Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

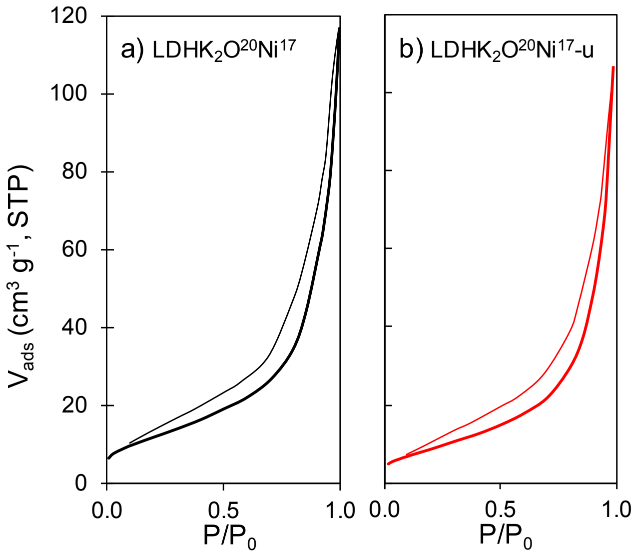

Figure S1: N_2 adsorption-desorption isotherms of the catalysts before (a) and after used (b) in BRM. Colors have been used for better visibility.

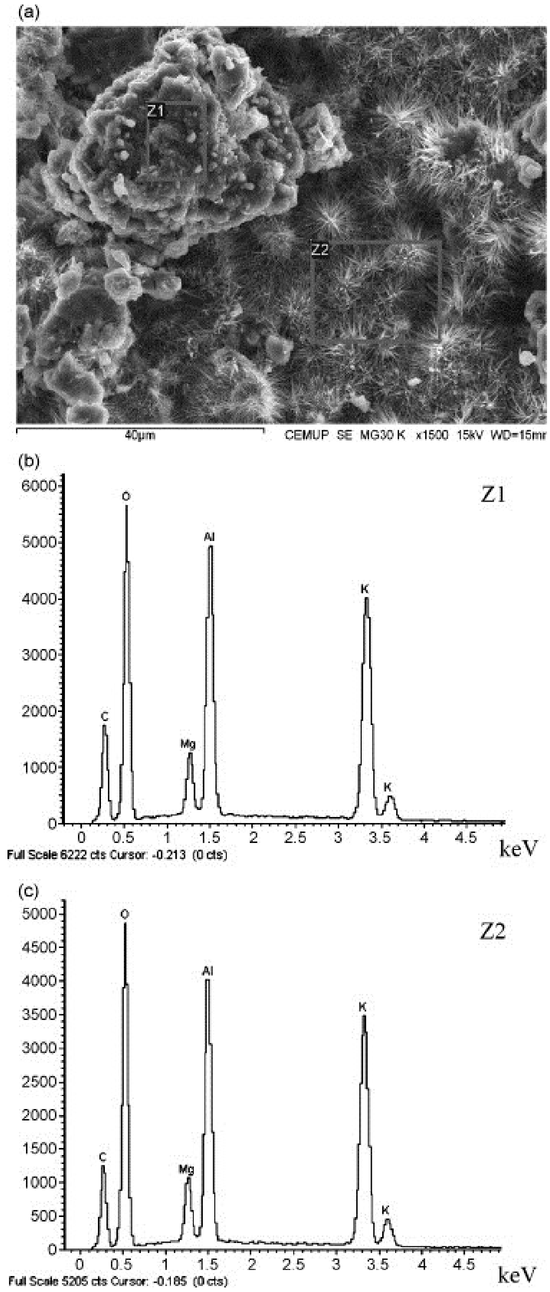

Figure S2: (a) SEM of MG30-K; EDX analysis of areas: (b) Z1 and (c) Z2. Adapted from [29].

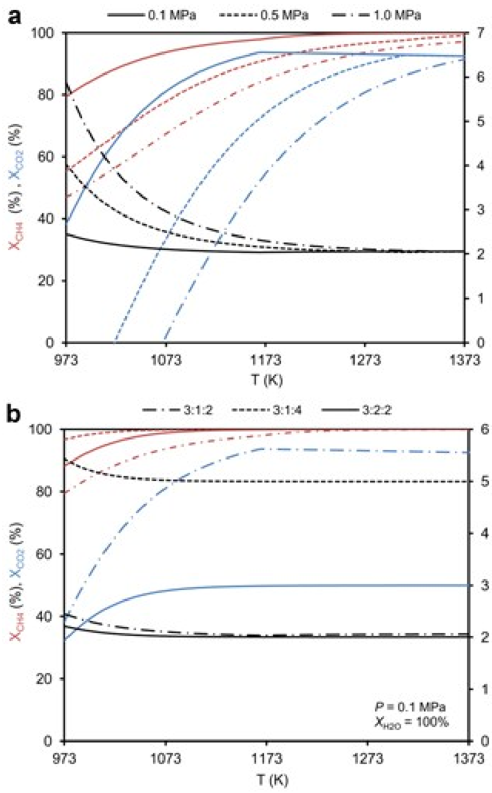

Figure S3: Conversion of CH_4 and CO_2, and H_2 to CO ratio versus temperature: (a) effect of *P* and (b) effect of CH_4:CO_2:H_2O ratios. Reproduced from [9].

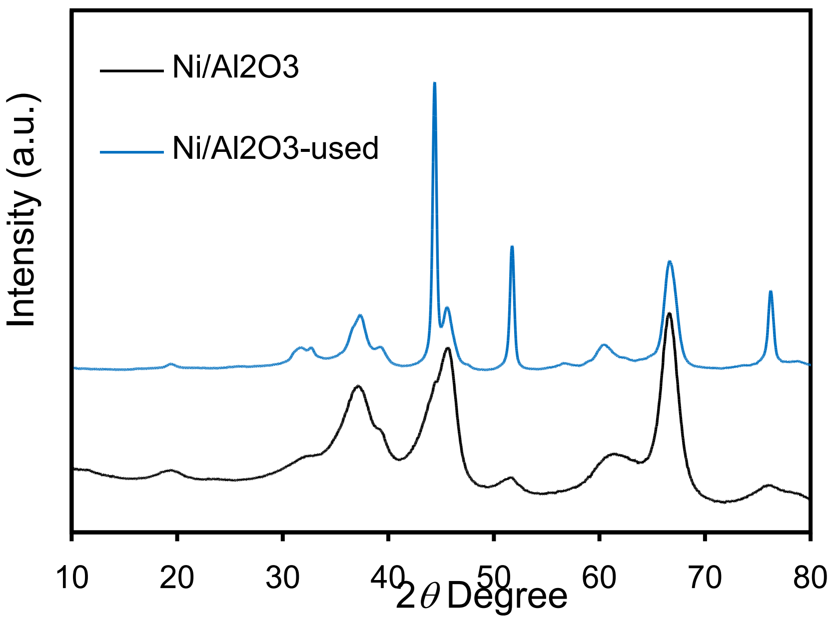

Figure S4: XRD patterns of catalyst used in a previous work [10].

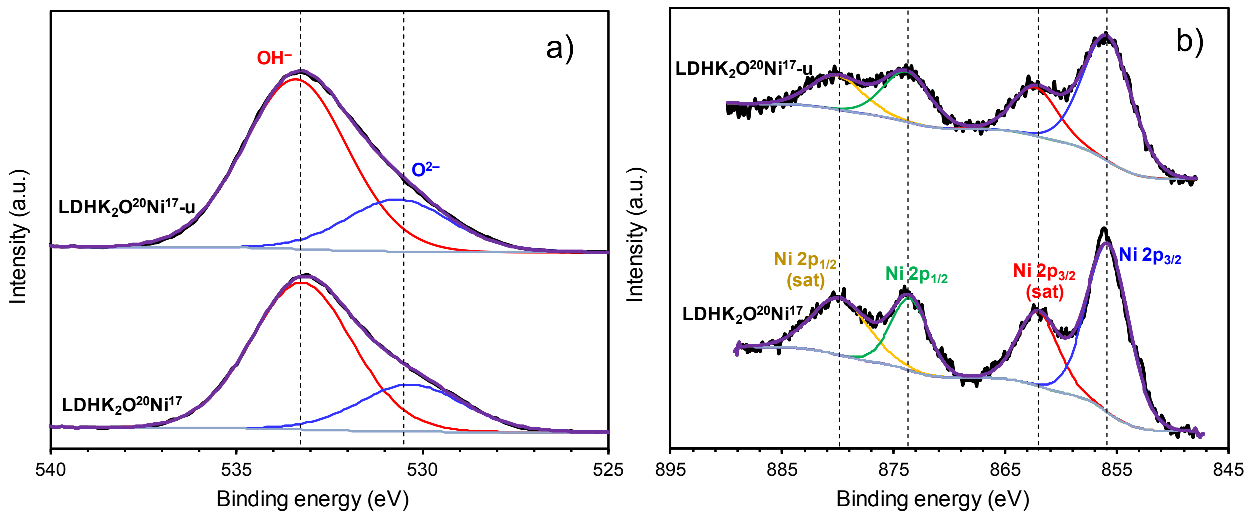

Figure S5: XPS spectra and deconvolution of the (a) O1s region and (b) Ni2p region of the different samples.

Table S1: Phases identified in the materials used before and after the BRM tests.

Material	Treatment	Phase	2θ positions and reflections	PDF-File ICDD
LDH	-	$Mg_2Al_2(OH)_8CO_3{\cdot}xH_2O$	12.4º (003), 29.8º (231), 34.9º (012), 38.8º (015), 60.9º (110)	43-0072
		AlO(OH)*	26.3º (010), 27.0º (110), 30.9º (101), 35.0º (200), 44.7º (210), 65.2º (002)	48-0890
$LDHK_2O^{20}$	H_2 reduced	$KOH{\cdot}H_2O$	22.1º (110), 30.5º (020), 33.3º (102), 45.8º (122)	36-0791
		KOH	32.2º (002), 32.4º (110), 65.2º (113)	21-0645
		$Mg_2Al_2(OH)_8CO_3{\cdot}xH_2O$	12.4º (003), 29.8º (231), 34.9º (012), 38.8º (015), 60.9º (110)	43-0072
		AlO(OH)*	26.3º (010), 27.0º (110), 30.9º (101), 35.0º (200), 44.7º (210), 65.2º (002)	48-0890
$LDHK_2O^{20}Ni^{17}$	H_2 reduced	Ni	44.5° (111), 52.0° (200), 76.4° (220)	70-1849
		NiO	37.3° (111), 43.5° (200), 75.3° (311), 79.3° (222)	73-1523
		$Mg_6Al_2(CO_3)(OH)_{16}{\cdot}4H_2O$	10.3° (006), 21.8° (018), 4.7° (024), 48.9° (211)	41-1428
		AlO(OH)*	14.6° (010), 28.2° (110), 8.4° (101), 49.6° (210), 65.4° (002)	49-0133
$LDHK_2O^{20}Ni^{17}$-u	After BRM	Ni	44.5° (111), 52.0° (200), 76.4° (220)	70-1849
		NiO	37.3° (111), 43.2° (200), 75.3° (311), 79.1° (222)	73-1523
		AlO(OH)*	14.8° (010), 28.7° (110), 49.2° (210), 65.7° (002)	49-0133
		$MgAl_2O_4$	30.9° (220), 36.8° (311), 65.4° (440)	77-1193

* Due to the high aluminum content, PURAL MG30 contains a significant amount of boehmite (*Product information*, Sasol Germany (formerly Condea), Hamburg **2012, http://www.sasoltechdata.com/tds/PURAL-MG.pdf**)

Supporting Experimental Section

Materials, characterization, and reaction studies.

The sorbent PURAL MG30 (aluminium magnesium hydroxide, 70% Al_2O_3) and PURAL MG30 K_2CO_3 (aluminium magnesium hydroxide, 70% Al_2O_3; potassium carbonate, 20% K) was provided by CONDEA Chemie Germany (now SASOL).

The support material was impregnated with nickel(II) nitrate hexahydrate (VWR international – Merck group Germany).

Tubular stainless steel fixed-bed continuous down-flow reactor was an home-made equipment produced by Neves&neves Lda. Trofa in Portugal.

The heating furnace with a PID temperature controller is a product from Termolab-Fornos Electricos Lda, Águeda in Portugal.

An HPLC pump (Merck L-2130 (Hitachi, Tokyo, Japan) was used to introduce the liquid water inside the reactor.

The composition of the outgoing off-gas stream was determined with a gas chromatograph (GC 1000, Dani Chromatographs) equipped with an on-line multiport 16-valve system for sample injection (Valco Instruments Company Inc.), a capillary column (Carboxen 1010 Plot, Supelco) and a thermal conductivity detector.

The gases were furnished by Air Liquid Portugal with the following purities: methane N35 (99.95%), hydrogen N35 (99.95%), carbon dioxide N48 (99.998%), nitrogen N35 (99.95%), carbon monoxide N35 (99.95%) and helium ALPHAGAZ 2 (99.9998%).