Supplementary Information

Regenerable Oxygen-Deficient Ni/γ-Al_2O_3 Catalyst for Efficient Glycerol Aqueous Phase Reforming

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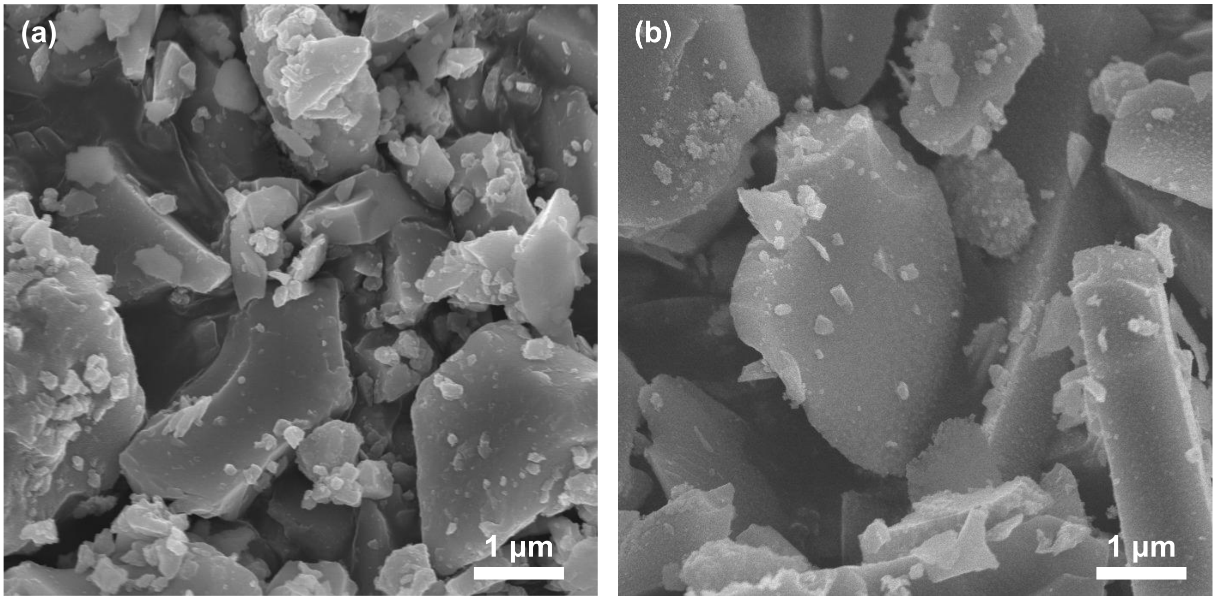

Figure S1. SEM images of (a) $NiAl_2O_4$ and (b) Ni/γ-Al_2O_3 powders.

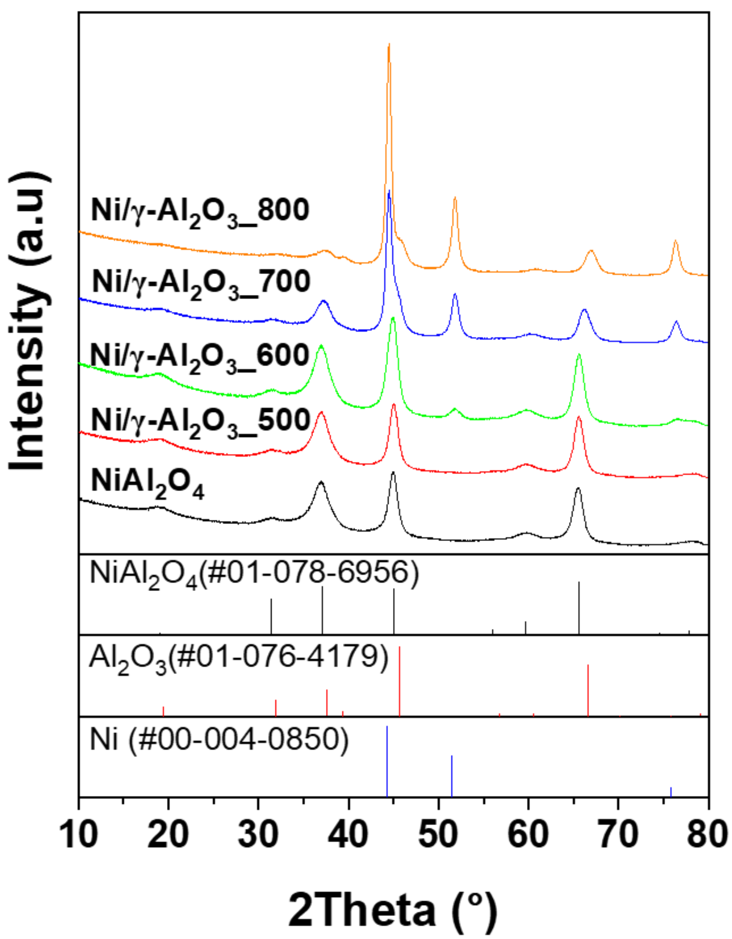

Figure S2. XRD spectra of the Ni/γ-Al_2O_3 prepared at different temperatures.

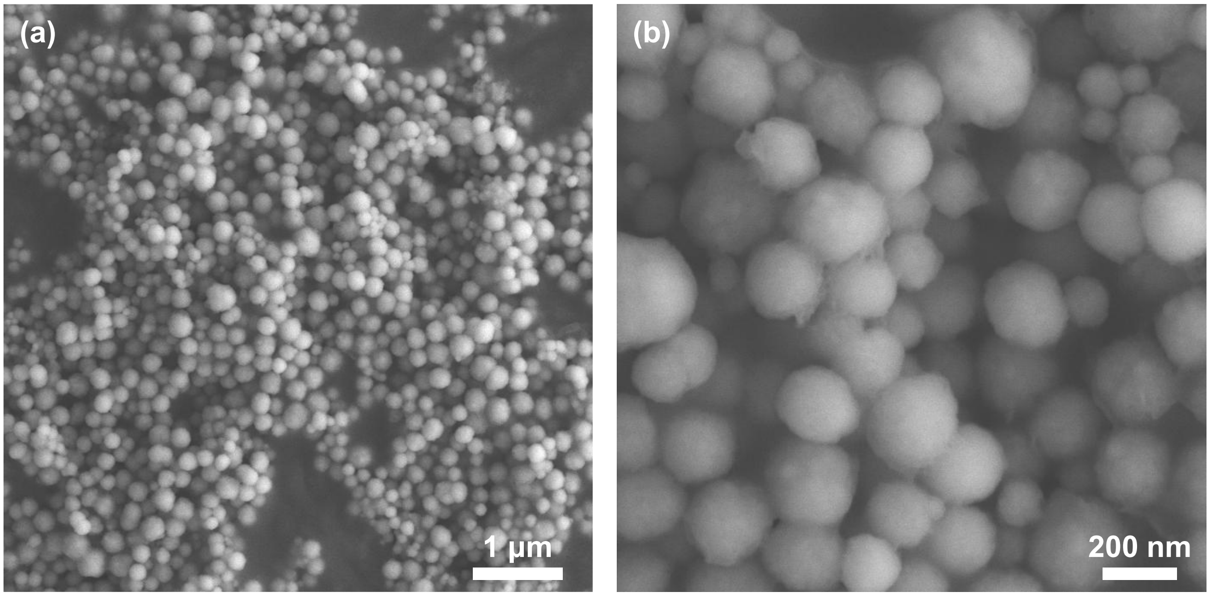

Figure S3. SEM images of the Ni nanoparticles prepared by hydrothermal process.

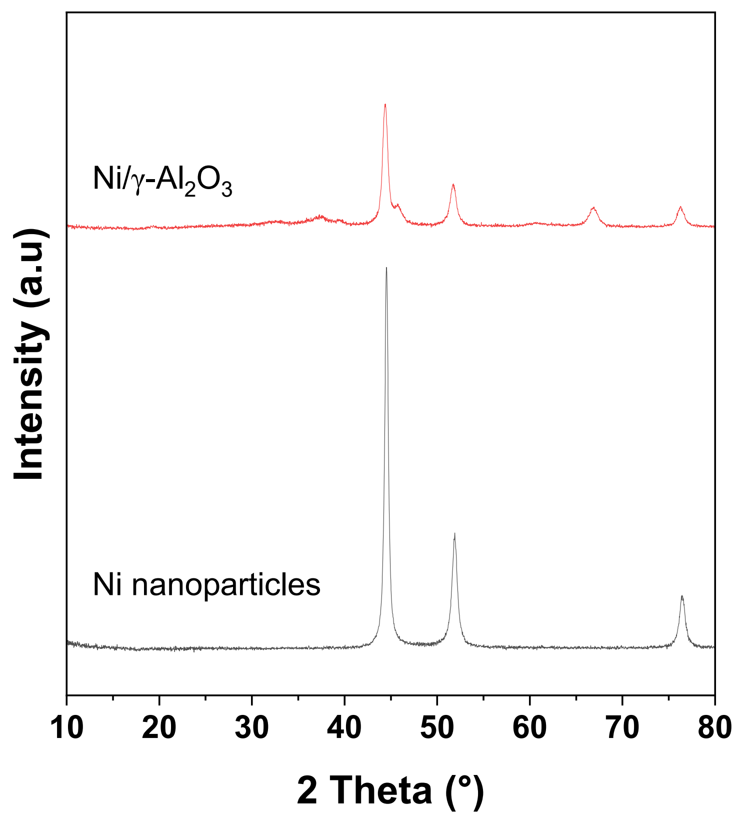

Figure S4. XRD spectra of the Ni/γ-Al_2O_3 prepared by exsolution (red) and Ni nanoparticles by hydrothermal process (black).

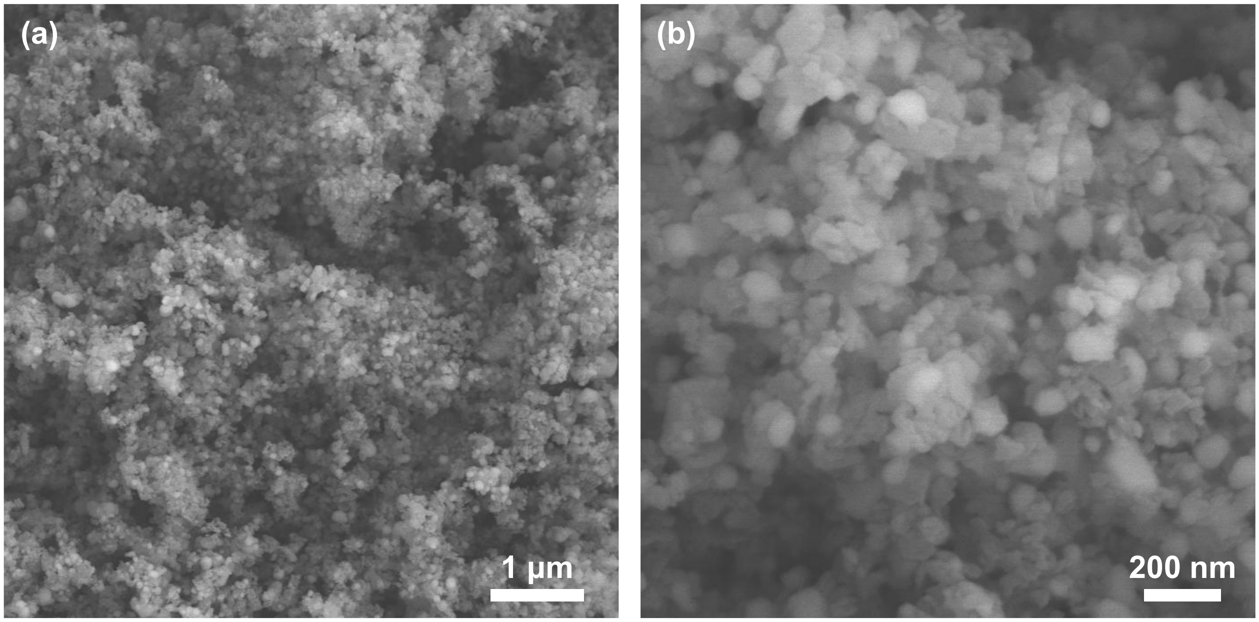

Figure S5. SEM images of the Ni/γ-Al_2O_3_w prepared by wet impregnation method.

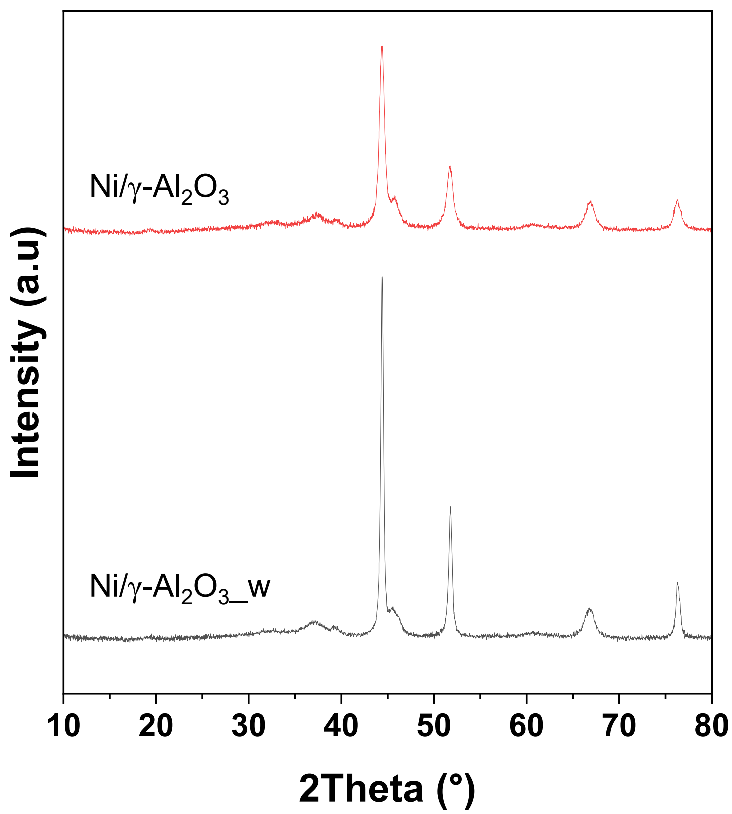

Figure S6. XRD spectra of the Ni/γ-Al_2O_3 prepared by exsolution (red) and wet impregnation (black).

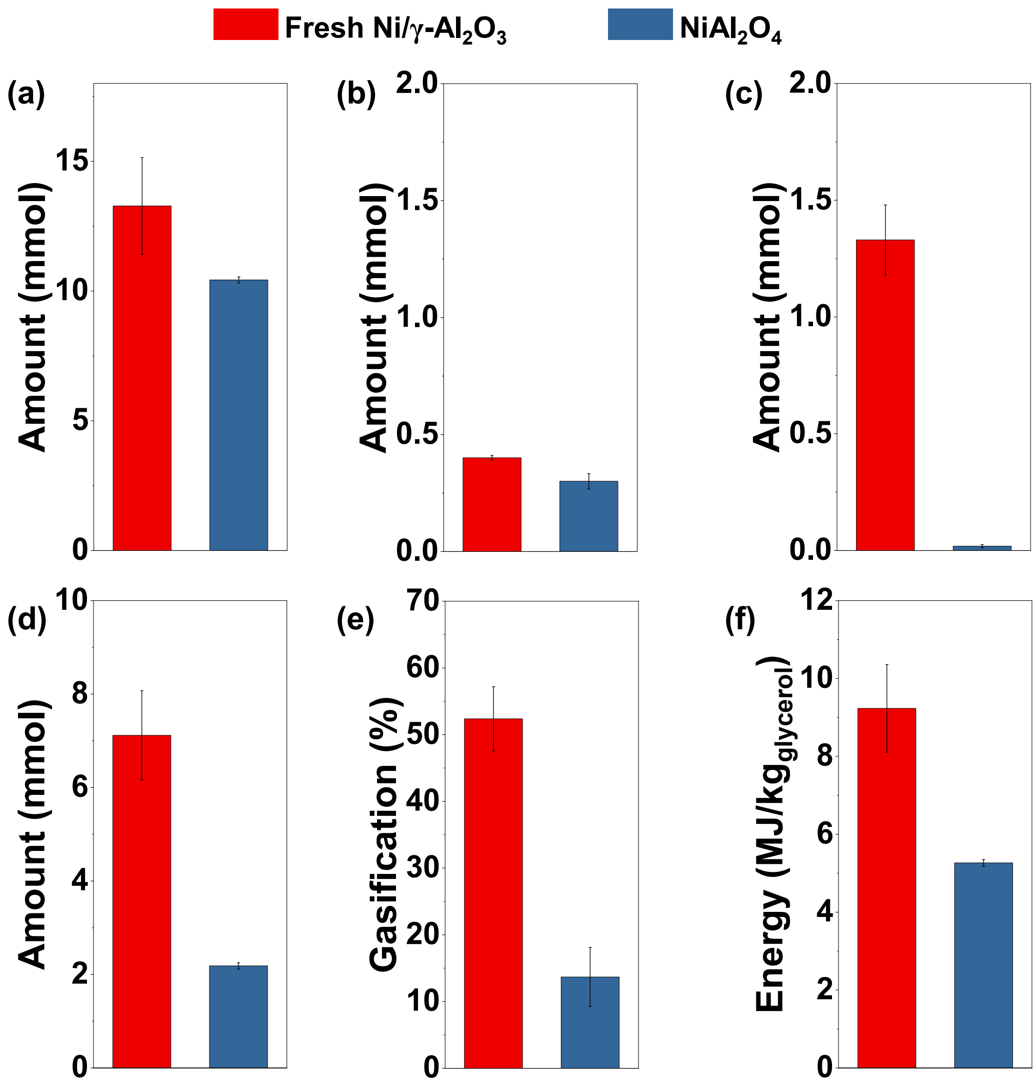

Figure S7. Catalytic performance comparison of fresh Ni/γ-Al_2O_3 and $NiAl_2O_4$: **(a)** hydrogen, **(b)** carbon monoxide, **(c)** methane, and **(d)** carbon dioxide production. **(e)** Gasification yields of glycerol on a carbon basis and **(f)** HHV of the gas products.

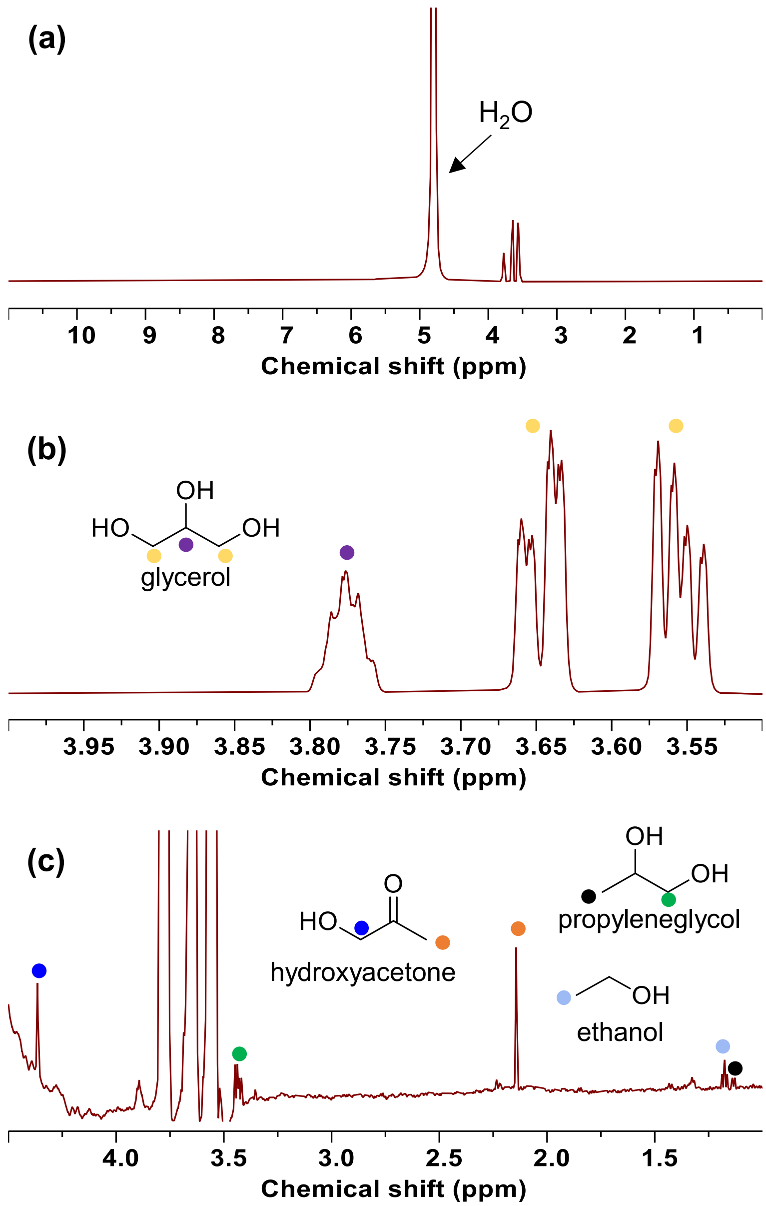

Figure S8. ^{1}H-NMR spectra of reaction mixture after APR of glycerol using γ-Al_2O_3. (a) Full spectrum and (b), (c) magnified spectra.

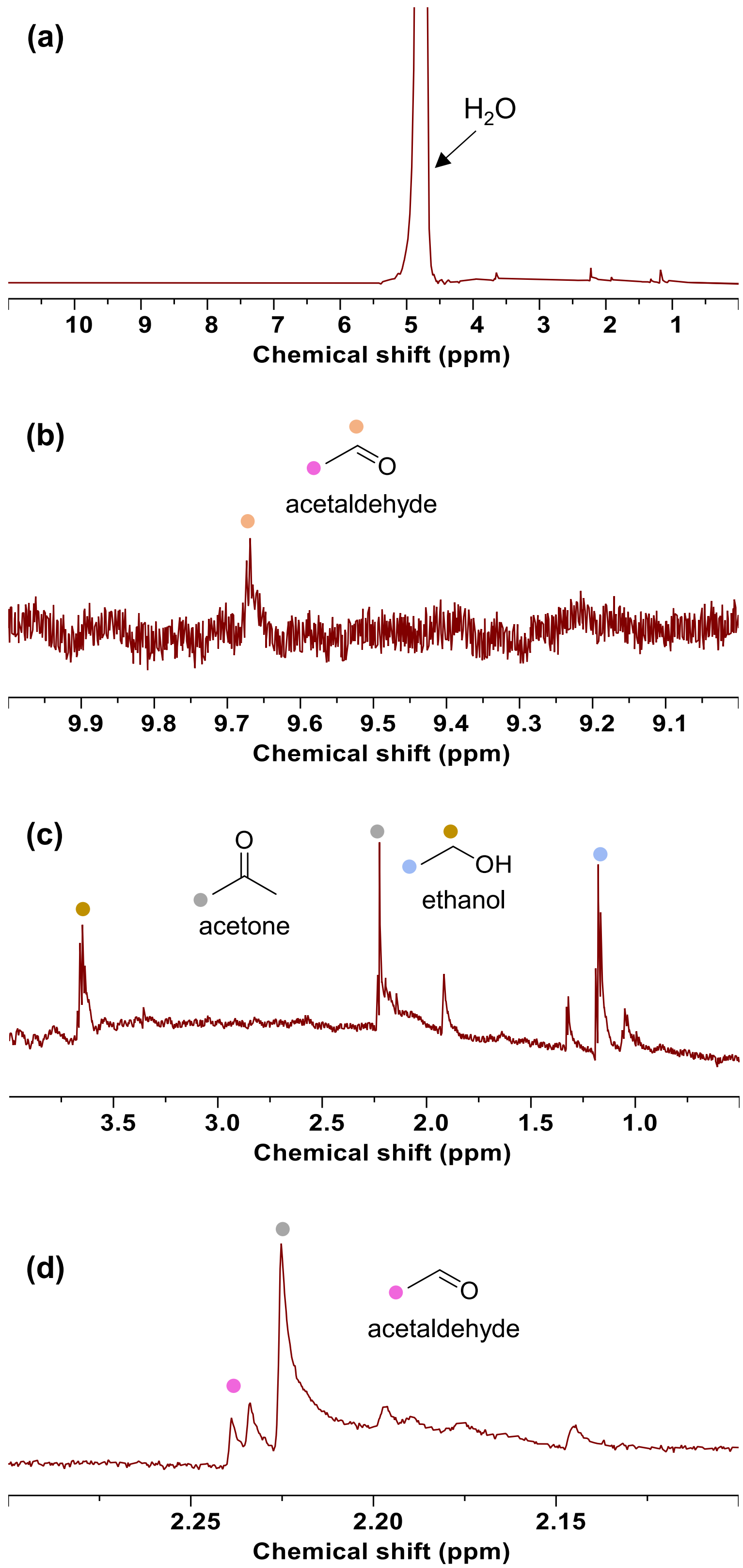

Figure S9. ^{1}H-NMR spectra of the reaction mixture after APR of glycerol using Ni nanoparticles. (a) Full spectrum and (b)-(d) magnified spectra.

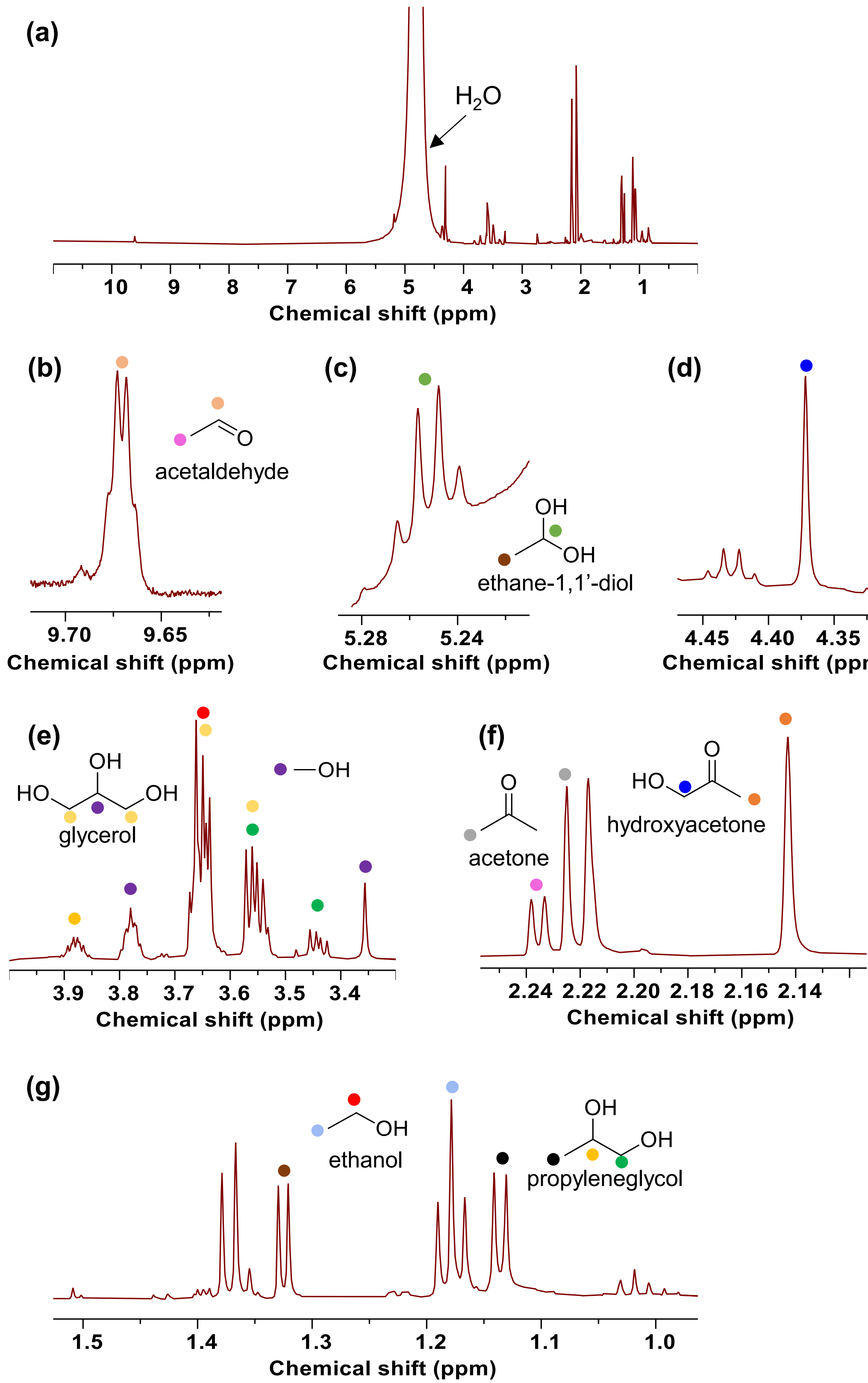

Figure S10. ^{1}H-NMR spectra of the reaction mixture after 30 min of APR of glycerol using Ni/γ-Al_2O_3. (a) Full spectrum and (b)-(g) magnified spectra.

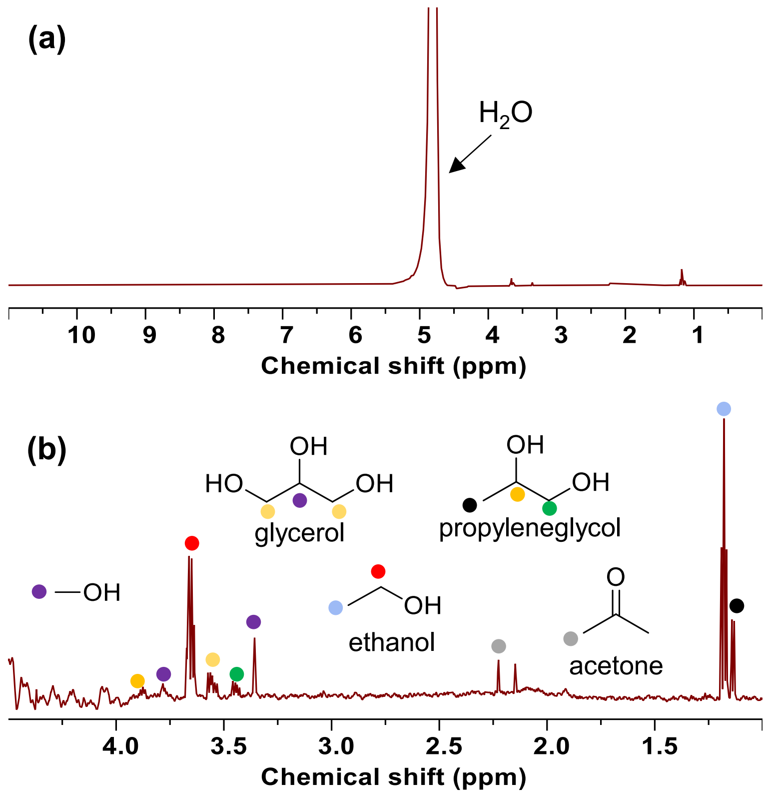

Figure S11. ^{1}H-NMR spectra of the reaction mixture after 45 min of APR of glycerol using Ni/γ-Al_2O_3. (a) Full spectrum and (b) magnified spectra.

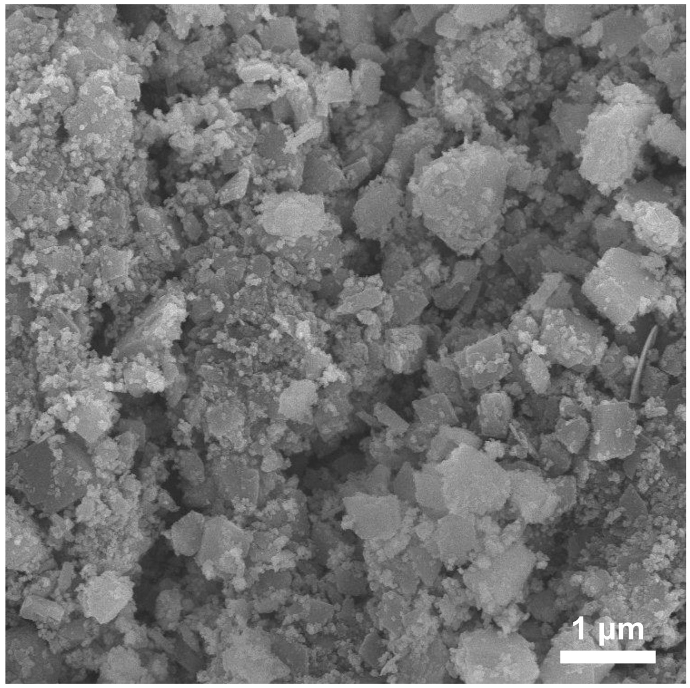

Figure S12. SEM image of the used Ni/γ-Al_2O_3.

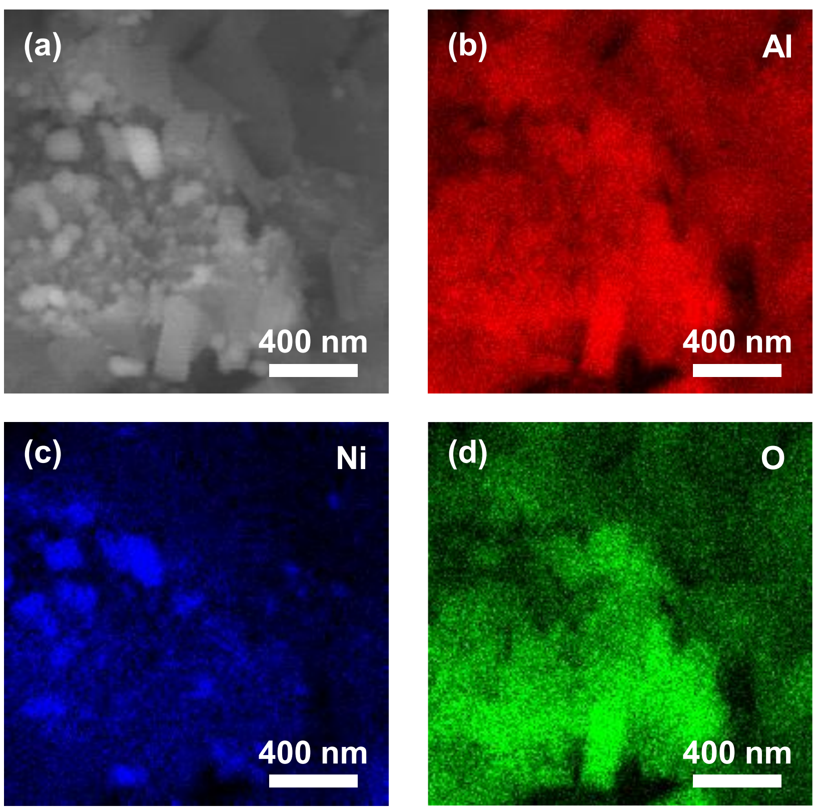

Figure S13. SEM-EDS mapping data of regenerated Ni/γ-Al_2O_3. (a) SEM image and elemental distribution mapping data of (b) aluminum, (c) nickel, and (d) oxygen.

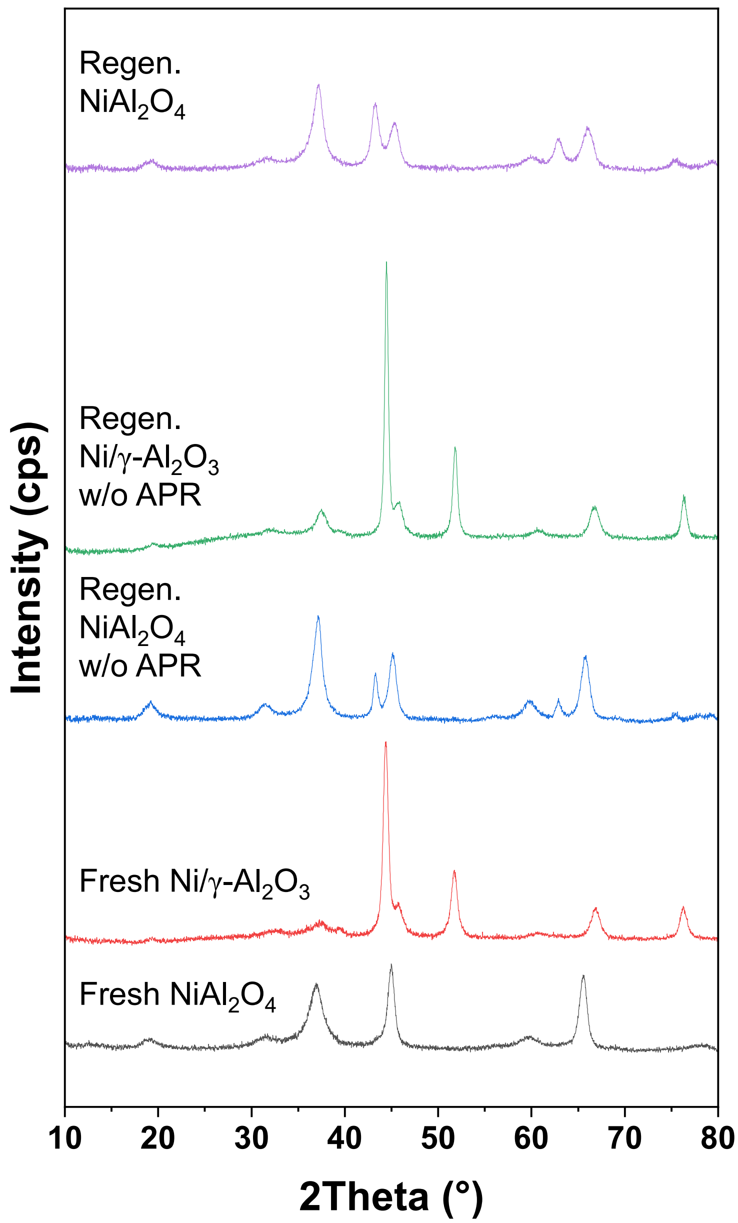

Figure S14. XRD spectra of the fresh $NiAl_2O_4$ (black), fresh Ni/γ-Al_2O_3 (red), $NiAl_2O_4$ regenerated without APR (blue), regenerated Ni/γ-Al_2O_3 without APR process (green), and regenerated $NiAl_2O_4$ (purple).

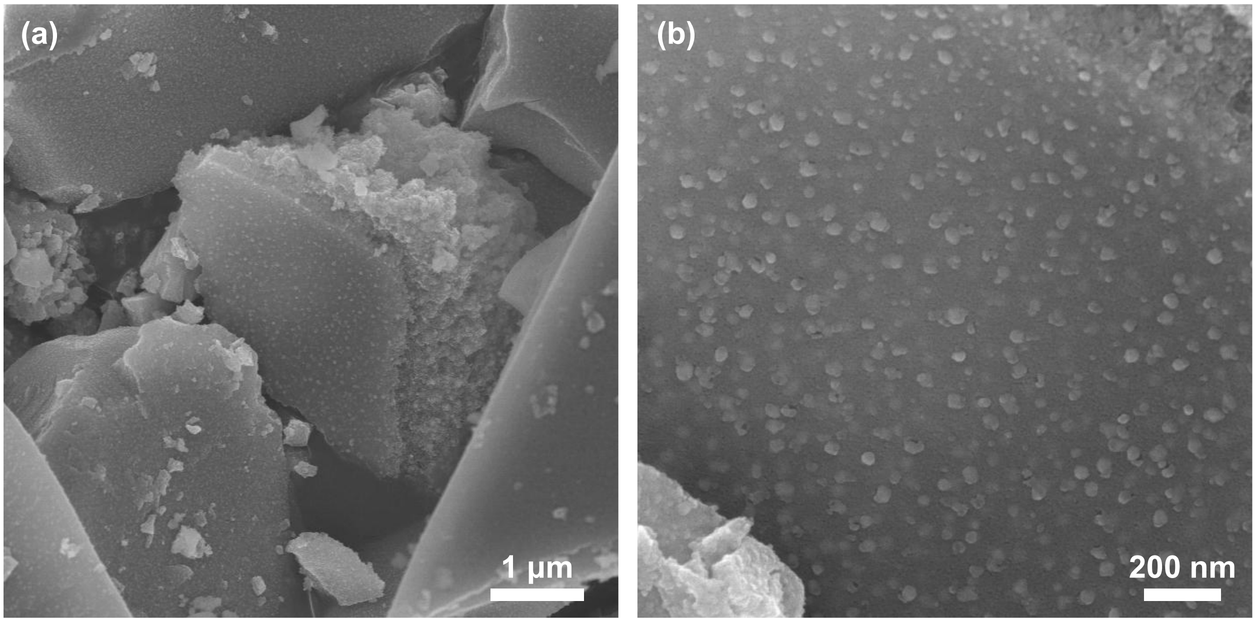

Figure S15. SEM images of regenerated Ni/γ-Al_2O_3 without APR.

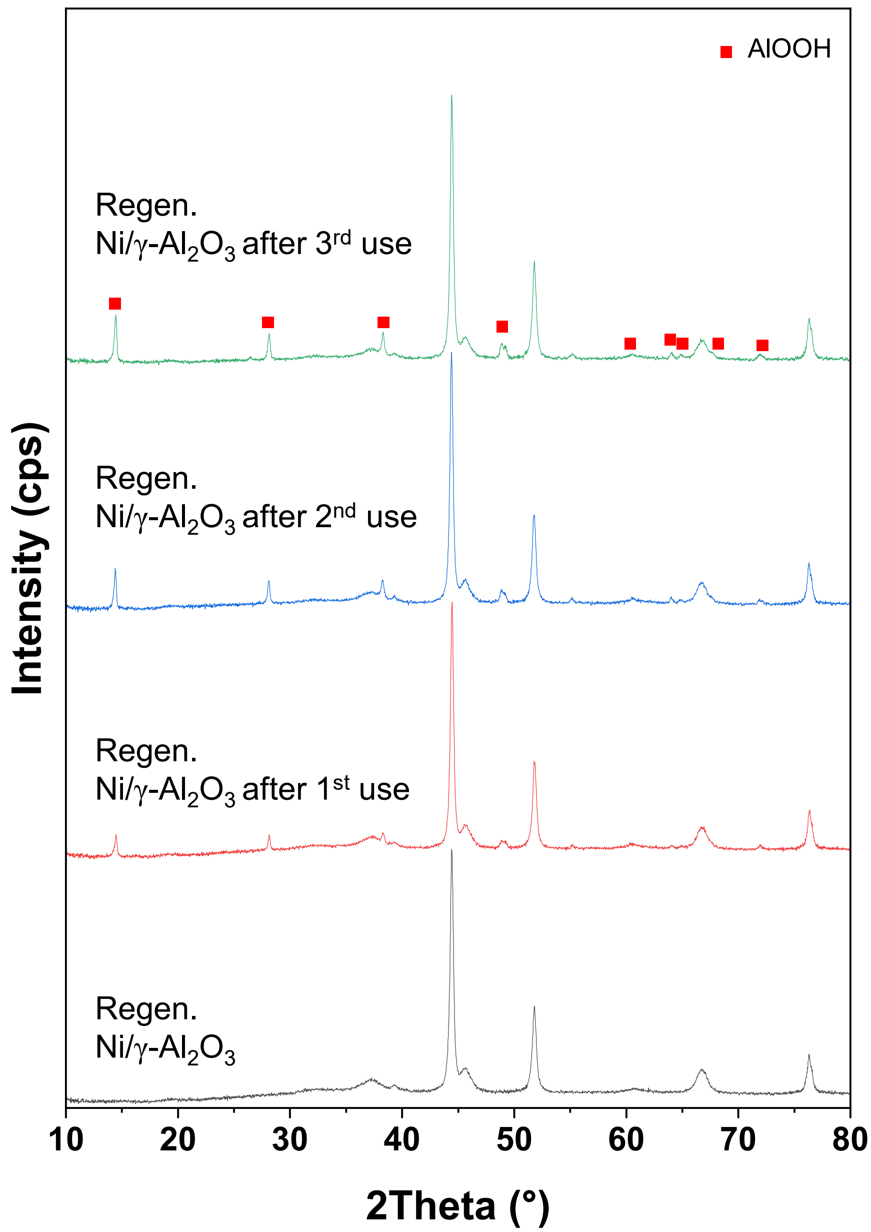

Figure S16. XRD spectra of the regenerated Ni/γ-Al_2O_3 for consecutive cyclic tests.

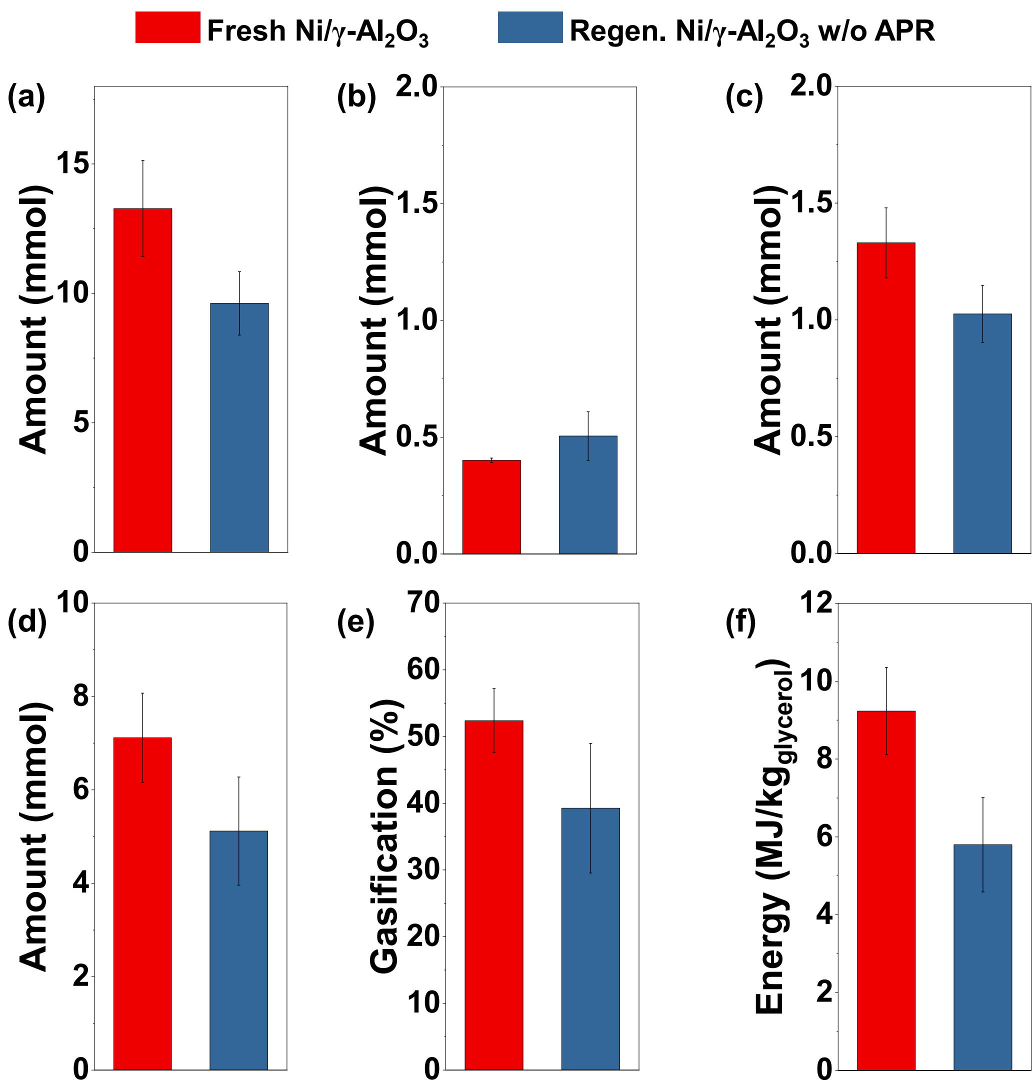

Figure S17. Catalytic performance comparison of fresh Ni/γ-Al_2O_3 and regenerated Ni/γ-Al_2O_3 without APR process (Regen. Ni/γ-Al_2O_3 w/o APR): (a) hydrogen, (b) carbon monoxide, (c) methane, and (d) carbon dioxide production. (e) Gasification yields of glycerol on a carbon basis and (f) energy of the gas products.

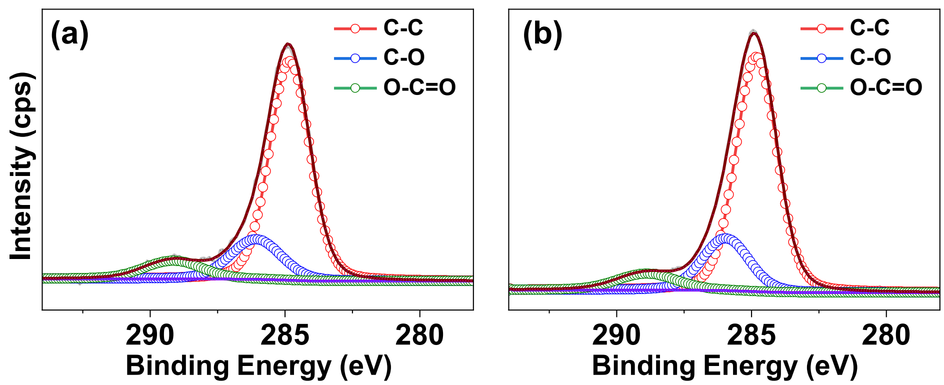

Figure S18. C 1s XPS spectra of (a) fresh Ni/γ-Al_2O_3 and (b) regenerated Ni/γ-Al_2O_3.

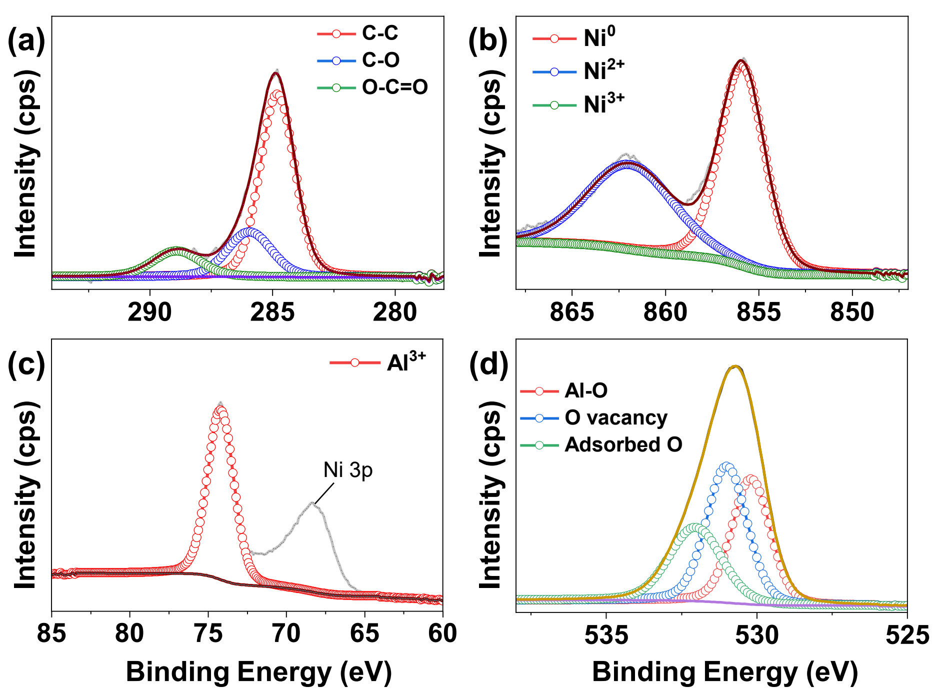

Figure S19. XPS spectra of fresh $NiAl_2O_4$ spinel oxide. (a) C 1s, (b) Ni 2p, (c) Al 2p, and (d) O 1s spectra.

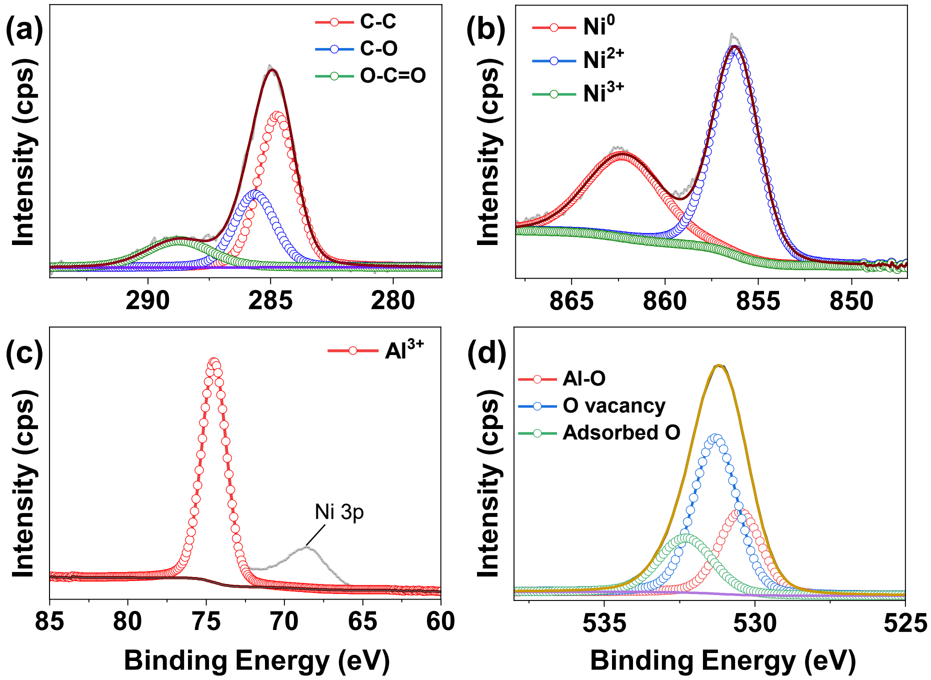

Figure S20. XPS spectra of regenerated $NiAl_2O_4$ spinel oxide. (a) C 1s, (b) Ni 2p, (c) Al 2p, and (d) O 1s spectra.

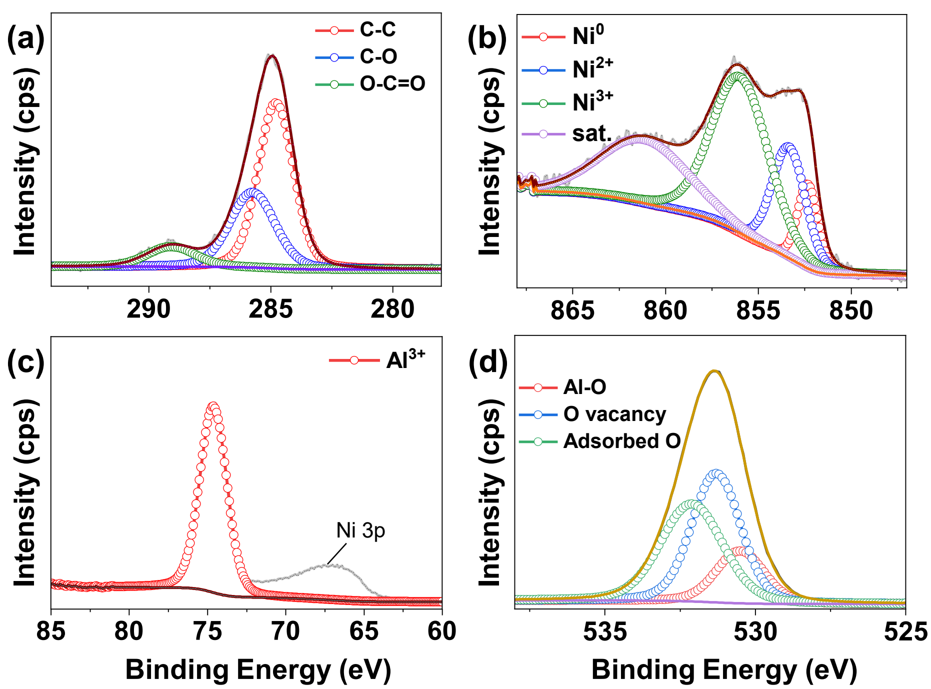

Figure S21. XPS spectra of regenerated Ni/γ-Al_2O_3 without APR. (a) C 1s, (b) Ni 2p, (c) Al 2p, and (d) O 1s spectra.

Table S1. Composition of gas products and high heating value (HHV)

	Gas product [%]				HHV
	H_2	CO_2	CH_4	CO	[MJ/kg$_{gas}$]
Ni/γ-Al_2O_3	60.01	32.17	6.01	1.81	13.7
Ni NPs	51.44	40.56	4.83	3.17	9.7
γ-Al_2O_3	6.22	89.02	3.1	1.66	1.2
Ni/γ-Al_2O_3_w	59.07	30.54	3.84	6.56	13
Regen. Ni/γ-Al_2O_3	59.98	29.78	6.77	3.47	14.8

Table S2. XPS analysis result of fresh $NiAl_2O_4$ and regenerated $NiAl_2O_4$

		Nickel			Aluminum	Oxygen		
		Ni^0	Ni^{2+}	Ni^{3+}		Al-O	O_v	O_{ad}
Fresh $NiAl_2O_4$	Binding energy [eV]		853.9		74.2	530.2	531.0	532.0
	Amount [%]					33.9	39.0	27.1
Regen. $NiAl_2O_4$	Binding energy [eV]		853.3		74.5	530.6	531.2	532.1
	Amount [%]					26.4	51.6	22.0
Regen. Ni/γ- Al_2O_3 w/o APR	Binding energy [eV]	852.3	853.3	856.0	74.6	530.5	531.3	532.1
	Amount [%]	12.1	26.0	61.9		16.9	42.4	40.7