Supplementary Materials

In the **Supplementary Material** the XRD analysis before and after testing, TEM-EDS images, fuel cell performance of different composition, Uv-visible and UPS data, and EIS fitted tables for ZnO, Mg-ZnO, and ZnO/Mg-ZnO at 420-520 ^{o}C have been presented in detail.

ZnO/MgZnO Heterostructure membrane with Type II Band Alignment for Ceramic Fuel Cells

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Fig.1 shows the XRD data of ZnO/Mg-ZnO before and after fuel cell measurements. The obtained data of XRD after fuel cell measurements reveal no apparent impurity peak confirming the formation of the pure phase. Besides, the peak at 43 degrees is related to NiO, which might be due to the reduction of NCAL in the H_2 environment at the anode side.

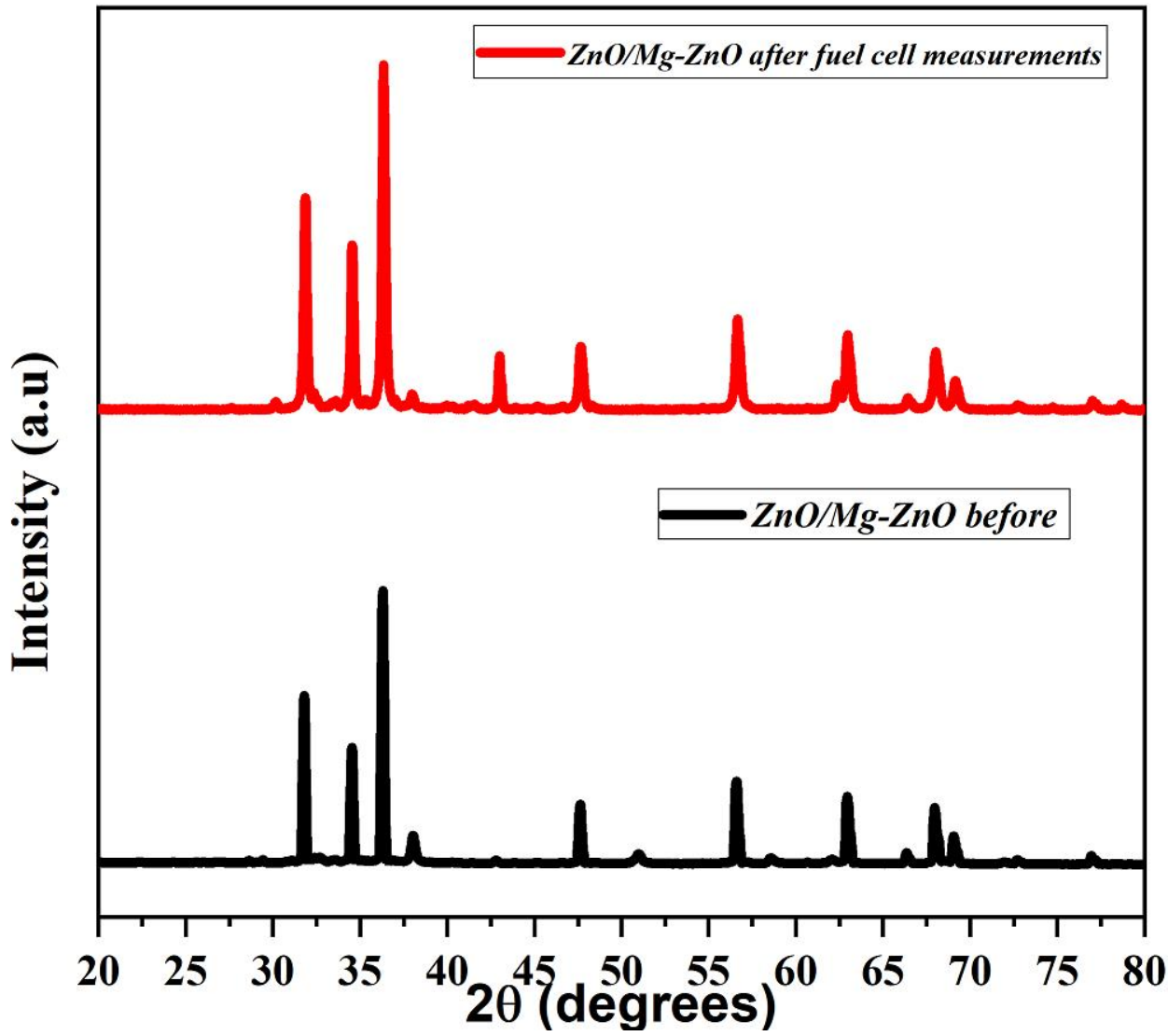


Fig. 1 XRD analysis of ZnO/Mg-ZnO before and after fuel cell characterization

EDS analysis

Fig. 2(a-e) shows the HR-TEM EDS images of HS revealing the existence of all elements in HS. Also, elemental rations have been extracted, as shown in Fig. 1(f). Also, the presence of each element probably confirms the establishment of heterostructure between ZnO and Mg-ZnO.

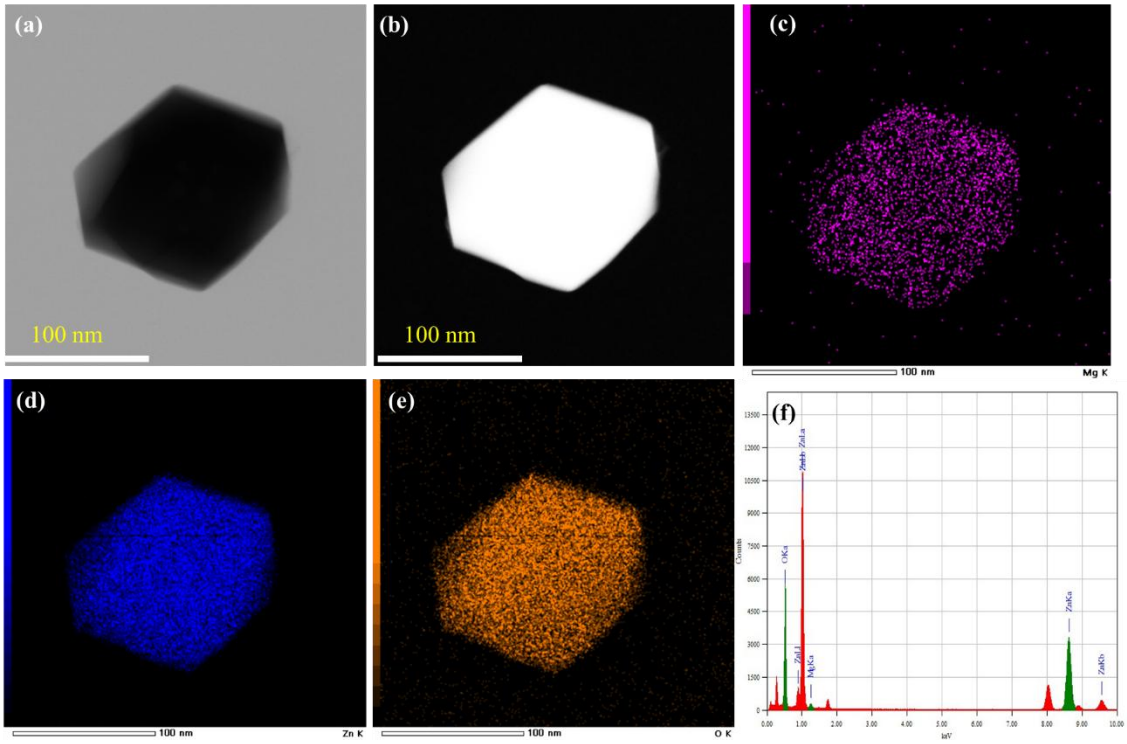
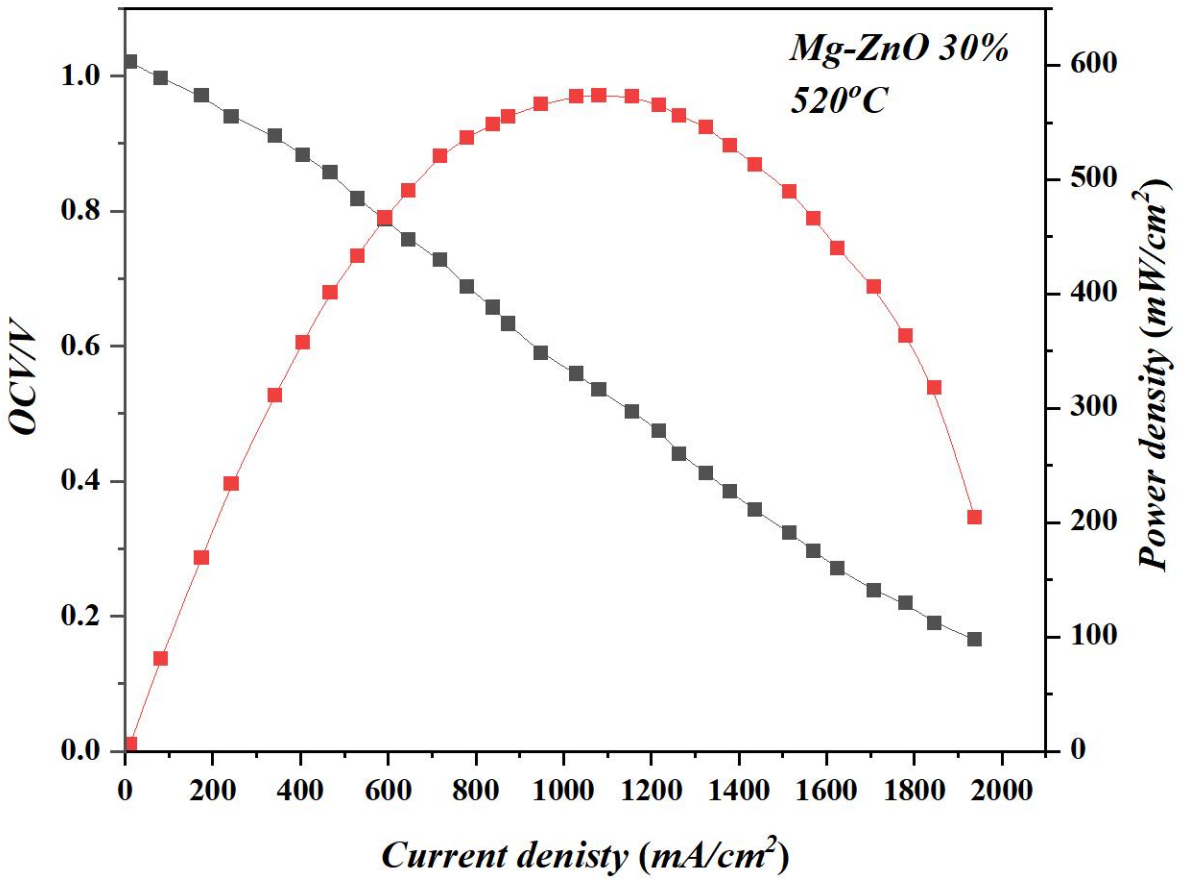


Fig. 2(a-f) HRTEM-EDX of ZnO/Mg-ZnO, revealing all elements' uniform distribution, including Zn, Mg, and oxygen.

Fig.3(a, b) shows the fuel cell performance of Mg-ZnO at 30% and different composite heterostructures with a different ratio of ZnO and Mg-ZnO. The fuel cell performance was performed in H_2 and Air environment at 520°C. The fuel cell performance of 30% doping of Mg into ZnO was executed and delivered the maximum fuel cell performance of 574 mW/cm^2 at 520°C. The obtained performance is lower than 20 % doping of Mg into ZnO 673 mW/cm^{2,} confirming that 20% is optimal for higher fuel cell performance. Furthermore, the heterostructure with different ratios like 2:1 and 1:2 have been performed and delivered the maximum fuel cell performance of 831 and 765 mW/cm^2 at 520°C. The obtained performance is lower than the 1:1 ratio of ZnO and Mg-ZnO (997 mW/cm^2), suggesting that 1:1 is an optimal ratio to deliver better fuel cell performance.


Mg-ZnO 30%
520°C
OCV/V
Power density (mW/cm²)
Current denisty (mA/cm²)
0.0
0.2
0.4
0.6
0.8
1.0
0
100
200
300
400
500
600
0
200
400
600
800
1000
1200
1400
1600
1800
2000

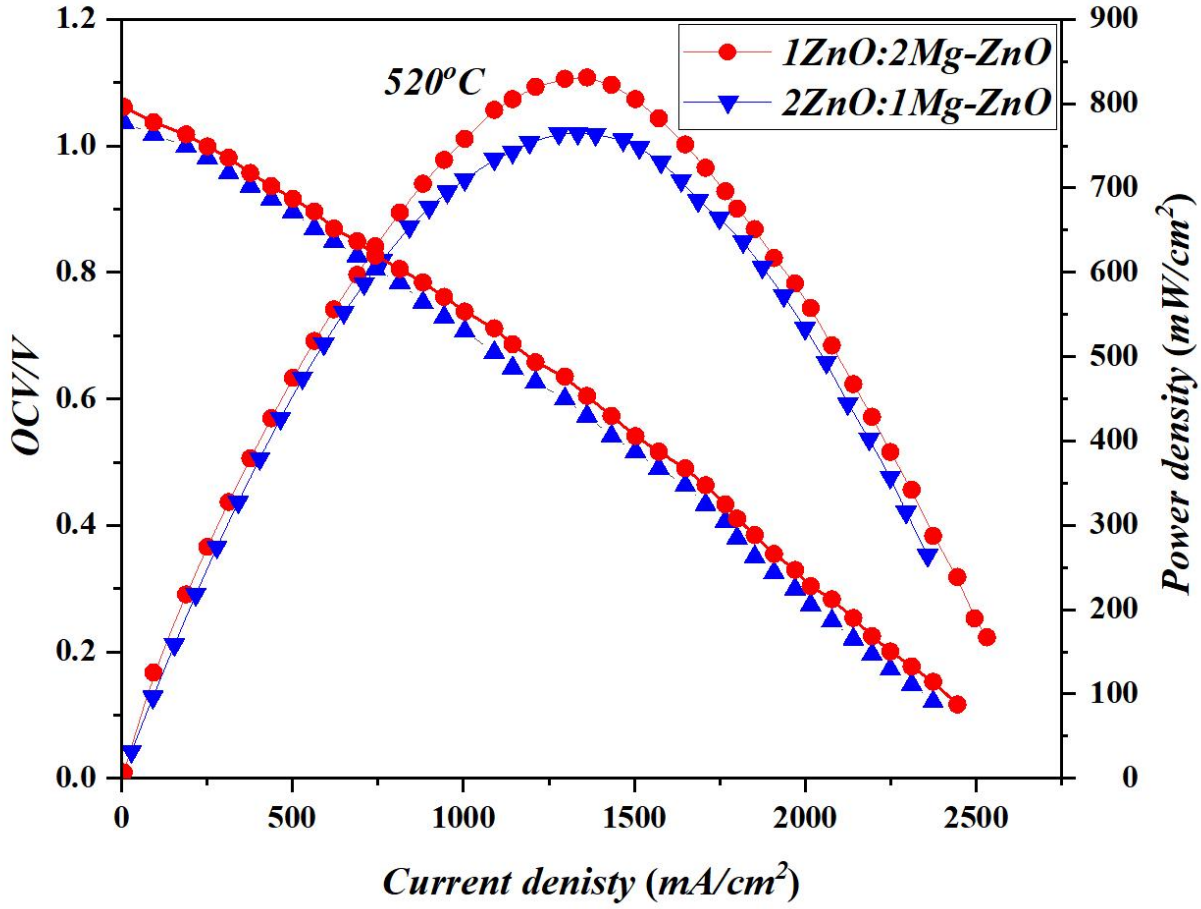


Fig.3(a, b) Fuel cell performance of Mg-ZnO 30% and different groups of heterostructure like 1:2 and 2:1 at 520 °C in H_2 and Air environments.

Optical Properties

To study the optical properties of prepared materials, Uv-visible and UPS analysis was performed to determine the energy bandgap and position of valence band maximum. The absorption of Mg-ZnO is higher than the ZnO, reducing the bandgap by incorporating Mg into ZnO, as shown in Fig.4 (a, b). The obtained bandgap of 3.12 and 2.85 eV belongs to the pure ZnO and Mg-ZnO, respectively. The attained valence band maximum position of ZnO and Mg-ZnO are 7.89 eV and 9.11 eV, as displayed in Fig.4 (c, d). The detailed evaluation of Uv-visible and UPS values is according to the previously discussed literature[1]. Based on obtained VB and energy bandgap values, the corresponding conduction band has been evaluated using the following equation ECB = EVB – Eg to be 4.77 and 6.26 Ev for ZnO and Mg-ZnO, respectively.

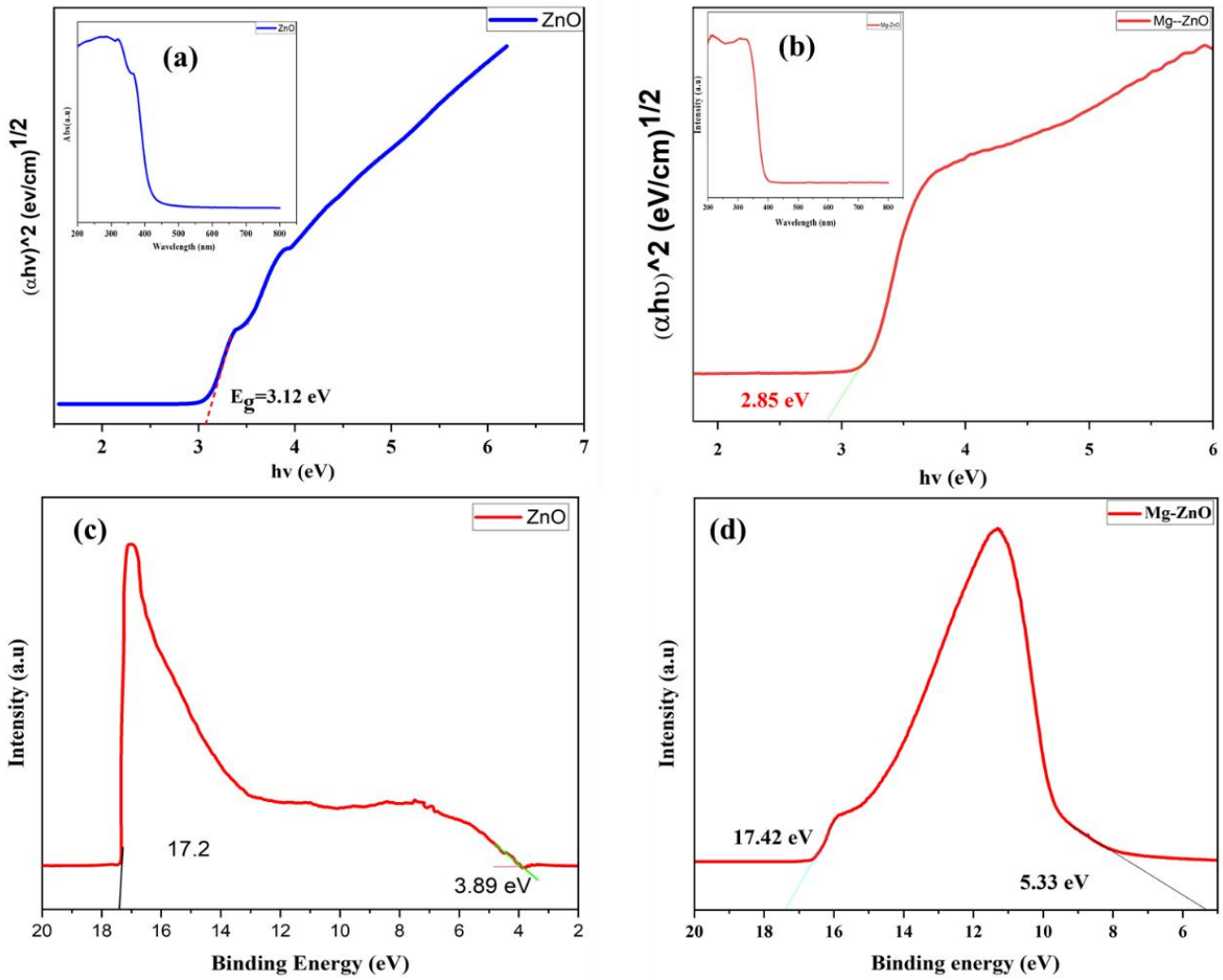


Fig. 4(a-d) Uv-visible and UPS data of ZnO and Mg-ZnO

Table. 1. The EIS fitted data of the ZnO electrolyte cell materials at different temperatures of 520-420 ºC.

T	R_o	R_1	R_2
520 °C	0.18	0.23	0.816
470 °C	0.30	0.32	1.88
420 °C	0.516	0.484	3.34

Table. 2. The EIS fitted data of the Mg-ZnO electrolyte cell materials at different temperatures of 520-420 °C.

T	R_o	R_1	R_2
520 °C	0.17	0.08	0.318
470 °C	0.218	0.093	0.43
420 °C	0.29	0.12	0.8

Table. 3. The EIS fitted data of the ZnO/Mg-ZnO HS electrolyte cell materials at different temperatures of 520-420 °C.

T	R_o	R_1	R_2
520 °C	0.12	0.05	0.25
470 °C	0.14	0.082	0.36
420 °C	0.17	0.1	0.713

References

1. B. Zhu, B. Wang, Y. Wang, R. Raza, W. Tan, J. Kim, P. A. van Aken, and P. Lund, *Nano Energy.* 37, 195 (2017).