Supplementary Information

Magnetic and Pseudocapacitive Enhancement in Gd-Substituted $NiFe_2O_4$

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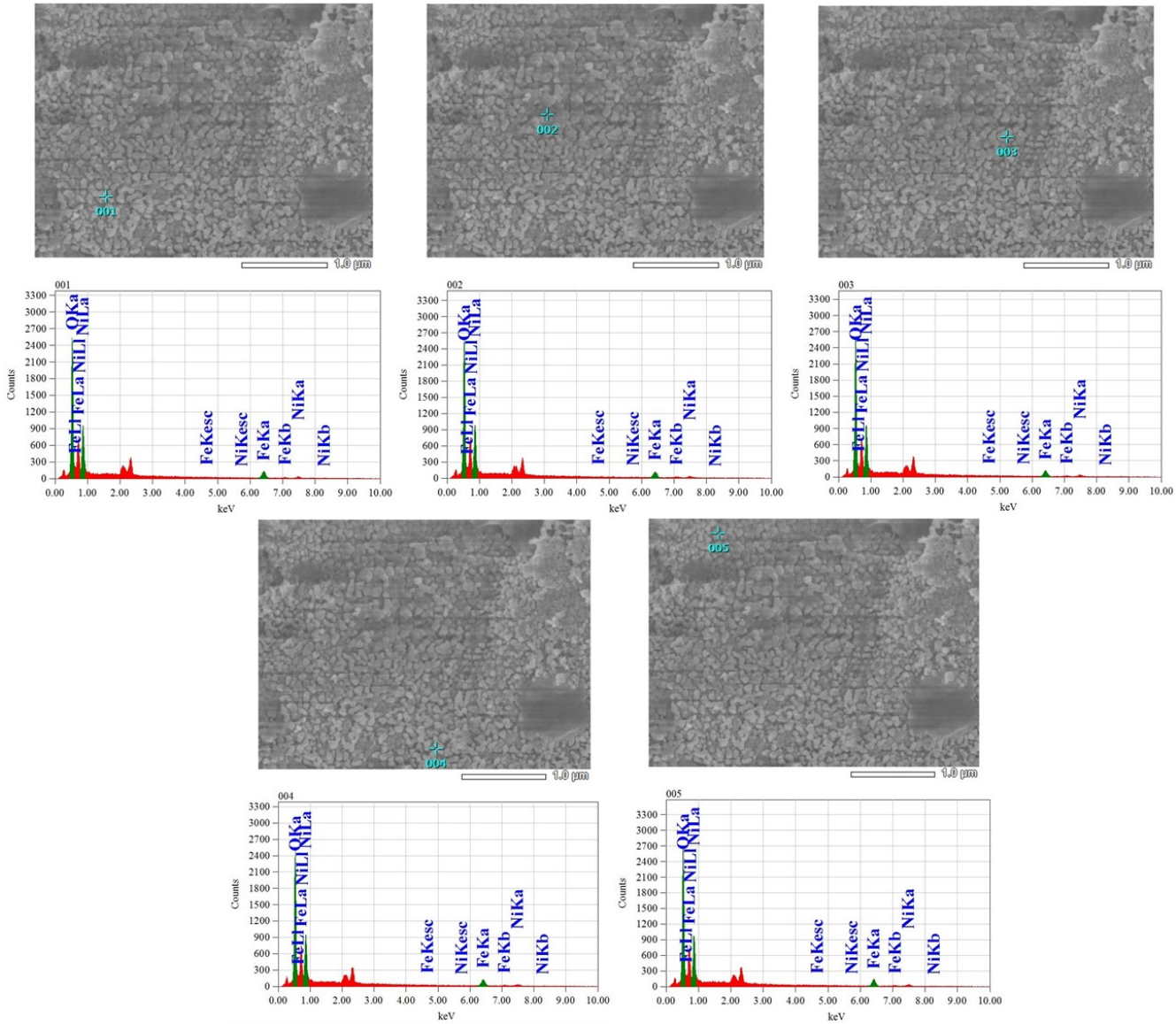
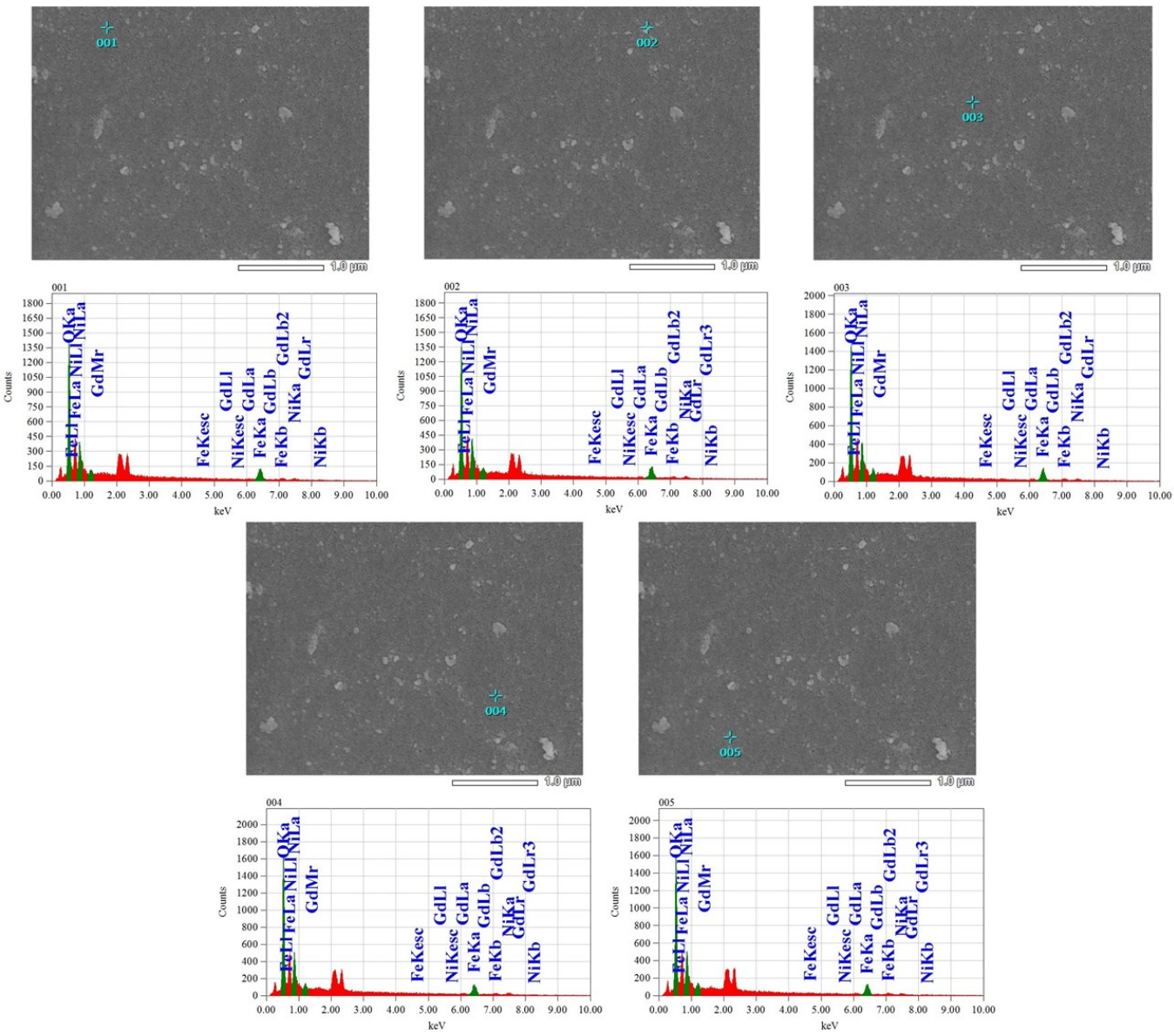


Figure S1: EDS spectra from multiple points of the NFO sample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002
003
004
005
1.0 μm
Counts
keV
OKa
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FeLl
FeLa
NiLl
GdMr
FeKesc
GdLl
NiKesc
GdLa
FeKa
GdLb
GdLb2
FeKb
NiKa
GdLr
GdLr3
NiKb

Figure S2: EDS spectra from multiple points of the NGFO2.5 sample

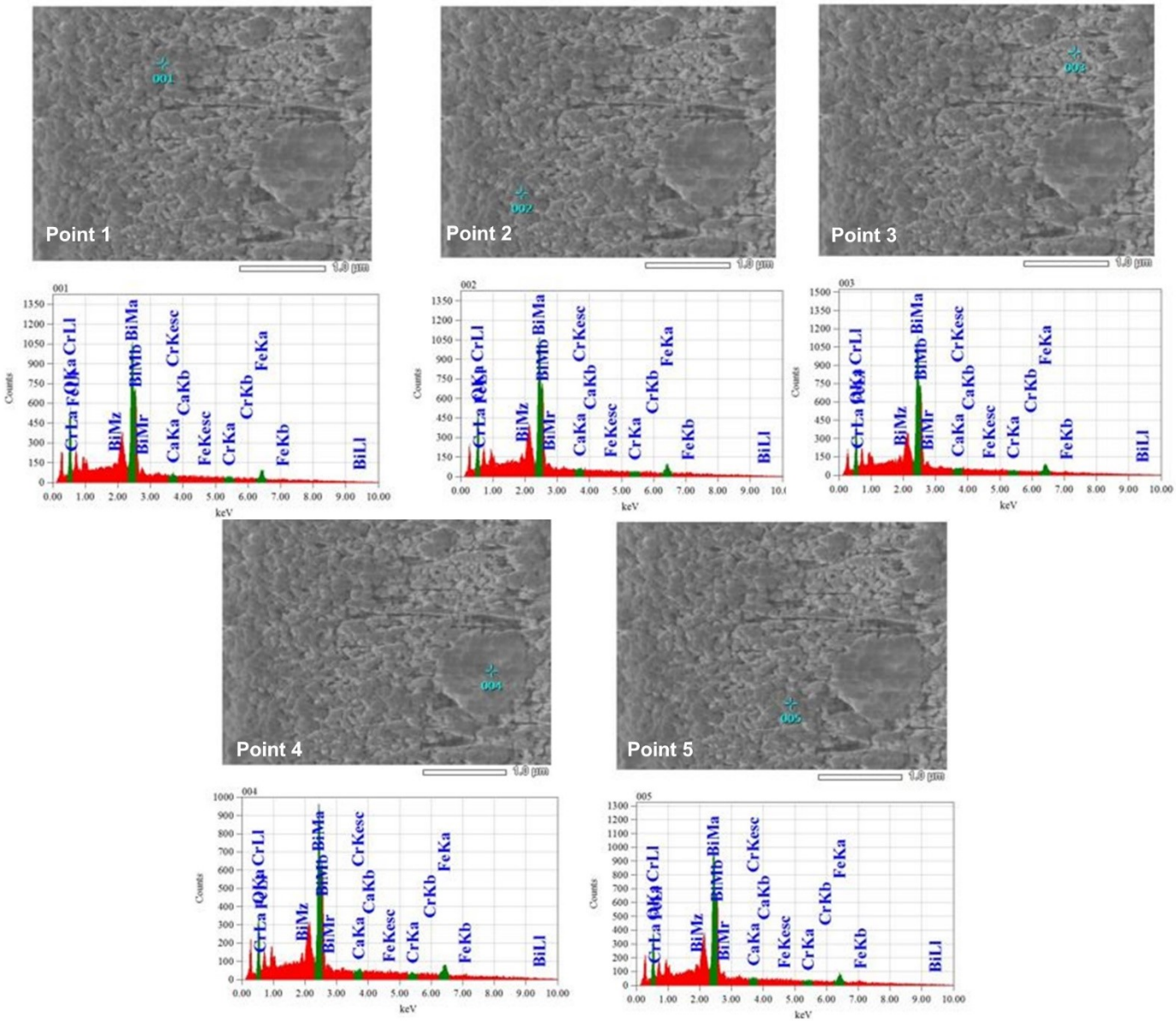

Figure S3: EDS spectra from multiple points of the NGFO7.5 sample

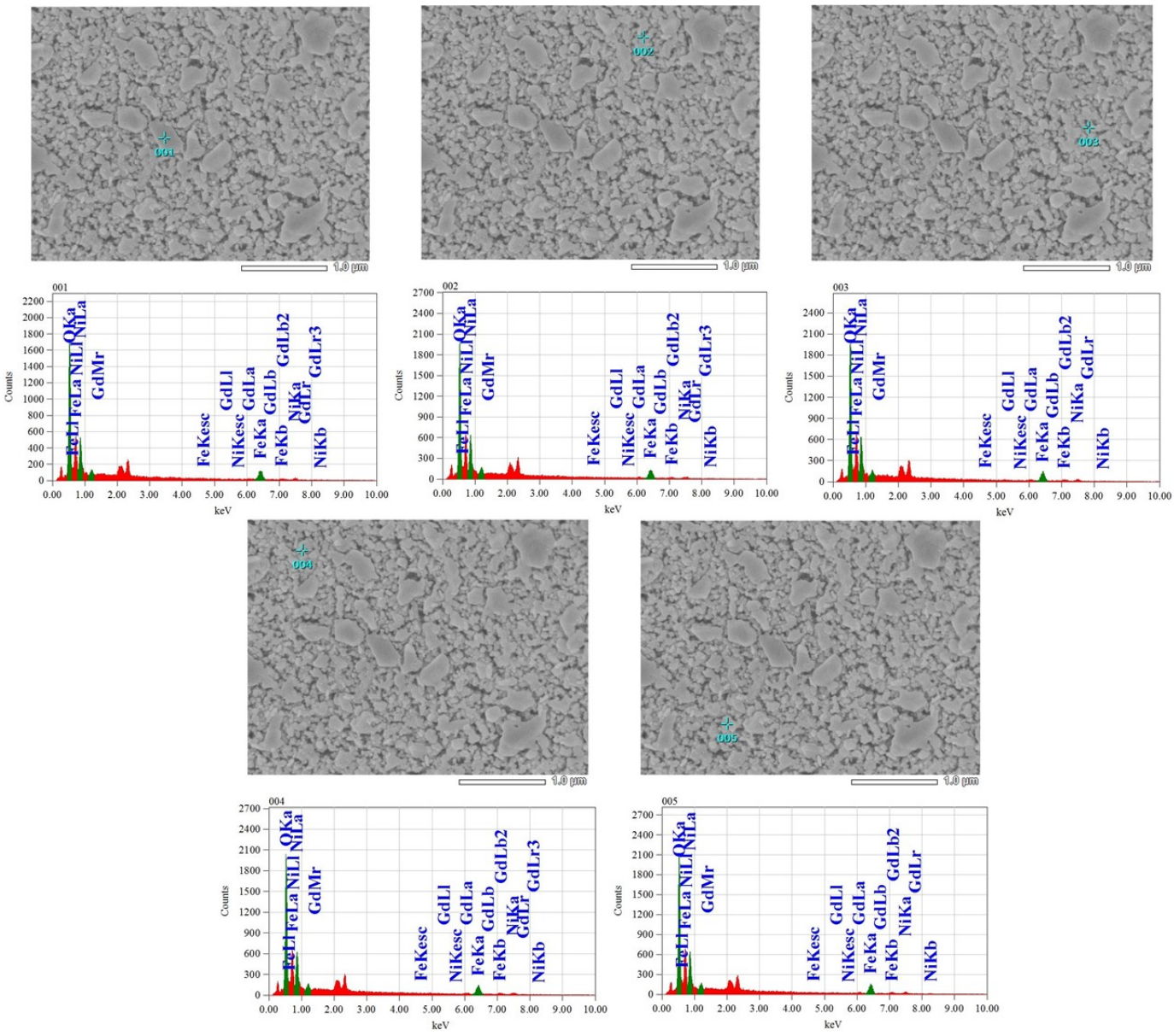

Figure S4: EDS spectra from multiple points of the NGFO12.5 sample

Cation Distribution Analysis and Magnetic Moment Estimation

To further support the interpretation of the enhanced saturation magnetization at low Gd concentration, a simplified cation distribution analysis was performed within the framework of the Néel two-sublattice model.

XPS analysis confirms that Ni^{2+} ions occupy both tetrahedral (A) and octahedral (B) sites in the present nanoparticle system. However, to illustrate the influence of Gd^{3+} substitution on magnetic behavior, two limiting boundary configurations were considered.

In the first limiting case, Ni^{2+} is assumed to occupy only the octahedral B sites (inverse spinel configuration), while Gd^{3+} preferentially substitutes Fe^{3+} at the B sites. The cation distribution is therefore represented as:

A (Tetrahedral) site	B (Octahedral) site
Fe_1	$Gd_xNi_1Fe_{1-x}$

Due to the super-exchange interaction between the two sites, the magnetic moments are antiparallelly aligned, hence,

$Net\ magnetic\ moment = 7x + 5(1-x) + 2 - 5 = 2x + 2$

When pure Nickel ferrite, $x = 0, and\ net\ moment = 2\ \mu_B$

When 2.5% Gd-doped Nickel ferrite, $x = 0.025, and\ net\ moment = 2(0.025) + 2 = 2.05\ \mu_B$

Thus, substitution of Gd^{3+} at B sites increases the net magnetic moment at low dopant concentration.

In the second configuration, consistent with our XPS results where Ni^{2+} occupies both A and B sites, the distribution is expressed as:

A (Tetrahedral) site	B (Octahedral) site
$Fe_1\ Ni_y$	$Ni_{1-y}Gd_xFe_{1-x}$

The corresponding net magnetic moment becomes,

$Net\ magnetic\ moment = 7x + 5(1-x) + 2(1-y) - 5 - 2y = 2x - 4y + 2$

Let, y=0.2,

When pure Nickel ferrite, $x = 0, and\ net\ moment = 2(0) - 4(0.2) + 2 = 1.2\ \mu_B$

When 2.5% Gd-doped Nickel ferrite, $x = 0.025, and\ net\ moment = 2(0.025) - 4(0.2) + 2 = 1.25\ \mu_B$

Even under this experimentally supported mixed-site configuration, low-level Gd substitution results in an increase in the net magnetic moment.

In both configurations, preferential occupation of Gd^{3+} at octahedral sites modifies the A-O-B superexchange pathways by altering the B-sublattice magnetic contribution. At low doping levels, this increases the imbalance between the two sublattices, resulting in enhanced saturation

magnetization. At higher dopant concentrations, weakening of Fe-O-Fe superexchange interactions and possible structural disorder can lead to the observed reduction in magnetization.

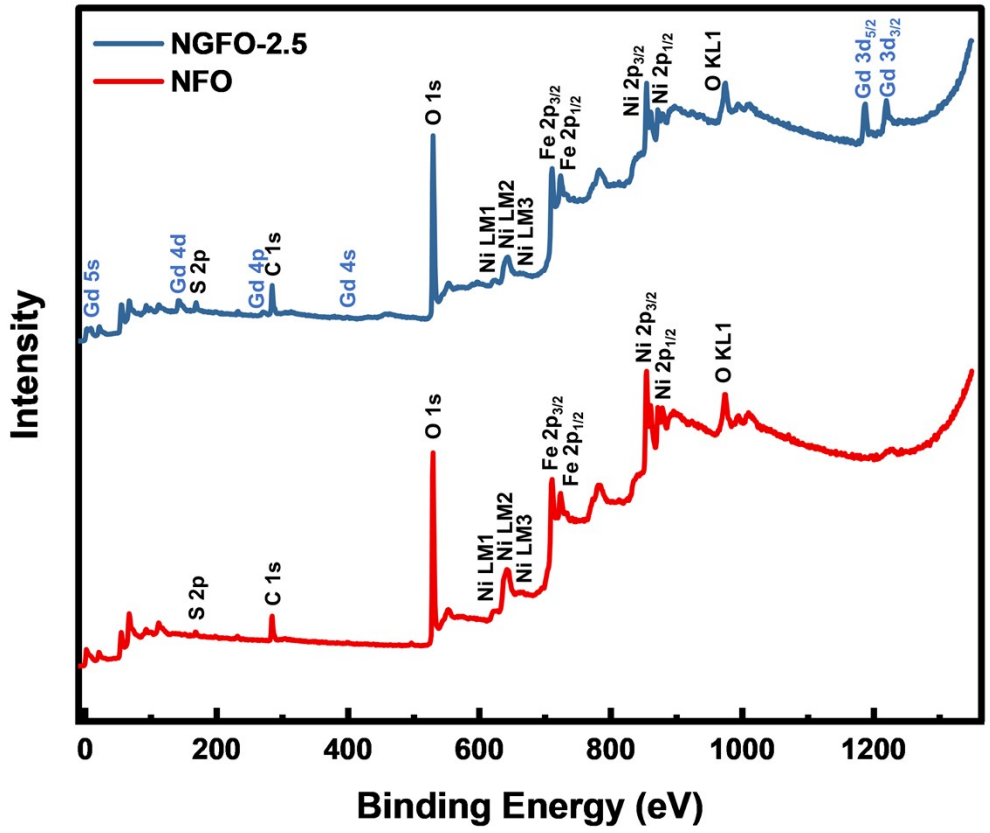

Figure S5: XPS survey spectra of NFO and NGFO2.5 samples

Table S1: Analysis Results of Ni 2p3/2 and Fe 2p3/2 XPS Spectra for NFO and NGFO-2.5 samples

Sample	Spectrum	BE (eV)	Assignment	FWHM (eV)	Atomic percentage (%)
NFO	Ni $2p_{3/2}$	854.42 ± 0.007	Ni^{2+} (B)	1.21 ± 0.02	30.5
		855.57 ± 0.220	Ni^{2+} (A)	2.74 ± 0.15	69.5
	Fe $2p_{3/2}$	710.05 ± 0.034	Fe^{3+} (B)	2.29 ± 0.09	28.5
		712.01 ± 0.185	Fe^{3+} (A)	3.71 ± 0.25	71.5
NGFO-2.5	Ni $2p_{3/2}$	854.55 ± 0.004	Ni^{2+} (B)	1.29 ± 0.01	35
		855.52 ± 0.015	Ni^{2+} (A)	2.78 ± 0.02	65
	Fe $2p_{3/2}$	710.04 ± 0.022	Fe^{3+} (B)	2.47 ± 0.09	43.5
		712.04 ± 0.184	Fe^{3+} (A)	4.09 ± 0.41	56.5