3}$*)*	100	1.8	1.8	1.8
Electron thermal velocity ($\times10^{7}$ *cms*$^{-1}$*)*	5.2	1.0	1.0	1.0
Hole thermal velocity ($\times10^{7}$ *cms*$^{-1}$*)*	5.0	1.0	1.0	1.0
Electron mobility (cm^{2}*v*$^{-1}$*s*$^{-1}$*)*	1.0×10^{9}	50	25	100
Hole mobility (cm^{2}*v*$^{-1}$*s*$^{-1}$*)*	10	75	100	20
Doping density ($\times10^{18}$ *cm*$^{-3}$*)*	9000	0.1	1.0	0.01

3. RESULTS AND DISCUSSION

3.1 Effect of absorber layer thickness

It is vital to investigate the absorber layer thickness to preserve the absorber layer materials and reduce manufacturing costs. Thickness is varied from 0.5 µm to 4.0 µm, and the optimized thickness is selected at 2.0 µm. The effect of absorber layer thickness without graphene is investigated for all the four parameters: open-circuit voltage (V_{oc}), short circuit current density (J_{sc}), fill factor (FF), and power conversion efficiency (η) and presented in Fig. 4. Since graphene improves the solar cell performance, the structure without GnP requires a thicker absorber layer to produce the same output parameters as with graphene inclusions. Alternatively, performance variation with thickness in the presence of graphene is displayed in Fig. 5 for all the four parameters (V_{oc}, J_{sc}, FF, and η). This figure shows that all these parameters of solar cells with graphene nanoplatelets (GnP) inclusions are increasing distinctly with increasing absorber layer thickness. For the optimized thickness of 2.0 µm, solar cells with GnP shows better performance than solar cells without GnP.

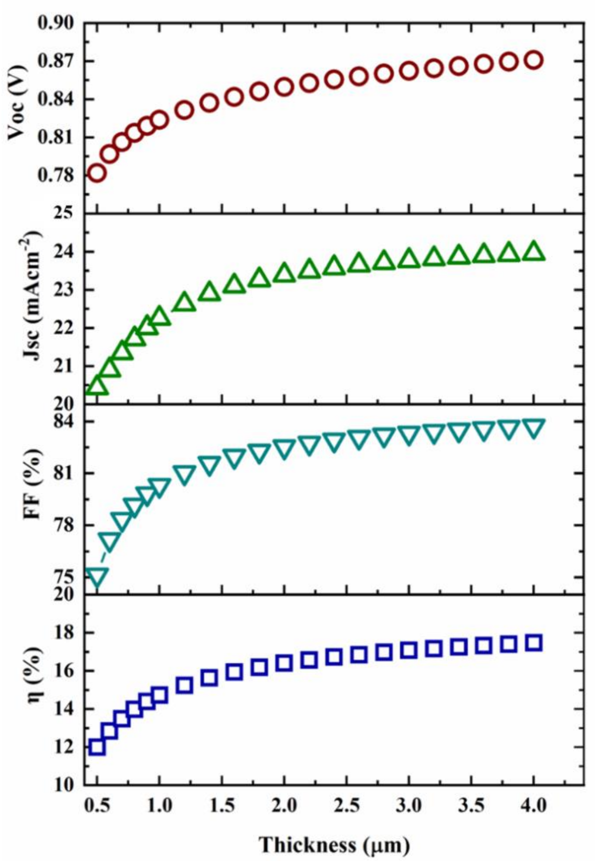

Fig. 4. Performance parameters vs. CZTS layer thickness without graphene.

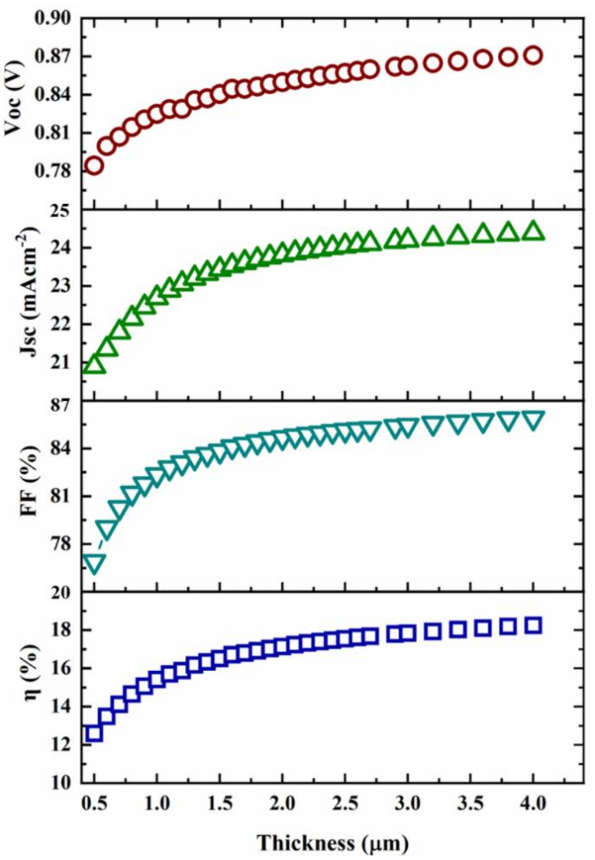

Fig. 5. Effect of CZTS layer thickness with graphene.

3.2 Doping concentration's effect in absorber layer

Further investigation is carried out to observe the effect of doping concentration using the SCAPS simulator. Doping density varies from 1×10^{11} cm^{-3} to 1×10^{18} cm^{-3}, and the results are shown in Fig. 6. It is found that V_{oc} is increased while J_{sc} is decreased with the increasing doping concentration of CZTS, indicating their dependence on doping density. This dependent relationship can be explained by Eq. 1.

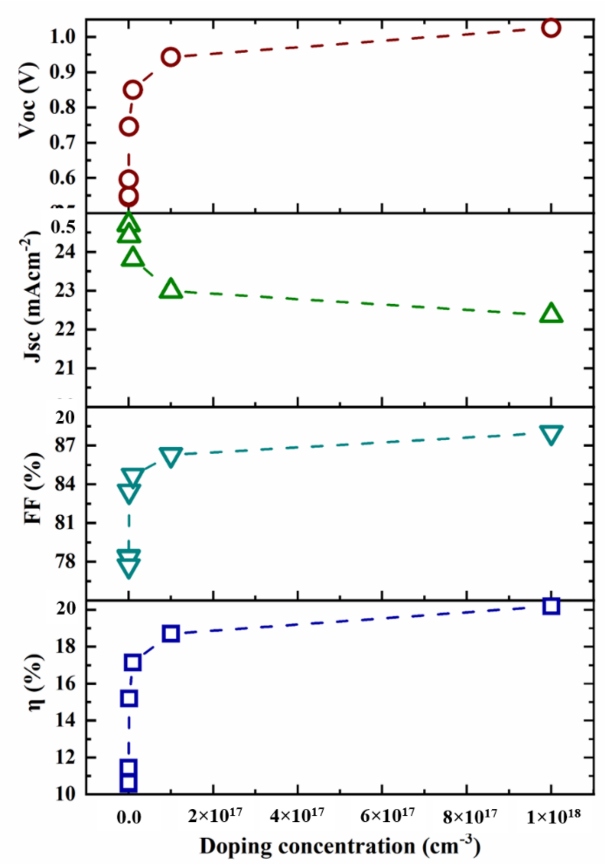

Fig. 6. Effect of CZTS doping concentration.

$$V_{oc} = \frac{kT}{q} \ln\left(\frac{I_L}{I_0} + 1\right) \quad (1)$$

Where k is Boltzmann constant in JK^{-1}, T is the temperature in K, q is the electronic charge in C, I_L is the photogenerated current in mA and I_0 is the diode saturation current in mA. It reveals that V_{oc} is inversely proportional to I_0. With increasing doping concentration, I_0 decreases; hence, V_{oc} increases. On the other hand, increasing doping density also increases the recombination profile, resulting in reduced J_{sc} with increasing absorber layer concentrations [6].

3.3 Effect of temperature

Temperature plays a major role in solar cell performance. Here, 300 K (~27 °C) is considered as the operating temperature. However, practically solar cells are placed outdoor where the temperature may change, causing unexpected output. For instance, excessive heat may deteriorate solar cell performance. The simulation is performed with temperature variation from 280 K to 400 K to explore the effect of temperature, and simulation results are reported in Fig. 7. This figure shows a decreasing trend of the output parameters. However, the output does not change significantly. So, it can be concluded that using graphene as a window layer increases the solar cell thermal stability [34].

3.4 Effect of graphene inclusions on QE

Quantum efficiency (QE) is the ratio of the number of carriers accumulated to the number of photons incident on a solar cell. It is expressed as a function of wavelength or energy and is represented in Fig. 8. It shows that by adding graphene, QE has been increased to approximately 90%. Since graphene increases the transparency resulting more photons are absorbed by the absorber layer, which in turn improved the output.

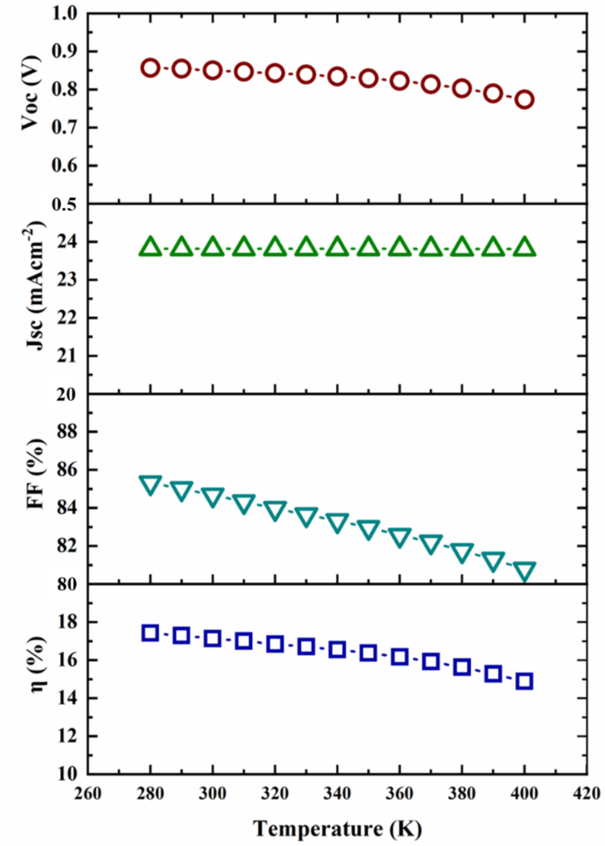

Fig. 7. Performance parameters versus temperature.

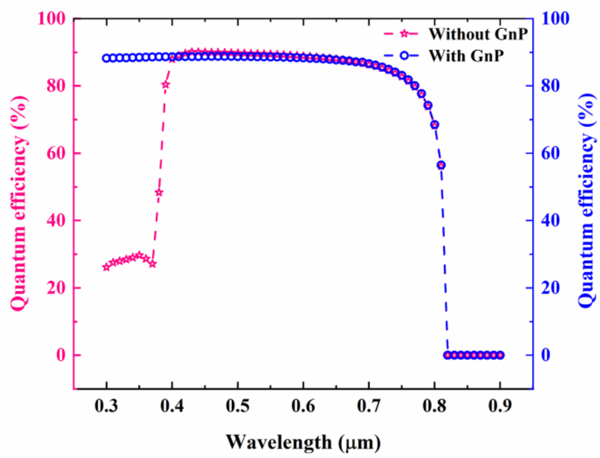

Fig. 8. QE characteristics with and without graphene.

3.5 Comparison of J-V characteristics

Fig. 9 shows that both J_{sc} and V_{oc} have increased due to the incorporation of graphene as a window layer. These results are promising to achieve a highly efficient solar cell. Moreover, the performance parameters for the cell with i-ITO as a window layer in the ITO/ZnO/CZTS structure [9] have been compared with the proposed structure (GnP/ZnO/CZTS) and subsequently presented in Table 2. A significant improvement in the conversion efficiency is achieved successfully.

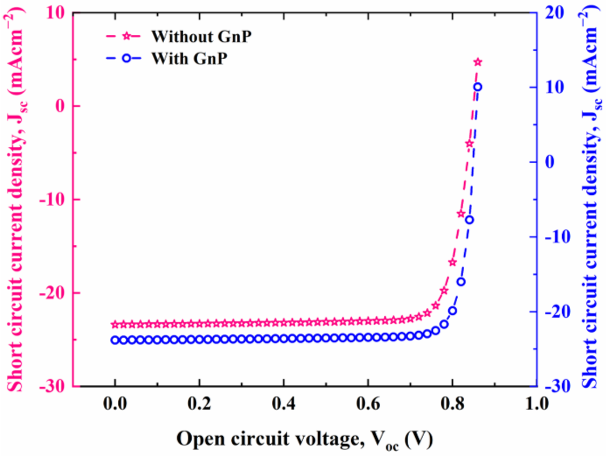

Fig. 9. J-V characteristics with and without graphene.

Table 2. Comparison of the performance parameters.

Structures	V_{oc}	J_{sc}	FF	η
	V	*mAcm*$^{-2}$	*%*	*%*
ITO/ZnO/CZTS	0.8496	23.39	82.5	16.4
GnP/ZnO/CZTS	0.8500	23.81	84.7	17.1

4. CONCLUSIONS

CZTS based solar cell has recently drawn the attention of researchers for its attractive optical, electrical properties and low cost. In this work, the effect of graphene inclusions as a window layer is explored extensively. The incorporation of graphene enhances the solar cell performance with the open-circuit voltage, short circuit current, fill factor, and efficiency as 0.85 V, 23.815 mAcm^{-2}, 84.68%, and 17.14%, respectively. Graphene addition also reduces the absorber layer thickness up to 0.2 µm. Moreover, the performance analysis with temperature variation reveals that the proposed cell is thermally stable.

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