Supporting Information

Spontaneous Formation of Hierarchically Structured Curly Films of Nickel Carbonate Hydrate through Drying**

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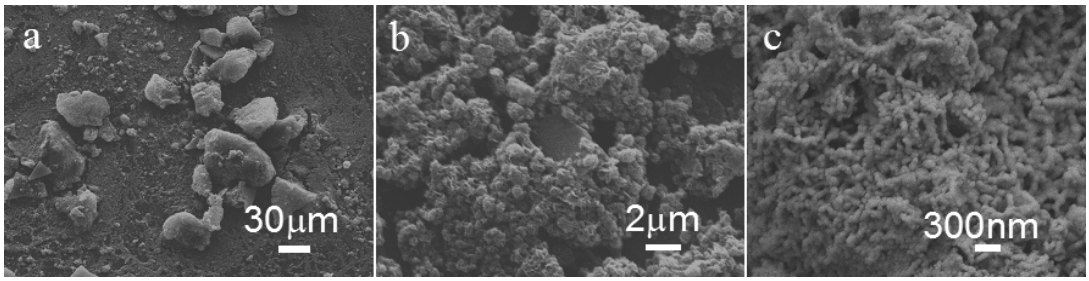

Figure S1. SEM images of nickel carbonate hydrate formed in bulk solution after crystallization for 2 days at ambient temperature in the presence of polymer A (PEG-PEI-$(CH_2$-$(CH$-$OH)_3$-$CH_2CONHNH_2)_3$). [polymer A] = 1.0 g·L^{-1}. [$NiCl_2$] = 10 mM. (a) a full view of irregular aggregates. (b) higher magnification SEM image of the aggregates. (c) high magnification SEM image of irregular slab-shaped aggregate.

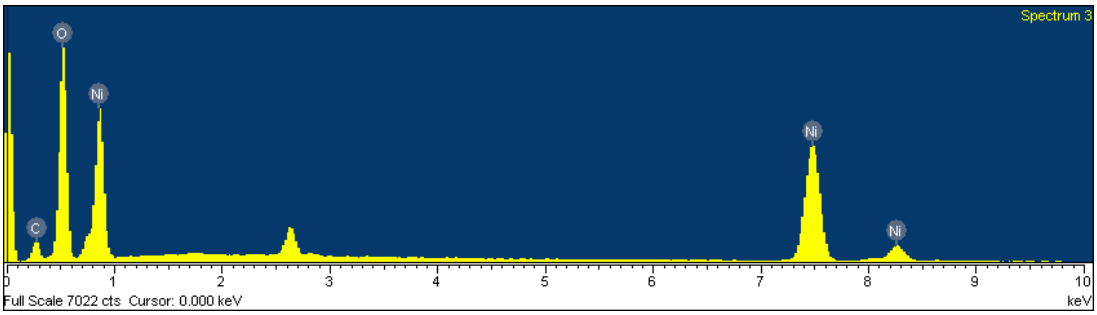

Figure S2. EDS result of the film-like sample of nickel carbonate hydrate formed at the air/glass slice after drying of the films formed at air/solution interface in the presence of polymer A (PEG-PEI-$(CH_2$-$(CH$-$OH)_3$-$CH_2CONHNH_2)_3$). [polymer A] = 1.0 g·L^{-1}, [$NiCl_2$] = 10 mM. The crystallization reaction proceeded for 2 days at ambient temperature.

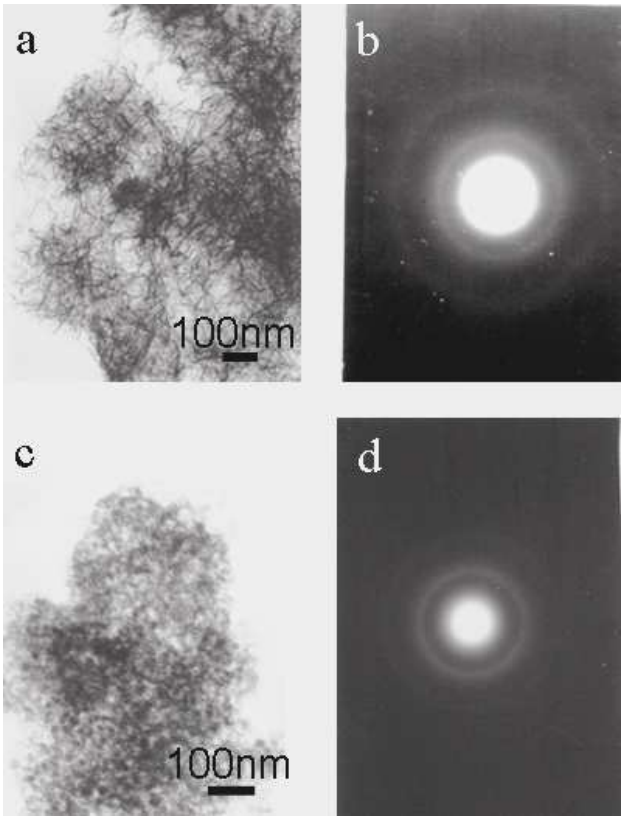

Figure S3. TEM images and electron diffraction (ED) patterns of the film-like sample of nickel carbonate hydrate formed at the air/glass slice after drying of the films formed at air/solution interface in the presence of polymer A (PEG-PEI-(CH_2-(CH-OH)$_3$-$CH_2CONHNH_2$)$_3$) with different concentration. [$NiCl_2$] = 10 mM. The crystallization reaction proceeded for 2 days at ambient temperature. (a) 1.0 g·L^{-1}, (b) ED pattern for the sample shown in (a). (c) 2.0 g.L^{-1}, and (d) ED pattern shown in (c).

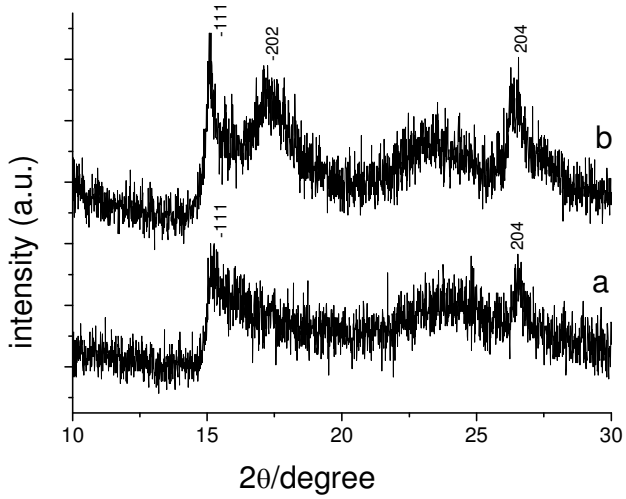

Figure S4. XRD Patterns of the film-like sample of nickel carbonate hydrate formed at the air/glass slice after drying of the films formed at air/solution interface in the presence of polymer A (PEG-PEI-(CH_2-(CH-OH)$_3$-$CH_2CONHNH_2$)$_3$) using different nickel salts as nickel resource. (a) Nickel nitrate and (b) Nickel acetate. [polymer A] = 1.0 g · L^{-1}. The crystallization reaction proceeded for 2 days at ambient temperature.

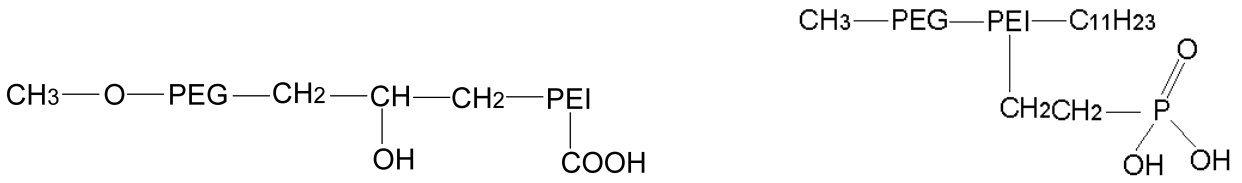

Figure S5. The chemical structural formula of two polymer additives shown in Table 1. Left: PEG-PEI-$(CH_2\text{-}COOH)_n$ (polymer B); Right, PEG-PEI-$(COC_{11}H_{23})(CH_2CH_2PO(OH)_2)_n$ (polymer C).

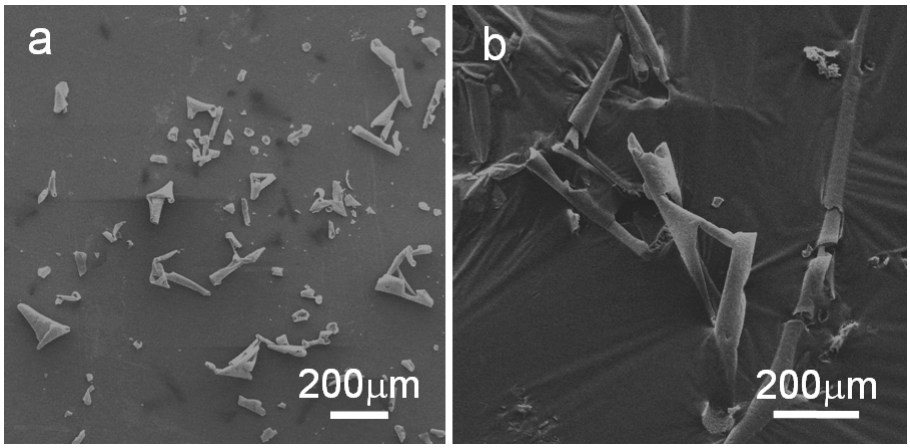

Figure S6. SEM images of the film-like sample of nickel carbonate hydrate formed at the air/glass slice after drying of the films formed at air/solution interface in the presence of other additives. (a) Dextrin, (b) PAA. The concentration of the additives used are 1 $g{\cdot}L^{-1}$, the other experimental conditions were kept the same to that depicted in Figure 1. $[NiCl_2]$ = 10 mM. The samples formed by mineralization for 2 days at ambient temperature.

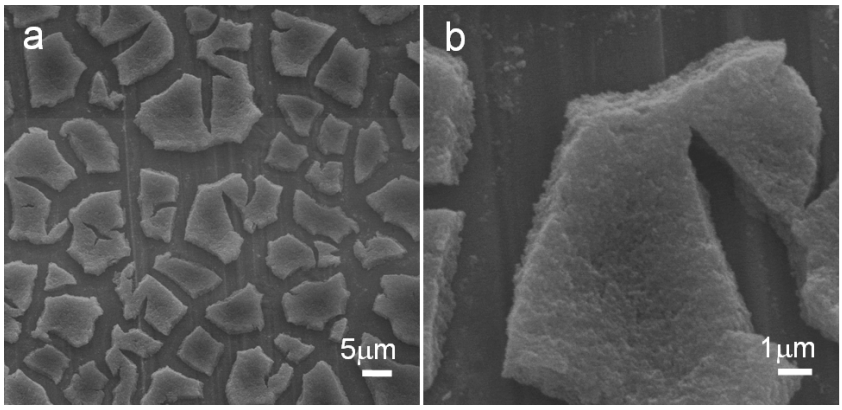

Figure S7. SEM images of nickel carbonate hydrate samples obtained through directly mixed nickel chloride and polymer A (1.0 $g.L^{-1}$), then slowly adjusting the pH value of the resulting solution to 8 by using sodium hydroxide. $[NiCl_2]$ = 10 mM.