Supporting Information

Nanocellulose reinforced organo-inorganic nanocomposite for synergistic and affordable defluoridation of water and an evaluation of its sustainability metrics

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SUPPORTING INFORMATION CONTENT

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Equation (1): The maximum uptake of F^- (q_e) by cellulosic composites was calculated using the equation given below:

$$Uptake\ (q_e) = \frac{(C_o - C_e)V}{m}$$

where q_e is the amount of F^- ions adsorbed per gram of the adsorbent (mg/g) at equilibrium, C_e is the equilibrium concentration of As(III)/As(V) in the bulk solution (mg/L), C_o is the initial F^- concentration (mg/L), V is the volume of solution (L) and m is the mass of the adsorbent (g).

Equation (2): The linearized form of Langmuir equation used in this work is defined as,

$$\frac{C_e}{q_e} = \frac{C_e}{q_{max}} + \frac{1}{bq_{max}}$$

where q_e is the amount of adsorption at the surface of the adsorbent (mg/g), C_e is the equilibrium concentration of the solution (mg/L), q_{max} is the maximum surface density at monolayer coverage and b is the Langmuir adsorption constant (L/mg) related to the free energy of adsorption and $1/q_{max}$ and $1/bq_{max}$ are the Langmuir constants.

Equation (3): The removal % of F^- was calculated using the equation mentioned below:

$$Removal\ \% = \frac{C_o - C_e}{C_o} \times 100$$

where C_o and C_e are the initial and equilibrium concentrations of the metal ions, respectively.

Sustainability metrics equations

Equation (4): $$Mass\ intensity = \frac{mass\ of\ all\ reactants\ used\ excluding\ water}{mass\ of\ product}\ kg/kg\ product$$

Equation (5): $Water\ intensity\ (W_P) = \frac{mass\ of\ all\ water\ used}{mass\ of\ product}\ kg/kg\ product$

Equation (6): $Reaction\ mass\ efficiency\ (RME) = \frac{\text{mass of product}}{\text{mass of all reactants}} \times 100\%$

Equation (7): $Energy\ Intensity = \frac{amount\ of\ non\ renewable\ energy\ used}{mass\ of\ product}\ kW.h/kg$

Equation (8): $E\ factor = \frac{[kg(raw\ materials) - kg(desired\ product)]}{kg(total\ product\ including\ water)}$

Equation (9): *Lagergren pseudo-first-order model:* $\ln(q_e - q_t) = \ln q_e - k_1 t$

Equation (10): *Ho's pseudo-second-order model:* $\frac{t}{q_e} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$

Equation (11): *Freundlich adsorption isotherm model*: $\log Qe = \log Kf + \frac{1}{n} \log Ce$

Equation (12): *Dubinin–Radushkevich isotherm model:* $lnQe = lnQ_s - K_{ad}\varepsilon^2$

Equation (13): *Gibbs free energy equation:* $\Delta G° = \Delta H° - T\Delta S°$

Equation (14): *Equation for thermodynamic parameters:* $\ln k = (-\Delta H°)/RT + (\Delta S°)/R$

(a) (b)

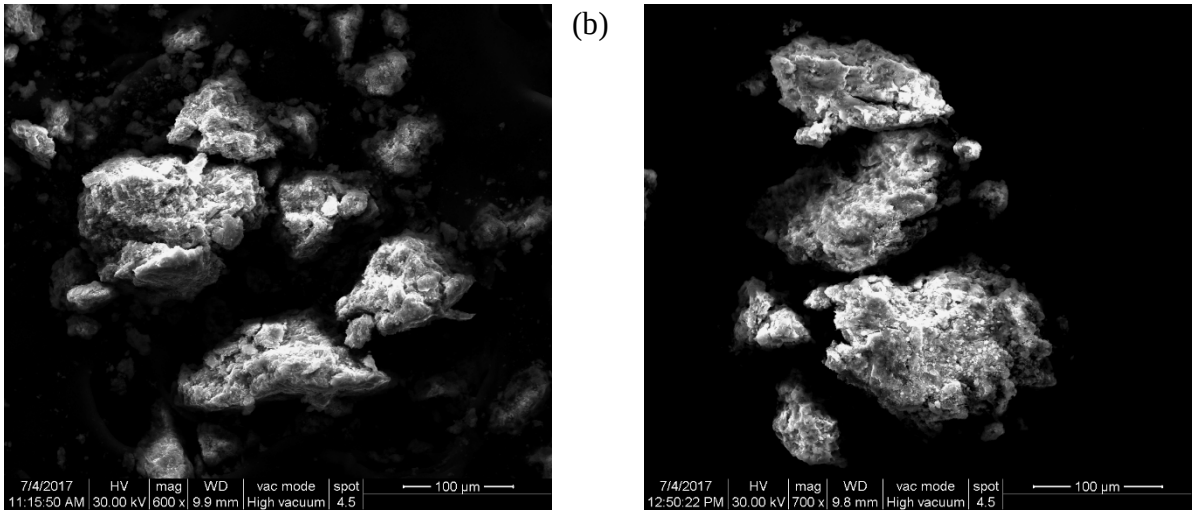


Figure S1: Large area SEM images of CNPFH (a) before and (b) after F- adsorption.

(a) (b)

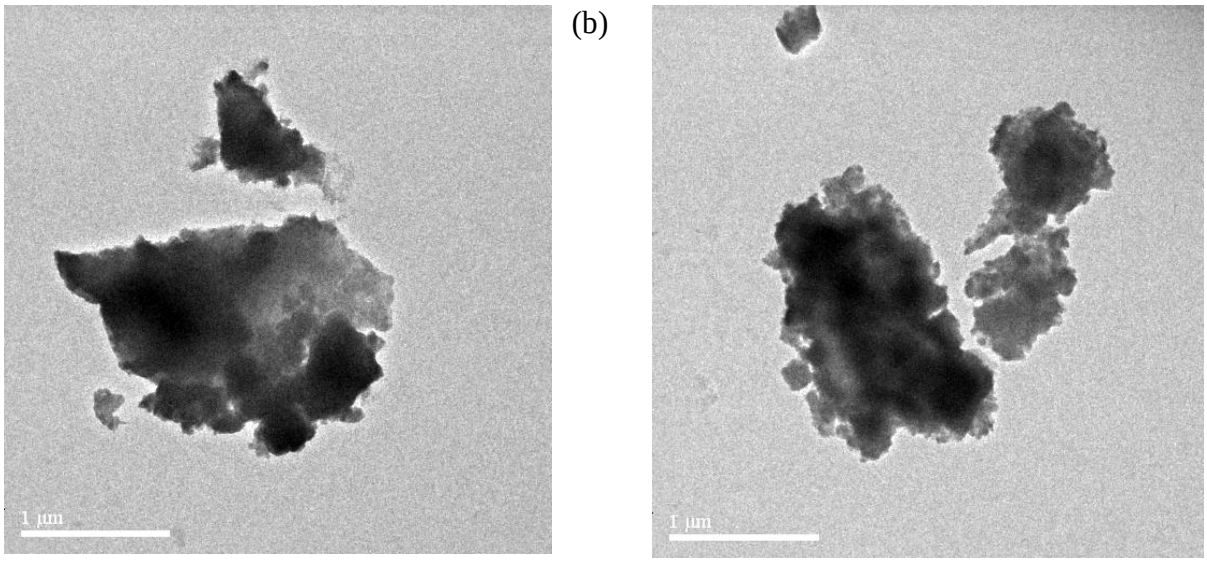


Figure S2: TEM images of CNPFH (a) before and (b) after F- adsorption, corresponding to elemental mapping shown in Figure 1 (C) and (D).

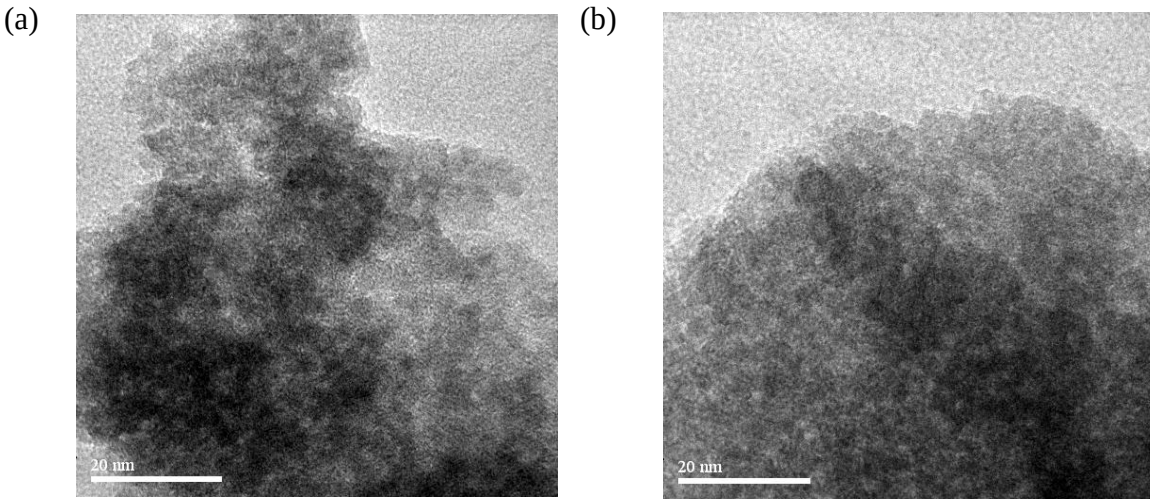


Figure S3. TEM images of as-synthesized CNPFH composite (a) before and (b) after F-adsorption.

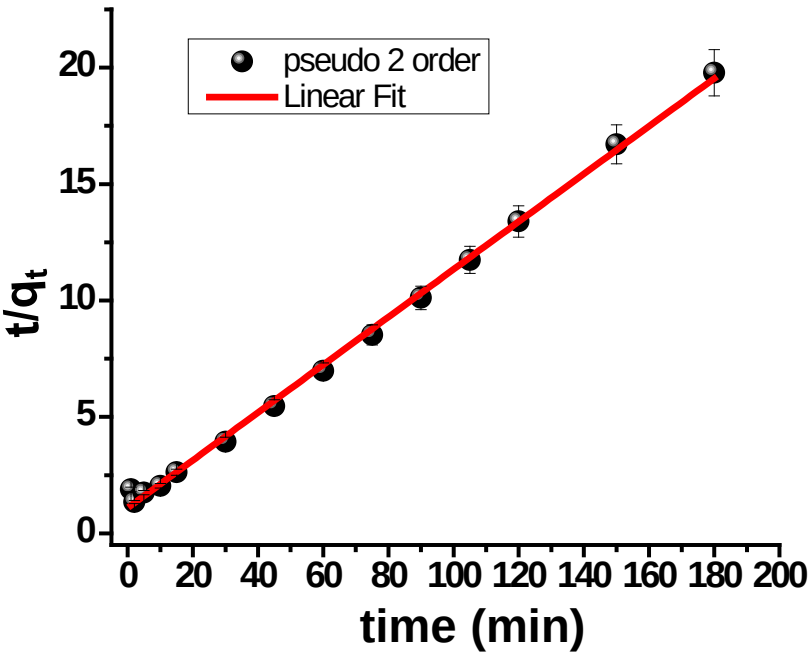


Figure S4. Pseudo second-order reaction kinetic plot for the adsorption of F^- on CNPFH, fitted with Eq. 10.

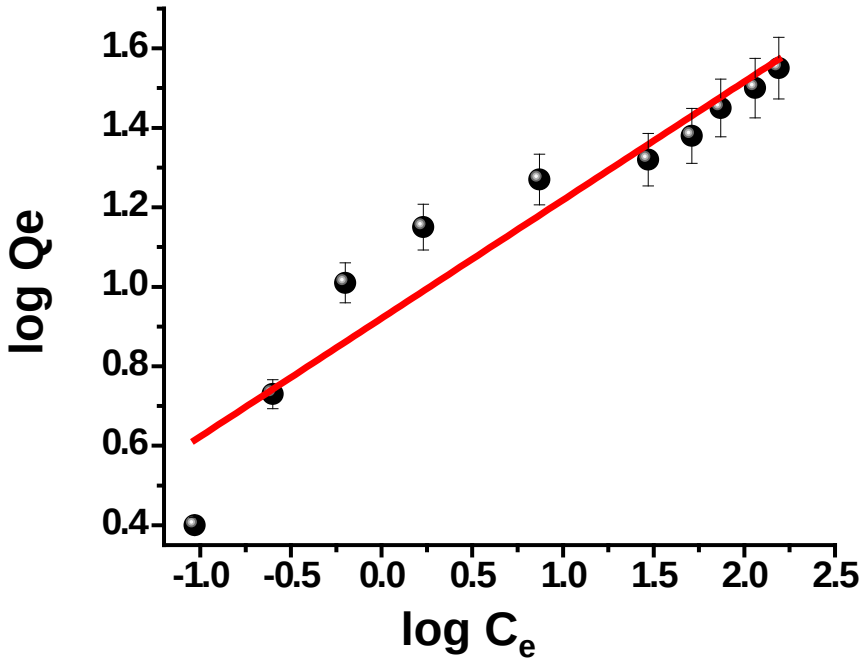
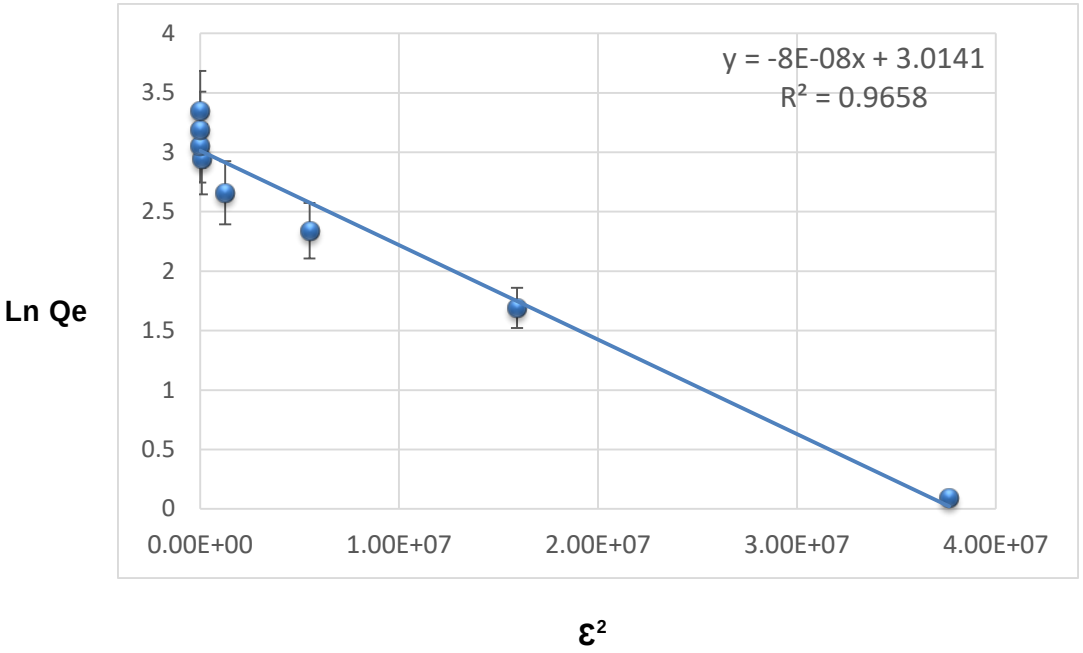


Figure S5. Freundlich isotherm plot for the adsorption of F^- on CNPFH, fitted with Eq. 11.

Table S1. Summary of isotherms and kinetic parameters of F^- adsorption on CNPFH.

	Model	Formulae	k (rate constant)	q_e (Adsorption capacity) (mg/g)	R^2
Kinetics	Pseudo 1st order	$\ln(q_e - q_t) = \ln q_e - k_1 t$	0.02	6.5	0.891
	Pseudo 2nd order	$\frac{t}{q} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$	0.01	9.8	0.997
	Model	**Formulae**	**n (isotherm constant)**	**Adsorption capacity**	**R^2**
Isotherm	Langmuir	$\frac{C_e}{q_e} = \frac{C_e}{q_{max}} + \frac{1}{n q_{max}}$	0.04	50.8 mg/g	0.969
	Freundlich	$\log q_e = \log K_f + \frac{1}{n} \log C_e$	3.3	8.3 ($mg^{1-1/n} L^{1/n}$ /g)	0.899



E_{DR} = 2500kJ/mol, R^2 = 0.96

Figure S6. Dubinin–Radushkevich isotherm model for F- adsorption on CNPFH using Eq. 12.

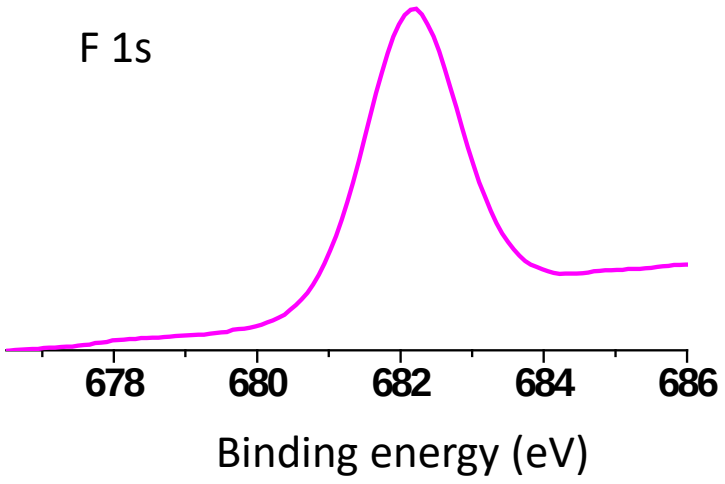


Figure S7. Deconvoluted XPS spectrum of F 1s for F^- adsorbed FeOOH-cellulose composite.

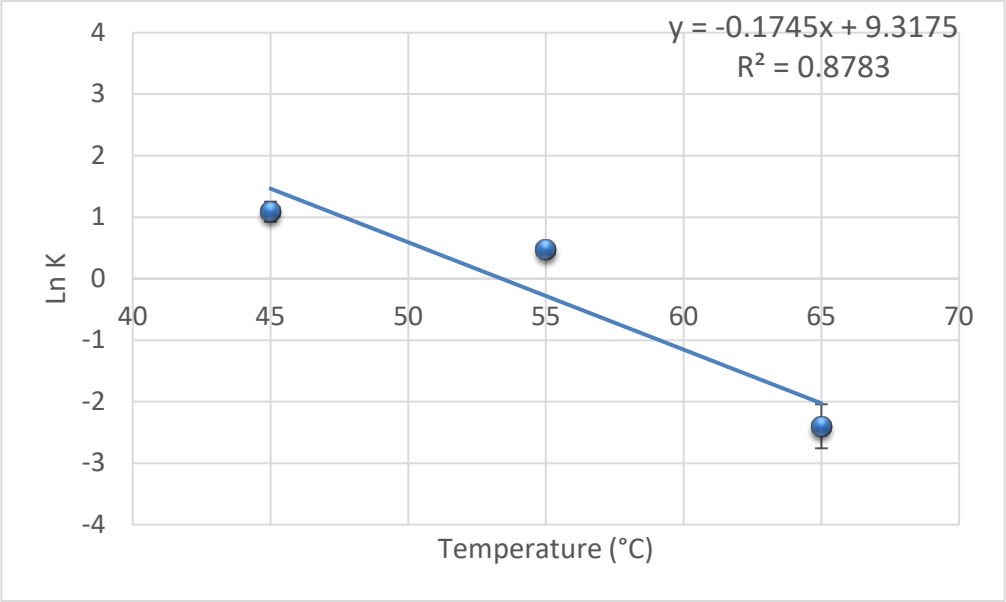


Figure S8. Thermodynamic plot for the adsorption of F^- on CNPFH, using Eq. 14.

ΔS = 0.077 kJ/mol .K, ΔH = 0.002 kJ/mol

Table S2.

Physicochemical characteristics of influent natural water.

Sl no.	Parameters	Value
1	Total coliforms (CFU/ml)	1-2 * 10^3
2	pH at 25°C	7.8
3	Conductivity (µS/cm)	640.0
4	Fluoride (mg/L)	0.57
5	Chloride (mg/L)	86.34
6	Nitrate (mg/L)	1.84
7	Sulfate (mg/L)	32.41
8	Silicate (mg/L)	15.87
9	Phosphate (mg/L)	55.83
10	Sodium (mg/L)	53.74
11	Potassium (mg/L)	2.33
12	Magnesium (mg/L)	14.34
13	Calcium (mg/L)	28.72