Supplementary materials

Layered-Double Hydroxides and derived oxide as CRM-free highly active catalysts for the reduction of 4-nitrophenol

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Table S1 - Values of apparent constants for pseudo-first-order reactions using Ni and Cu oxides.

Sample	K_{app} (min^{-1})	**K_{app} (min^{-1}/mg)**
Ni(II)O	0	**0**
Cu(II)O	0.18 ± 0.02	**0.018 ± 0.02**

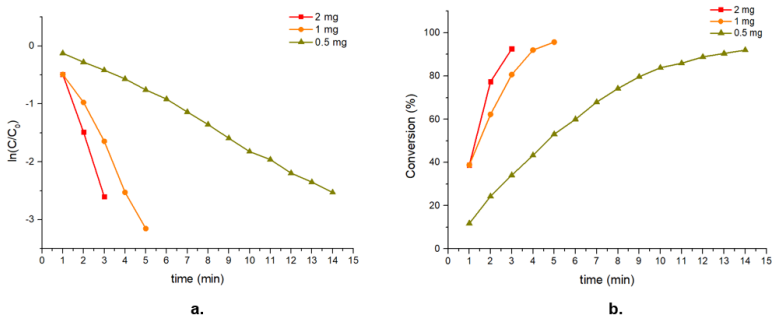


Figure S1 - Pseudo-first-order kinetic plot (a), and conversion plot (b) for the comparison of the activity of NiCuAl 1.5:1.5:1 oxide used in different amounts.

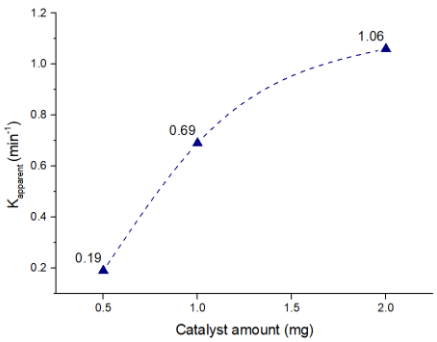


Figure S2 - Relationship between the apparent kinetic constant of the 4NP reduction and the amount of NiCuAl 1.5:1.5:1 oxide used as catalyst.

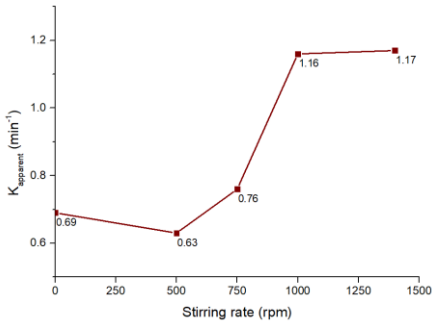


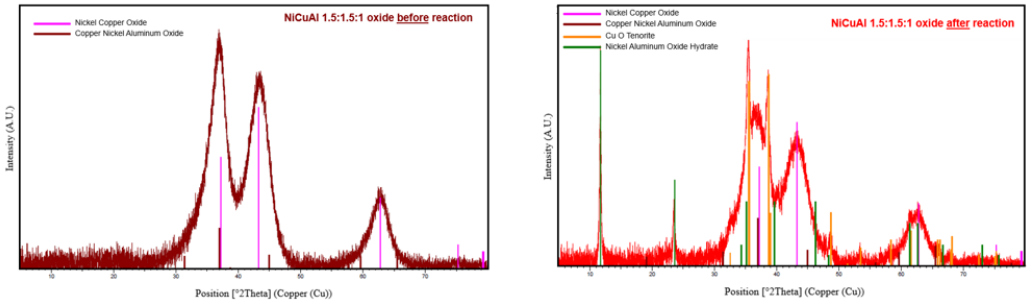
Figure S3 - Relationship between the apparent kinetic constant of the 4NP reduction and the stirring rate applied when NiCuAl 1.5:1.5:1 oxide is used as catalyst.

Table S2 – Apparent kinetic constant and conversion using NiCuAl 1.5:1.5:1 oxide as catalyst.

	σ [1]	K_{app} (min^{-1})
4-nitrophenol	1.22	0.69 ± 0.05
2-nitrophenol	0.13	0.48 ± 0.03
3-nitrophenol	-0.38	0.40 ± 0.05

Table S3 - Comparison of NiCuAl 1.5:1.5:1 oxide with other similar catalysts employed for 4NP reduction tests.

Catalyst	K_{app} (s^{-1})	Amount of catalyst (mg)	K (s^{-1}/mg)	Ref.
C_2H_2-treated Pd NS	$2.17 \cdot 10^{-3}$	-	-	J. Am, Chem. Soc., 2014 [2]
Au/OMS composite	$0.97 \cdot 10^{-3}$	20	**$4.85 \cdot 10^{-5}$**	J. Am, Chem. Soc., 2011[3]
p(Aam-co-TMT)@Au	$2.30 \cdot 10^{-3}$	-	-	Appl. Catal. B Environ., 2019 [4]
Au@g-C_3N_4	$1.5 \cdot 10^{-2}$	2.0	**$7.50 \cdot 10^{-3}$**	Appl. Catal. B Environ., 2019 [5]
Pt_3Au_1-PDA/RGO	$9.58 \cdot 10^{-3}$	-	-	Appl. Catal. B Environ., 2016 [6]
Au/g-C_3N_4-6 (500 W xenon lamp, λ>420 nm)	$7.990 \cdot 10^{-3}$	1.0	**$7.990 \cdot 10^{-3}$**	Appl. Catal. B Environ., 2017 [7]
α-CD-capped Au	$4.65 \cdot 10^{-3}$	-	-	J. Phys. Chem. C., 2009 [8]
PCP@Au-Ag	$2.87 \cdot 10^{-3}$	0.2	**$1.44 \cdot 10^{-2}$**	Master. Chem. Phys., 2018 [9]
PAM/PPY/GO-Ag	$2.10 \cdot 10^{-2}$	4.0	**$5.25 \cdot 10^{-3}$**	Appl. Surf. Sci., 2018 [10]
Au/CeO_2@ZrO_2	$2.41 \cdot 10^{-2}$	0.14	**$1.71 \cdot 10^{-1}$**	Appl. Catal. B Environ., 2015 [11]
Fe_3O_4@COF-Au	$3.7 \cdot 10^{-3}$	3.0	**$1.23 \cdot 10^{-3}$**	Appl. Catal. B Environ., 2020 [12]
NiCuAl oxide	**$1.2 \cdot 10^{-2}$**	**1.0**	**$1.2 \cdot 10^{-2}$**	**This work**



a. **b.**

Figure S4 - XRD patterns of NiCuAl 1.5:1.5:1 oxide before (a) and after (b) the 4NP reaction.

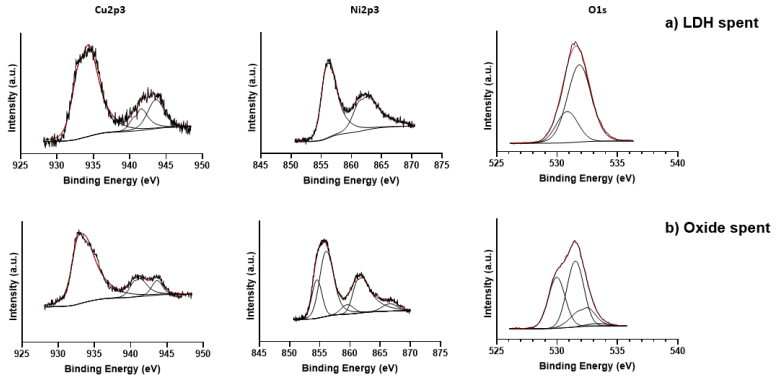


Figure S5 - XPS spectra of NiCuAl 1.5:1.5:1 a) LDH spent b)oxide spent.

Table S4 - Weight percentage (% w) of the species presents on the top-surface obtained by XPS analysis

Sample	O1s (% w)	Al2p (% w)	Ni2p3 (% w)	Cu2p3 (% w)
LDH	36.64	22.53	21.51	19.32
LDH spent	35.34	19.99	23.18	21.42
Oxide	24.92	28.37	24.71	25.75
Oxide spent	25.74	23.89	24.33	26.40

Table S5 - Weight Ratio of the species observed on the surface, by XPS analysis, for all the catalysts investigated

Sample	Ni/Al	Cu/Al	Cu/Ni	Cu/O	Ni/O
LDH	0.95	0.86	0.90	0.53	0.59
LDH spent	1.16	1.07	0.92	0.61	0.66
Oxide	0.87	0.91	1.04	1.03	0.99
Oxide spent	1.01	1.11	1.09	1.03	0.94

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