Supplementary Information for

A new material built with alternating Cu sulfide and (Al,Mg) hydroxide molecular sheets: hydrothermal synthesis and selected characteristics

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Figure S1. A 3D (a) and 2D (b) representation of tapping mode AFM images along with corresponding phase contrast one acquired from a group of flakes of material synthesized at the Cu/Mg/Al/S precursor ratio of 4/2/1/15 (sample a, see Table 1 of the manuscript).



Figure S2. A TEM image with marked areas corresponding to EDS analysis results summarized in Table S1 (see below).

Spectrum	In stats.	0	Mg	Al	S	Ni*	Cu
Spectrum 1	Yes	57.27	2.21		0.91	6.43	33.19
Spectrum 2	Yes	56.36	1.80		1.62	4.95	35.27
Spectrum 3	Yes	55.56	25.61	10.30	3.76	2.61	2.16
Spectrum 4	Yes	29.84	15.25	6.23	19.84	5.67	23.17
Spectrum 5	Yes	37.45	13.48	4.71	16.72	4.01	23.62

Table S1. Atomic concentrations of the product as determined using EDS.

*Ni signal appears due to Ni TEM grid



Figure S3. Two typical HRTEM images acquired from sample a. On the left panel, a fragment of HRTEM image of valleriite superimposed on that of the new synthesized $Cu_{4-x}S_2@(Mg,Al)(OH)_2$ phase is shown. An enlarged picture with both sulfide and hydroxide sheets designations is displayed on the right panel.



Figure S4. XRD pattern and corresponding Le Bail refinement for the products of heating of sample a to 1000 °C.



Figure S5. A common view SEM image (left top panel) acquired from sample f along with corresponding Cu K α , S K α , Mg K α , Al K α , and O K α maps.



Figure S6. XRD patterns acquired from sample f (curve a), and sample g (curve b).



Figure S7. UV-vis-NIR spectra recorded from sample a (curve a), sample f (curve b), and sample g (curve c) (A), along the corresponding Tauc plots used for the optical band gap (E_g) calculations. Numbers on the right plot are the calculated E_g values. From the Tauc plots, it can be seen that although absolute values of absorbance are somewhat different (due to somewhat different colloidal stability of suspensions under examination), the points in which linear fitting curves cross the *x*-axis are near to each other, giving rise to the near values of band gaps.