

High-Temperature Syngas Desulfurization and Particulate Filtration by ZnO/Ceramic Filters

T.-C. Wang, Ling-Wei Wei, H.-L. Huang, Kuen-Song Lin,* and H. Paul Wang*



ABSTRACT: Combustible gas (e.g., gasification syngas) cleaning at high temperatures can obtain further gains in energy efficiency for power generation and importantly leads to a simplified process and lower cost as a commercially viable source of clean energy. Thus, a feasibility study for high-temperature desulfurization (HTDS) and additional high-temperature particulate filtration (HTPF) of a raw syngas using ZnO sorbent-dispersed Raney CuO (ZnO/R-CuO) and ceramic filter (ZnO/CF) has been carried out. By synchrotron X-ray absorption near-edge structure (XANES) spectroscopy, mainly Zn(II) and Cu(II) are found in the ZnO/R-CuO sorbents. Both ZnO and R-CuO in the sorbents are involved in HTDS (1% H₂S) at 873 K to form ZnS, Cu₂S, and a small amount of CuS and reach relatively high HTDS efficiencies (82–90%). In addition, regeneration of the sulfurized sorbent by oxidation with O₂ at 873 K (HTRG) for 1 h can restore ZnO and CuO for continuous and repetitive HTDS-HTRG cycles. To facilitate the HTDS engineering applications by the ZnO/R-CuO



sorbents, their reaction rate constant ($8.35 \times 10^4 \text{ cm}^3/\text{g/min}$) and activation energy (114.8 kJ/mol) at 873 K have also been determined. Furthermore, the ZnO/CF sorbent/filter can perform HTDS and additional HTPF at 873 K with very high particulate removal efficiencies (>98%). This demonstrates the feasibility for hot-syngas cleaning with a much better energy efficiency and lesser cost for cleaner power generation.

1. INTRODUCTION

An integrated gasification combined cycle (IGCC) for clean electricity generation exhibits a relatively high efficiency and low environmental pollution over conventional coal-fired power plants.^{1,2} Before feeding the gasification synthetic gas (syngas) to a gas turbine for electricity, sulfur and particulates therein must be reduced. Sulfur species such as H_2S in a raw syngas are conventionally desulfurized by wet chemical absorption at relatively low temperatures (<400 K).³

Although IGCC with cold-gas desulfurization for gas cleanup has generally better effectiveness than the traditional coalfired boilers, further gains in energy efficiency are possible if syngas can be cleaned up at high temperatures.^{4–6} Accordingly, high-temperature desulfurization (HTDS) has many advantages such as elimination of the need and expense for equipment of cooling hot-gas streams prior to desulfurization, which can lead to a simplified process and thus improve the IGCC thermal efficiencies.⁷ Generally, a 3% reduction in the heat rate, a 6% decrease in capital cost, and a 6% decrease in the levelized cost of electricity could be achieved if HTDS was carried out.⁸

ZnO is an effective desulfurization sorbent for removal of <10 ppm sulfur in the flue gas.⁹ CuO sorbents have also been frequently used for desulfurization.¹⁰ Raney metal oxides have a skeletal, sponge-like structure with very high surface areas.

Raney catalysts generally show high activities in dehydrogenation due to the presence of relatively high surface active species.¹¹ In addition, Raney catalysts have a low initial cost per unit mass of metal and the lowest ultimate cost per unit mass in catalysis applications. Better resistance to catalytic poisoning with Raney catalysts has been found.¹² For example, Raney copper and iron have often been used in selective hydrogenation of aldehydes or esters to alcohols and dehydrogenation of alcohols to aldehydes or ketones.¹³ Raney copper or iron can also be used in a wide range of gas- and liquid-phase hydrogenation, dehydrogenation, and water-shift processes.¹⁴

In addition to sulfur (H_2S), particulates are frequently found in the combustible off-gas streams. Particulates may cause plugging and abrasion of downstream piping and equipment during combustion in gas turbines or internal engines.^{15,16} Desulfurization prior to combustion of combustible gas is also

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critical as H_2S therein is converted to SO_x (during combustion), which may also contribute to the formation of secondary inorganic aerosol gases and fine particulates.¹⁷ Ceramic filters (CFs) with high thermal resistance can be used in the removal of particulates from off-gases at T > 673 K. Rigid ceramic filters have also been practical in combustion and gasification of fossil fuels and biomass for environmentally clean power generation.¹⁸ Direct applications of high-temperature particulate controls with CFs are expected to be beneficial to the advanced fuel processing technologies.¹⁹

Effective sorbents like ZnO dispersed on porous CFs can accomplish the goal of simultaneous HTDS and high-temperature particulate filtration (HTPF) in a single reactor. Thus, the main objective of this work was to study the feasibility of HTDS and HTPF using the novel ZnO sorbent dispersed on CFs for high-efficiency power generation processes. In addition, the main scientific issues concerning speciation of select elements ultimately depend on their molecular-scale structures.²⁰ Accordingly, speciation of the ZnO-based sorbents after the hot-gas clean-up treatments was investigated by extended X-ray absorption fine structure (EXAFS) and near-edge structure (XANES) spectroscopy for a better understanding of the zinc species interconversion during the repetitive desulfurization, particulate filtration, and regeneration processes.

2. MATERIALS AND METHODS

2.1. Preparation of Sorbents and Filters. Raney copper oxide (R-CuO) was prepared by the oxalate gel coprecipitation method. Briefly, copper nitrates (Fluka) (0.1 M) and oxalic acid (Bake) (0.1 M, >20%) were dissolved in ethanol and mixed well at 298 K. The precipitates were separated by a centrifuge, dried at 283 K for 8 h, and calcined at 873 K for 2 h. The ZnO/R-CuO sorbents (ZnO: 20 wt %) were prepared by impregnation of $Zn(NO_3)_2$ (Fluka) (0.3 M) onto R-CuO at 298 K. The sorbents were dried at 378 K for 8 h and calcined at 873–1073 K for 2 h.

For simultaneous HTDS and HTPF, ZnO $(Zn(NO_3)_2 (Fluka))$ was coated onto a ceramic filter (CF) (Cerafil XT-650) at 298 K. The ZnO/CF adsorbent/filter (ZnO: 20 wt %) was dried at 378 K for 8 h and calcined at 873 K for 2 h prior to HTDS and HTPF.

2.2. High-Temperature Desulfurization and Particulate Filtration. The HTDS, HTPF, and high-temperature regeneration (HTRG) experiments by ZnO/R-CuO and ZnO/CF were carried out using a vertical fixed-bed reactor (OD 1" × 16"), which was operated at the temperatures between 873 and 1073 K. The ZnO/R-CuO sorbent (1.5 g) was packed in the fixed-bed reactor. A simulated syngas (40% N₂, 20% H₂, 20% CO, 19% CO₂, and 1% H₂S) at a flow rate of 50 mL/min (determined by digital flow rate meters) entered from the top of a fixed-bed reactor at 873–1073 K for HTDS. After HTDS, HTRG with O₂ (50 mL/min) was carried out at 873 K. The volume space velocity for the HTDS and HTRG reactor was 6000 h⁻¹. An *in situ* FTIR (Bio-Rad FTS-40) gas cell was used to monitor the off-gas from the HTDS and HTRG reactions.

Phosphor particulates (<10 μ m) (50 mg/m³) (obtained from electronic waste-recycling plants) carried by the simulated syngas containing 1% H₂S (flows: 50–200 mL/ min) were fed into the reactor. About 1.5 g of the ZnO/CF sorbent/filter was used for HTDS as well as HTPF at 873– 1073 K. After HTDS, HTRG by oxidization with O₂ (50 mL/ min) was carried out at 873-1073 K. The volume space velocity (STP) for HTDS and HTRG was also 6000 h^{-1} . The FTIR gas cell was used to monitor the off-gas from the reactor.

2.3. Characterization of Sorbents and Filters. The crystalline structure of the ZnO/R-CuO sorbent was determined by X-ray powder diffraction (XRD) (RIGAKU Model D/MAX III-V) with CuK α radiation, which was scanned from 10° to 80° (2 θ) at a scan rate of 3°/min. The FTIR spectra of the off-gases from HTDS, HTRG, and HTPF reactions, recorded on an FTIR spectrometer (Bio-Rad FTS-40) with a computerized data handling system, were determined (64-scan data accumulation with a resolution of 4 cm⁻¹).

Synchrotron X-ray absorption spectroscopy (XAS) spectra, i.e., XANES and EXAFS, of copper and zinc in ZnO/R-CuO and Zn/CF were collected on the Wiggler beamline at the Taiwan National Synchrotron Radiation Research Center (NSRRC). A Si(111) double-crystal monochromator was used for providing highly monochromatized photon beams (energy between 1 and 9 keV). The electron storage ring was operated at an energy of 1.3 GeV (current, 100–200 mA). Data were collected in the fluorescence mode in the region of the Zn (9669 eV) and Cu (8979 eV) K edge at 298 K. The absorption spectra were collected in ion chambers that were filled with argon gas.

The EXAFS spectra for copper and zinc in the sorbents and filters were analyzed on the measured oscillatory structure, which appeared at 50-1000 eV above the absorption edge. The raw absorption data in the region of 50–200 eV below the edge position were fit to a straight line by the least-square algorithms. The fitted pre-edge background curves were extrapolated throughout all data ranges and subtracted and normalized to minimize the effect of sample thickness. The isolated EXAFS data were normalized to the edge jump and converted to the wavenumber scale. Fourier transformation of the scattering curve yielded a radial structure function (RSF). The k^2 -weighted EXAFS spectra were Fourier-transformed to the R space over the range between 2.5 and 11.5 Å^{-1} . The peak positions were 0.3-0.5 Å less than the true distance. The multiple-scattering XAS modeling program FEFF 8 was used in the RSF calculation for the model structure. The EXAFS data were also analyzed using the UWXAFS 3.0 simulative program.²⁰

The XANES spectra of ZnO/R-CuO and Zn/CF extended to an energy of 50 eV above the edge were analyzed with the principal component analysis and least-squares fitting (PCA/ LSF) as a quantitatively analytical tool for speciation of dilute and/or amorphous multicomponents, which was not easily attainable with conventional methods. Semiquantitative analyses of the XANES spectra were conducted by the LSF of linear combinations of the standard spectra to the spectrum of the sample. The XANES spectra of zinc and copper model compounds such as ZnO, ZnS, CuO, and Cu₂S were also measured on the Wiggler beamline.

3. RESULTS AND DISCUSSION

X-ray diffraction patterns of the ZnO/R-CuO sorbents that were calcined at 873–1073 K for 2 h are shown in Figure 1a– c. The sorbents with different pretreatment temperatures (873–1073 K) have a similar diffraction pattern. The intense peaks at 32°, 36.5°, and 34.6° and 35.5° and 38.5° (2θ) for ZnO and CuO,²¹ respectively, are observed, suggesting the



Figure 1. XRD patterns of the (a) ZnO/R-CuO sorbent calcined at (b) 873, (c) 973, and (d) 1073 K for 2 h and (e) sulfurized (with H_2S) at 873 K for 1 h (1: ZnO; 2: CuO; 3: ZnS; 4: Cu₂S; 5, Cu₂O; 6: Cu).

existence of mainly ZnO and CuO in the ZnO/R-CuO sorbent.

Basic understanding at the molecular scale is of great importance and interest in developments of effective HTDS and HTPF materials and methods. Speciation data such as bond distance, coordination numbers (CNs), and chemical identity of elements in complicated sorbent and filter matrixes can be determined by synchrotron EXAFS and XANES. The PCA/LSF XANES data of copper and zinc in the ZnO/R-CuO sorbents that were calcined, sulfurized with H₂S (1%), and regenerated with O₂ are shown in Figure 2. After HTDS at 873 K for 2 h, additional Zn(II) (ZnS), Cu(II) (CuS), Cu(I) (Cu₂S), and Cu(0) (Cu) species are found (Figure 2c), signifying that both ZnO and Raney CuO are involved in HTDS. It should be noted that metallic copper (Cu) is also formed after HTDS. The XANES data in Table 1 show that

Table 1. Fractions of Zinc and Copper Compounds in ZnO/
R-CuO and ZnO/CF after High-Temperature
Desulfurization (HTDS) (with $H_2S(1\%)$) and High-
Temperature Regeneration (HTRG) (with O ₂) at 873 K

treatment	species	ZnO/R-CuO fraction	ZnO/CF fraction
calcined (at 1073 K)	ZnO	1.00	1.00
	CuO	1.00	0
HTDS	ZnO	0.57	0.33
	ZnS	0.43	0.67
	Zn	0	0
	CuO	0	0
	CuS	0.04	0
	Cu_2S	0.55	0
	Cu	0.41	0
HTRG	ZnO	0.81	0.96
	ZnS	0.16	0.04
	Zn	0.03	0
	CuO	0.65	0
	CuS	0.33	0
	Cu ₂ S	0.02	0

ZnS (43%) and Cu₂S (55%) with a small amount of CuS (4%) are yielded from the HTDS of ZnO and R-CuO in the ZnO/ R-CuO sorbents, respectively. Metallic copper (Cu) was generated by self-reduction of R-CuO, which may occur in the reducing environment during HTDS.²²

To have continuous HTDS operations, without changing the reaction temperature, at 873 K, the high-temperature regeneration (HTRG) of the HTDS-treated ZnO/R-CuO sorbent was carried out by oxidation with flowing O₂ (50 mL/min). Figure 2e shows that the HTRG of the sulfurized sorbent at 873 K for 1 h leads to regeneration of ZnO (81%) and CuO (65%) (Table 1) from ZnS, Cu, and Cu₂S (after HTDS) for continuous and repetitive HTDS-HTRG cycles.



Figure 2. Normalized (A) zinc and (B) copper k-edge spectra of the ZnO/R-CuO sorbents calcined at (a) 973 and (b) 1073 K for 2 h, (c) sulfurized (with H_2S (1%)) at 873 K, and (d) regenerated (with O_2) at 873 K for 2 h.

The refined copper and zinc EXAFS data of the calcination-, HTDS-, and HTRG-treated ZnO/R-CuO sorbents are shown in Table 2. The average bond distances of Zn–O (first shell)

Table 2. Speciation Parameters of Zinc and Copper in ZnO/ R-CuO and ZnO/CF after High-Temperature Desulfurization (HTDS) (with H_2S (1%)) and High-Temperature Regeneration (HTRG) (with O_2) at 873 K

absorbent	treatment	shell	R (Å)	CN
ZnO/R-CuO	calcination at 1073 K	Zn-O	1.94	5.6
		Zn-(O)-Zn	3.02	8.6
		Cu-O	1.94	5.3
		Cu-(O)-Cu	2.63	7.2
	HTDS	Zn-S	2.32	3.9
		Zn-(S)-Zn	3.08	10
		Cu-S	2.61	6.6
		Cu-(S)-Cu	2.52	8.3
	HTRG	Zn-O	1.95	3.1
		Zn-(O)-Zn	3.01	7.7
		Cu–O	1.96	1.4
		Cu-(O)-Cu	2.71	3.8
ZnO/CF	calcination at 1073 K	Zn-O	1.94	5.4
		Zn-(O)-Zn	3.21	8.2
	HTDS	Zn-S	2.31	4.0
		Zn-(S)-Zn	3.31	9.6
	HTRG	Zn-O	1.95	4.4
		Zn-(O)-Zn	3.20	8.0

and Zn-(O)-Zn (second shell) in ZnO (calcined at 1073 K) are 1.94 and 3.02 Å with CNs of 5.6 and 8.6, respectively. After HTDS at 873 K, by substitution of oxygen in ZnO with sulfur, the bond distances of Zn-S and Zn-(S)-Zn increase.²¹ The successive HTRG at 873 K leads to restoration of the bond distances and CNs of Zn-S and Zn-(S)-Zn for the next HTDS operations. Nevertheless, after HTRG, increases in the Cu-O (first shell) and Cu-(O)-Cu (second shell) bond distances with decreases in their CNs for the skeletal R-CuO are observed. This is caused by HTDS and HTRG that may cause a slight chemical structure destruction of the skeletal R-CuO.²³

Time dependence for HTDS at 873-1073 K with the simulated syngas containing H₂S (1%) by the ZnO/R-CuO sorbent in a fixed-bed reactor is shown in Figure 3. Little axial dispersion occurs in the HTDS experiments with the ZnO/R-CuO sorbent at 873-1073 K for the elapsed time of 1 h approximately $(t/t_0 = 0.4 - 0.5)$. At high temperatures, ZnO as well as R-CuO is involved in desulfurization to an increasing extent. For example, within 20 min, a high HTDS efficiency (90%) can be achieved. The HTDS kinetics with the ZnO/R-CuO sorbent were studied, assuming negligible mass diffusion and no intermediate.²⁴ Kinetic parameters such as the rate constant (k) and active energy (E_a) were determined by the nonlinear regression of the experimental data. Table 3 summarizes the reaction kinetic parameters for HTDS with the ZnO/R-CuO sorbents. Their HTDS rate constants at 873-1073 K are 8.35×10^4 to 183×10^4 cm³/min·g. Although the HTDS reaction rate at 1073 K (see Figure 3a) is greater than that at 873 K, the higher temperature (i.e., 1073 K) operation may cause a slight chemical structure destruction of the sorbent and the regeneration, on the contrary, prefers to be processed at mild temperatures, e.g., 873 K. Since the elapsed time for HTDS at 873 and 973 K by the sorbent is about 1 h



Figure 3. Time-dependent HTDG efficiencies by the ZnO/R-CuO sorbents at (a) 873, (b) 973, and (c) 1073 K and (d) ZnO/CF with H_2S (1%) in a simulated syngas under a flow rate of 100 mL/min at 873 K for 2 h.

Table 3. High-Temperature Desulfurization (HTDS) Efficiencies (ε) and Reaction Kinetic Parameters at 873– 1073 K for the ZnO/R-CuO Sorbent

	873 K	973 K	1073 K
HTDS ε			
ZnO	0.90	0.85	0.47
CuO	0.82	0.73	0.35
reaction kinetic parameter			
$k (10^4 \text{ cm}^3/\text{min}\cdot\text{g})$	8.35	19.5	183
$E_{\rm a}~({\rm kJ/mol})$	115		
$A (10^{11} \text{ cm}^3/\text{min g})$	7.4		

(Figure 3a,b), it is thus more feasible for HTDS and HTRG operated at 873 K.

To have simultaneous HTDS and HTPF functions of a hot raw syngas in a single reactor, the ZnO sorbent-dispersed ceramic filter (ZnO/CF) was prepared. The PCA/LSF XANES data (Table 1) show that mainly ZnO is present in ZnO/CF for HTDS at 873 K. A relatively high fraction of ZnS (67%) from the HTDS of the ZnO sorbent on the CF if compared to the ZnO/R-CuO sorbent (ZnS (43%)) is also shown in Table 1. The XANES data in Table 1 also indicate that ZnO/CF has a very high HTRG efficiency, and about 96% of ZnO can be regenerated for three continuous HTDS-HTRG cycles. In addition, the molecular-scale data (by EXAFS) of the regenerated ZnO (Table 2) also show that the Zn-O (first shell) and Zn-(O)-Zn (second shell) bond distances and CNs on the CF are reformed in the three-cycle regeneration at 873 K. Consequently, ZnO/CF has a higher HTDS efficiency at the elapsed time of about 1.7 h ($t/t_0 = 0.84$) at 873 K than ZnO/R-CuO (Figure 3).

Table 4 shows the average HTDS and HTPF efficiencies (for three runs) of the CF and ZnO/CF at 873–1073 K under the flow rates of 50–200 mL/min at 873–1073 K. The HTPF efficiencies for the CF and ZnO/CF are very high (>96%). The high efficiencies for the filters are found regardless of the temperature and flow rate ranging from 873 to 1073 K and from 50 to 200 mL/min, respectively. Nevertheless, at T > 973, the HTDS efficiencies decrease (<55%) with a slight decrease in the HTPF efficiencies. While the ZnO/R-CuO sorbent has relatively high HTDS efficiencies, the ZnO/CF sorbent/filter

Table 4. Efficiencies (ε) of High-Temperature Desulfurization (HTDS) and Particulate Filtration (HTPF) by ZnO/CF at 873–1073 K

filter	T (K)	HTDS ϵ	FR (mL/min)	HTPF ε	FR (mL/min)	HTPF ε
CF	873		100	0.98	50	0.99
	973		100	0.98	100	0.98
	1073		100	0.98	200	0.97
ZnO/	873	0.63	100	0.99	50	0.98
CF	973	0.55	100	0.98	100	0.97
	1073	0.30	100	0.98	200	0.96

can simultaneously perform HTPF at 873-1073 K for 1 h to reach high particulate removal efficiencies (>96%) (Table 4). Table 5 shows the efficiencies for desulfurization and

Table 5. Efficiencies (ε) of Desulfurization (DS) and Particulate Filtration (PF) Using Comparable Filters

sorbent/filter	Т (К)	DS e	FR (mL/min)	PF ε	reference
ZnO/CF	873	0.63	50	0.98	this work
Ni/ZnO	573	0.59	100	0.4	25
Ni/ZnO/-SiO ₂ -Al ₂ O ₃	675	0.11	400		26
ZnO/TiO_2	973	0.19	50		27

particulate filtration of hot off-gases using comparable sorbents and filters.²⁵⁻²⁷ It reveals that ZnO/CF has much higher efficiencies for desulfurization and particulate filtration at a high temperature (873 K) than reported sorbents and filters at 573–973 K.

The main chemical reactions in the HTDS processes by ZnO/R-CuO and/or Zn/CF can be more clearly described in Scheme 1. As the syngas containing H_2S (1%) passes through the ZnO/R-CuO and/or ZnO/CF sorbent beds at 873 K, $H_2S_{(g)}$ can be captured by desulfurization with ZnO and CuO

Scheme 1. High-Temperature Desulfurization and High-Temperature Regeneration Processes"





^{*a*}In the high-temperature desulfurization (HTDS) process, as the syngas containing H₂S (1%) passes through the ZnO/R-CuO and/or ZnO/CF sorbent beds at 873–1073 K, H₂S_(g) can be captured by desulfurization with ZnO and CuO to form ZnS, CuS, and Cu₂S. In the regeneration of the sorbents by O₂ at 873 K for high-temperature regeneration (HTRG), ZnO and CuO can be restored with a formation of SO₂ gas that may be collected by dissolution in H₂O_{2(aq)} for potential recycling of high-purity H₂SO₄ solution.

to form ZnS, CuS, and Cu₂S. Regeneration of the sorbents by O_2 at 873 K for HTRG, ZnO, and CuO can be restored with a formation of SO₂ gas that may be collected by dissolution in $H_2O_{2(aq)}$ for potential recycling of high-purity H_2SO_4 solution.

4. CONCLUSIONS

A feasibility study for HTDS of a simulated syngas using the ZnO sorbent-dispersed Raney CuO (ZnO/R-CuO) and ceramic filter (ZnO/CF) has been carried out. The principal component analysis and least-squares fitted XANES data show that mainly Zn(II) and Cu(II) are found in the ZnO/R-CuO sorbents. At high temperatures (873 K), ZnO as well as R-CuO is involved in desulfurization to form ZnS, Cu₂S, and CuS. The HTDS reaction kinetic parameters such as rate constant $(8.35 \times 10^4 \text{ cm}^3/\text{g/min})$ and activation energy (114.8) kJ/mol) by the ZnO/R-CuO sorbents have been obtained for engineering applications. Regeneration of the sulfurized sorbents by oxidation with O2 at 873 K for 1 h can restore ZnO and CuO for continuous and repetitive HTDS-HTRG cycles. While the ZnO/R-CuO sorbents have relatively high HTDS efficiencies (82–90%), the ZnO/CF sorbent/filter can simultaneously perform HTPF at 873 K for 1 h to reach high efficiencies (>98%). This demonstrates the feasibility for hotsyngas clean-up processes with a much better energy efficiency, lesser cost, and lower carbon footprint for cleaner power generation.

ASSOCIATED CONTENT

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

AUTHOR INFORMATION

Corresponding Authors

- Kuen-Song Lin Department of Chemical Engineering and Materials Science, Yuan Ze University, Chung-Li 32003, Taiwan; Email: slin@saturn.yzu.edu.tw
- H. Paul Wang Department of Environmental Engineering, National Cheng Kung University, Tainan City 70101, Taiwan; orcid.org/0000-0001-7272-8031; Email: wanghp@ncku.edu.tw

Authors

- T.-C. Wang Department of Environmental Engineering, National Cheng Kung University, Tainan City 70101, Taiwan
- Ling-Wei Wei Department of Environmental Engineering, National Cheng Kung University, Tainan City 70101, Taiwan
- H.-L. Huang Department of Safety, Health and Environmental Engineering, National United University, Miaoli 36003, Taiwan

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.2c08260

Notes

The authors declare no competing financial interest.

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