Supporting Information

A Cellulosic Ternary Nanocomposite for Affordable and Sustainable Fluoride Removal

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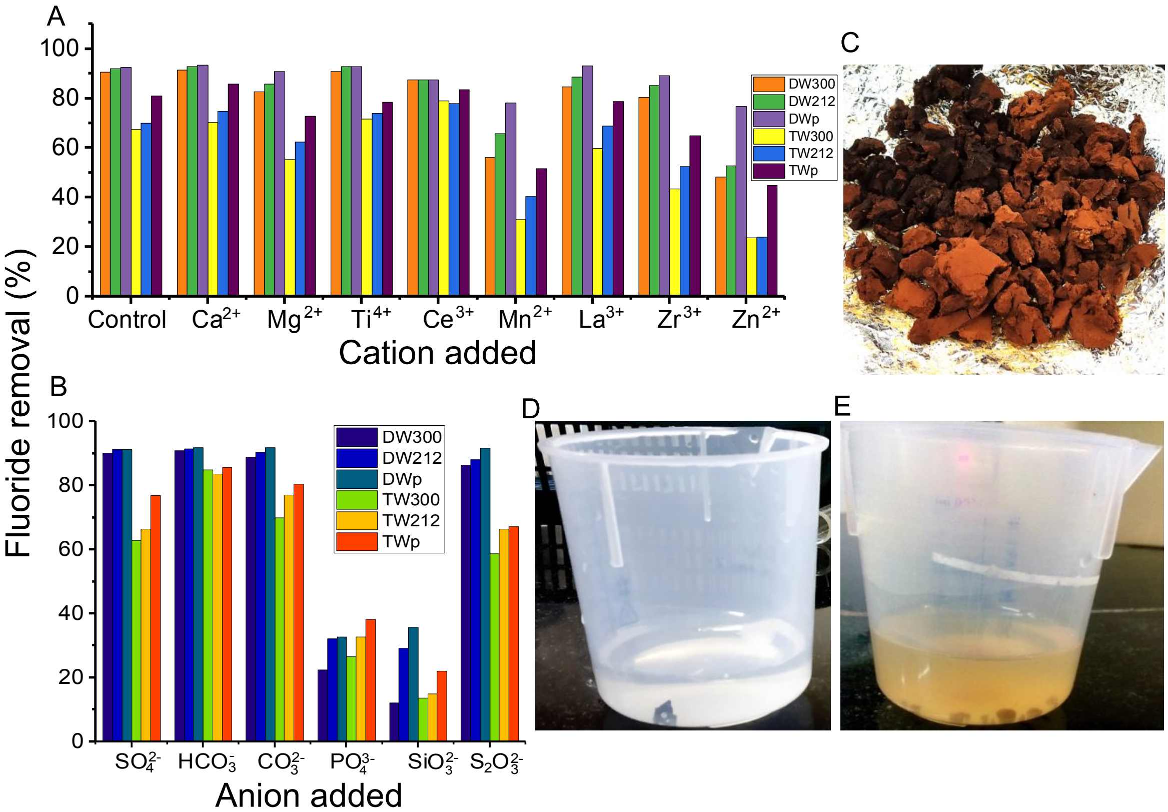


Figure S1. Effect of addition of other polyvalent cations (A) and other polyatomic anions (B) on the fluoride removal capacity of CAlFeC, tested with fluoride solution in distilled water (DW) and tap water (TW) and with particle sizes of 300 µm, 212 µm and powder, respectively. (C) Physical appearance of dried CAlFeC. (D and E) Show the stable and unstable composite, respectively, when shaken with water.

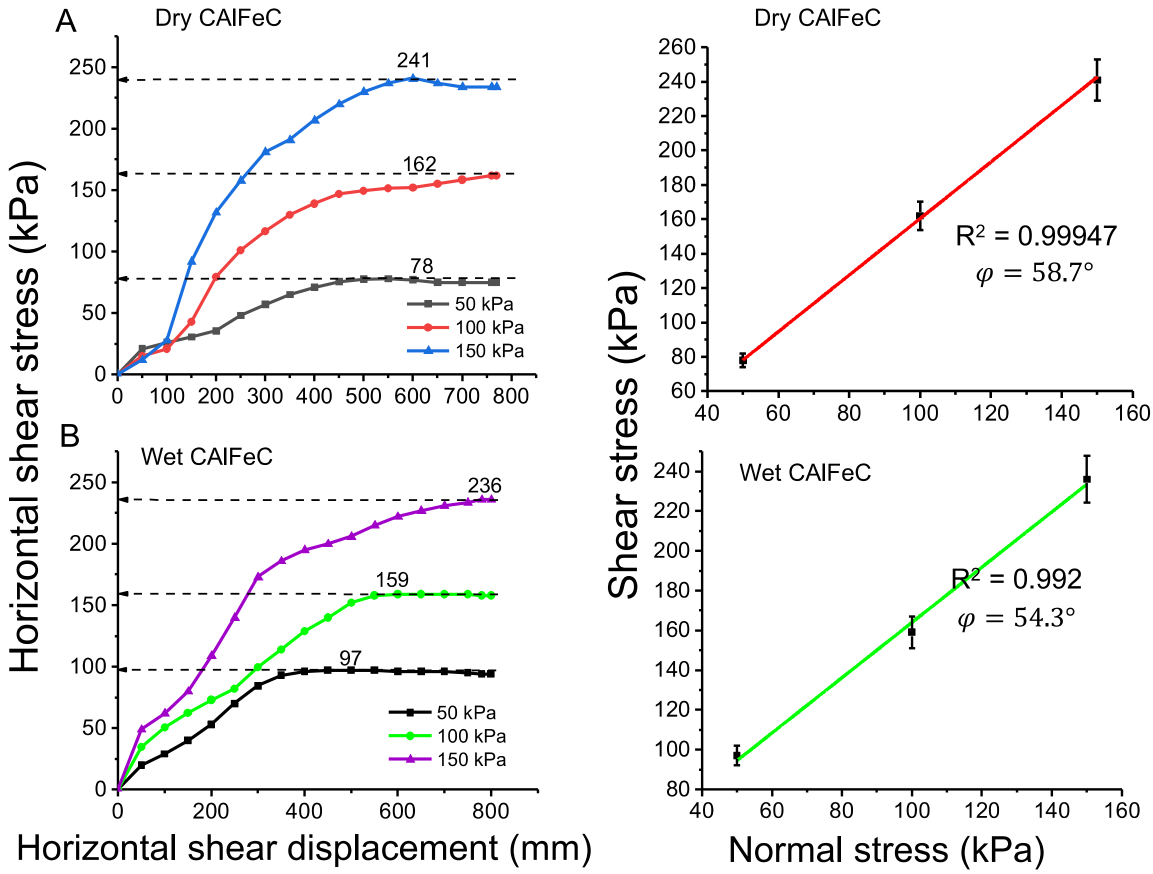


Figure S2. Direct shear stress test results of dry CAlFeC (A) and wet CAlFeC (B) and their corresponding Mohr-Coulomb failure patterns.

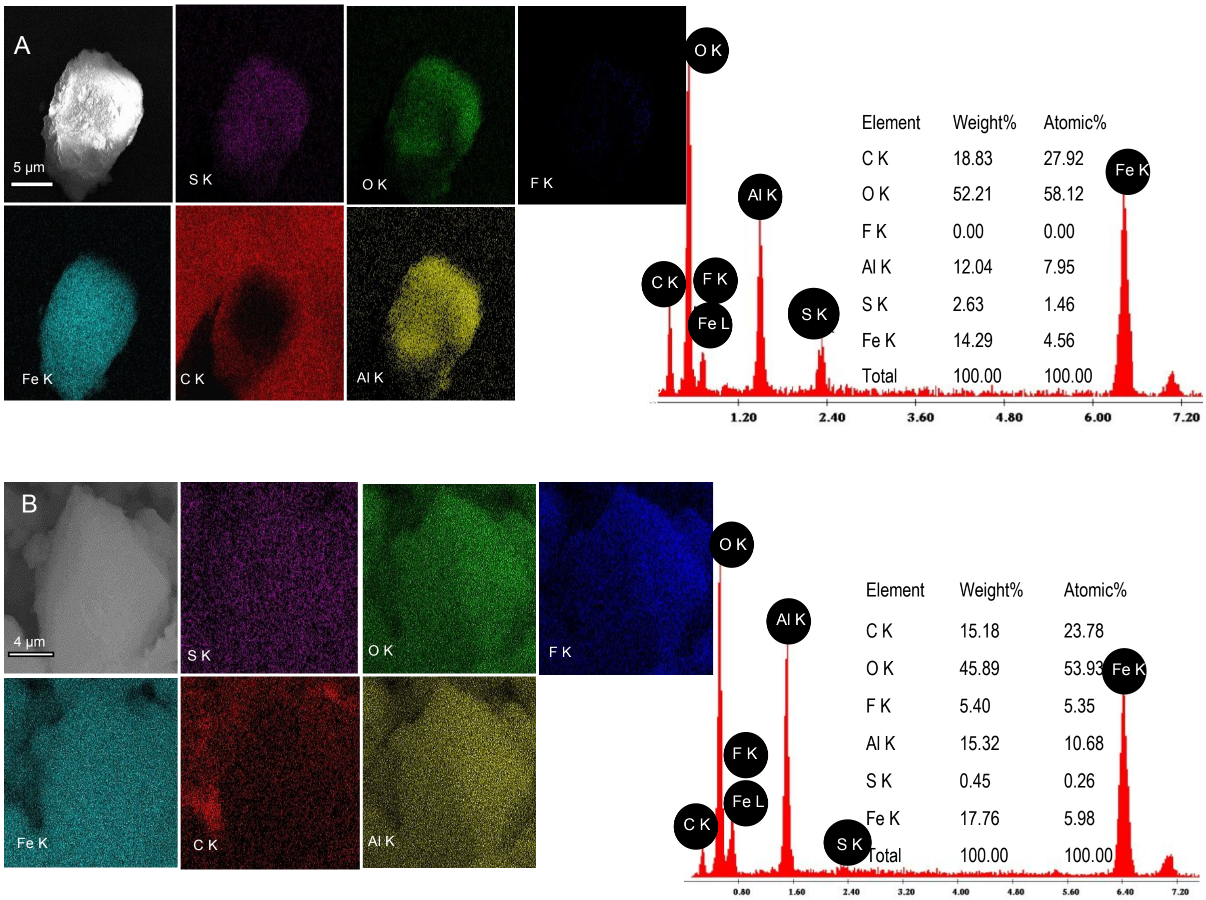


Figure S3. SEM-EDS elemental maps and spectra of CAlFeC (A) and CAlFeC-F (B).

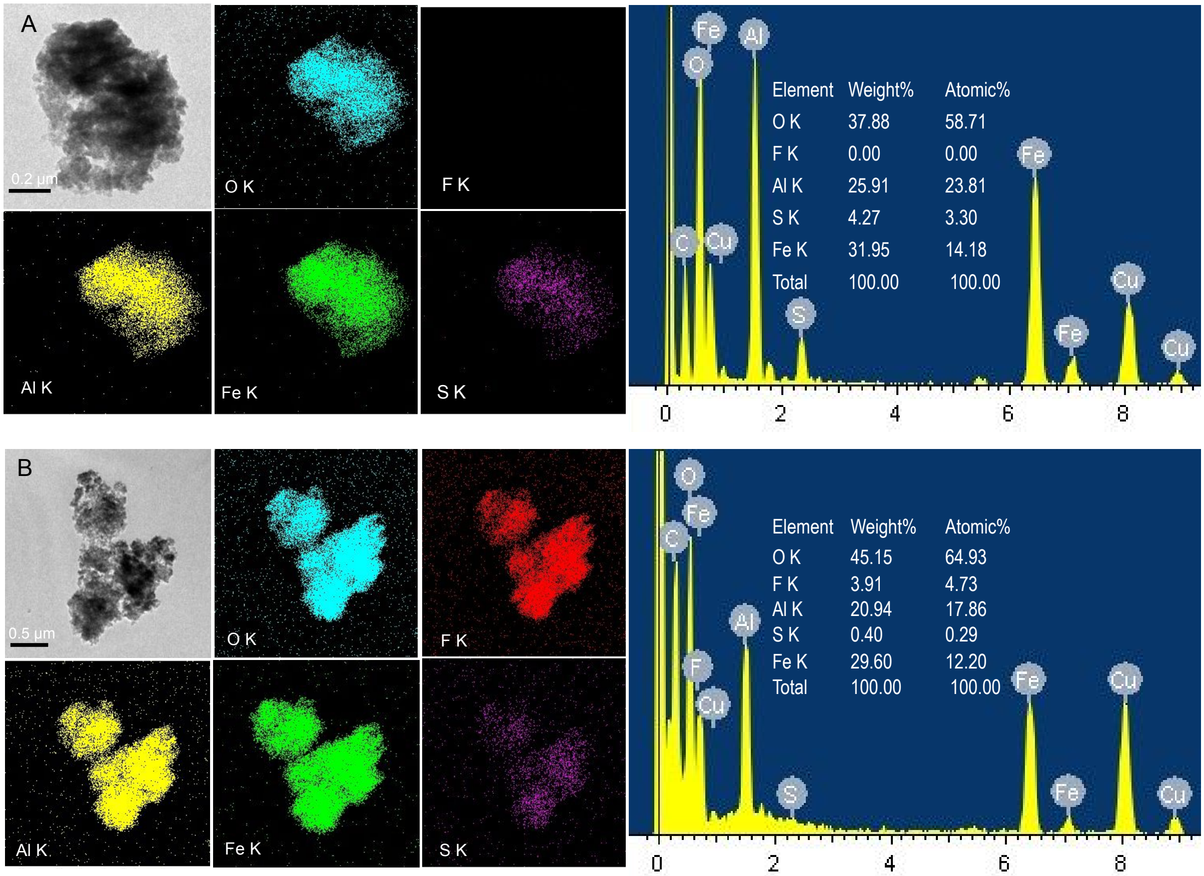
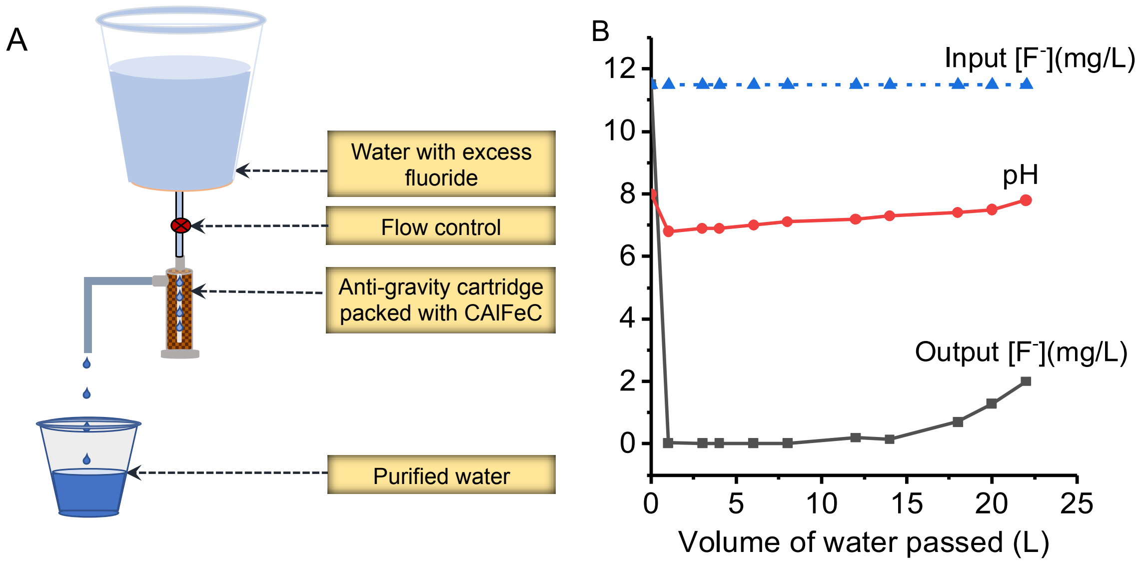


Figure S4. HRTEM-EDS element maps and spectra of CAlFeC (A) and CAlFeC-F (B).



C

	TOC/mg/L	TC/mg/L	IC/mg/L
Ultrapure water	-0.208	0.000	0.208
Ultrapure water + CAlFeC	0.258	0.341	0.83
10 mg/L F^- in ultrapure water + CAlFeC	2.725	3.054	0.3289

Figure S5. Cartridge experiment setup (A), changes in pH and fluoride levels during cartridge run (B) and the results of TOC leaching measurements (C).

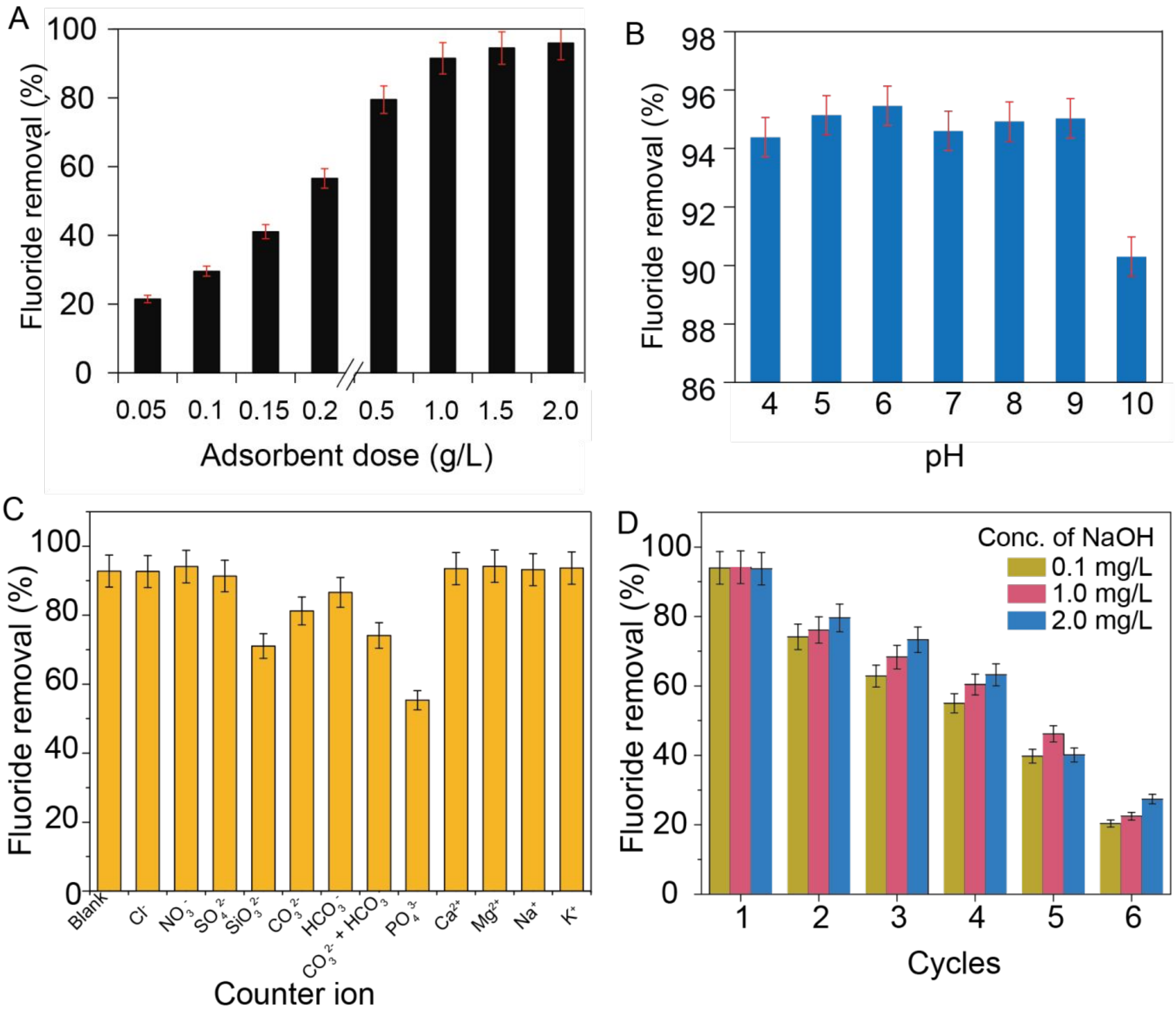


Figure S6. Fluoride adsorption characteristics of CAlFeC showing the effect of adsorbent dose (A), pH of the medium (B), counter-ions (C), and the effect of regeneration (D).

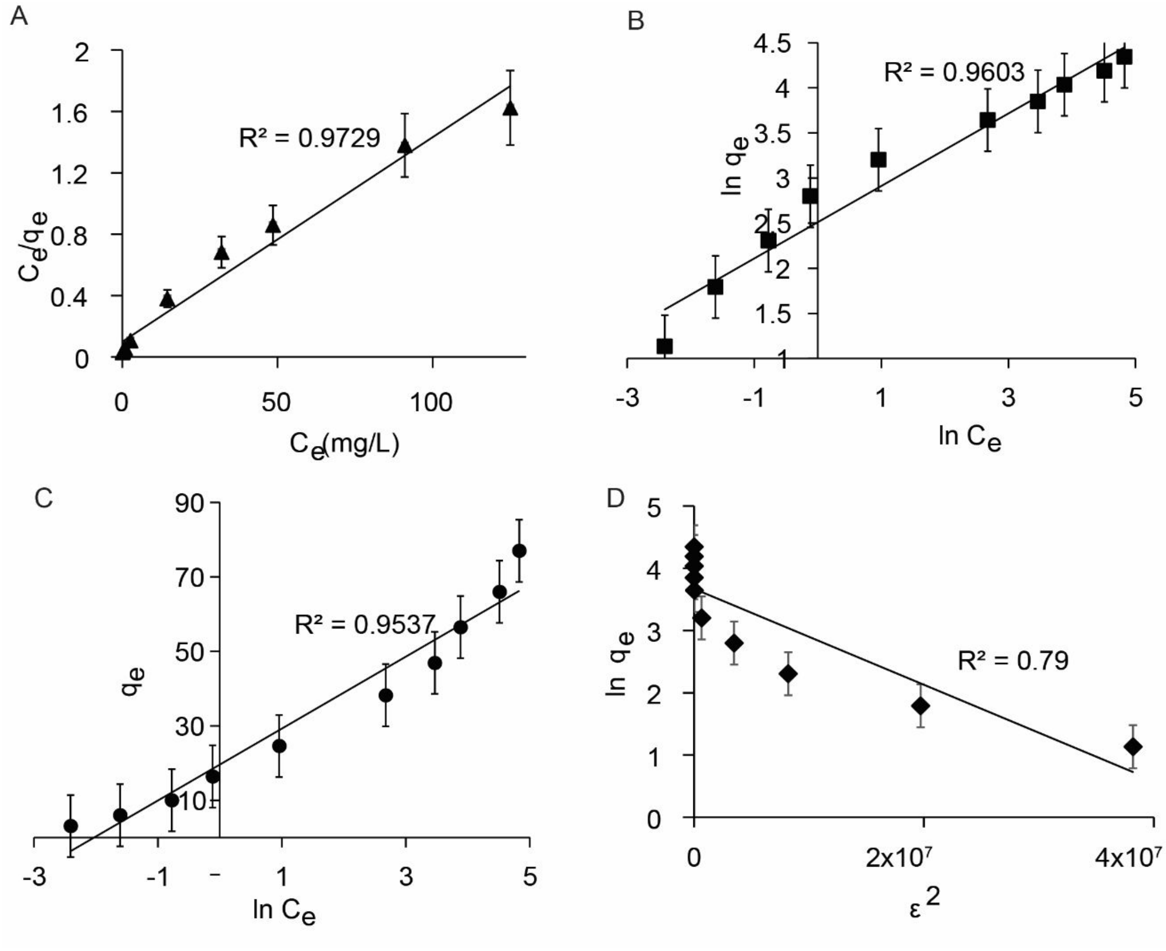


Figure S7. Adsorption isotherms of CAlFeC for fluoride showing Langmuir (A), Freundlich (B), Temkin (C) and Dubinin-Radushkevich (D) isotherm fittings.

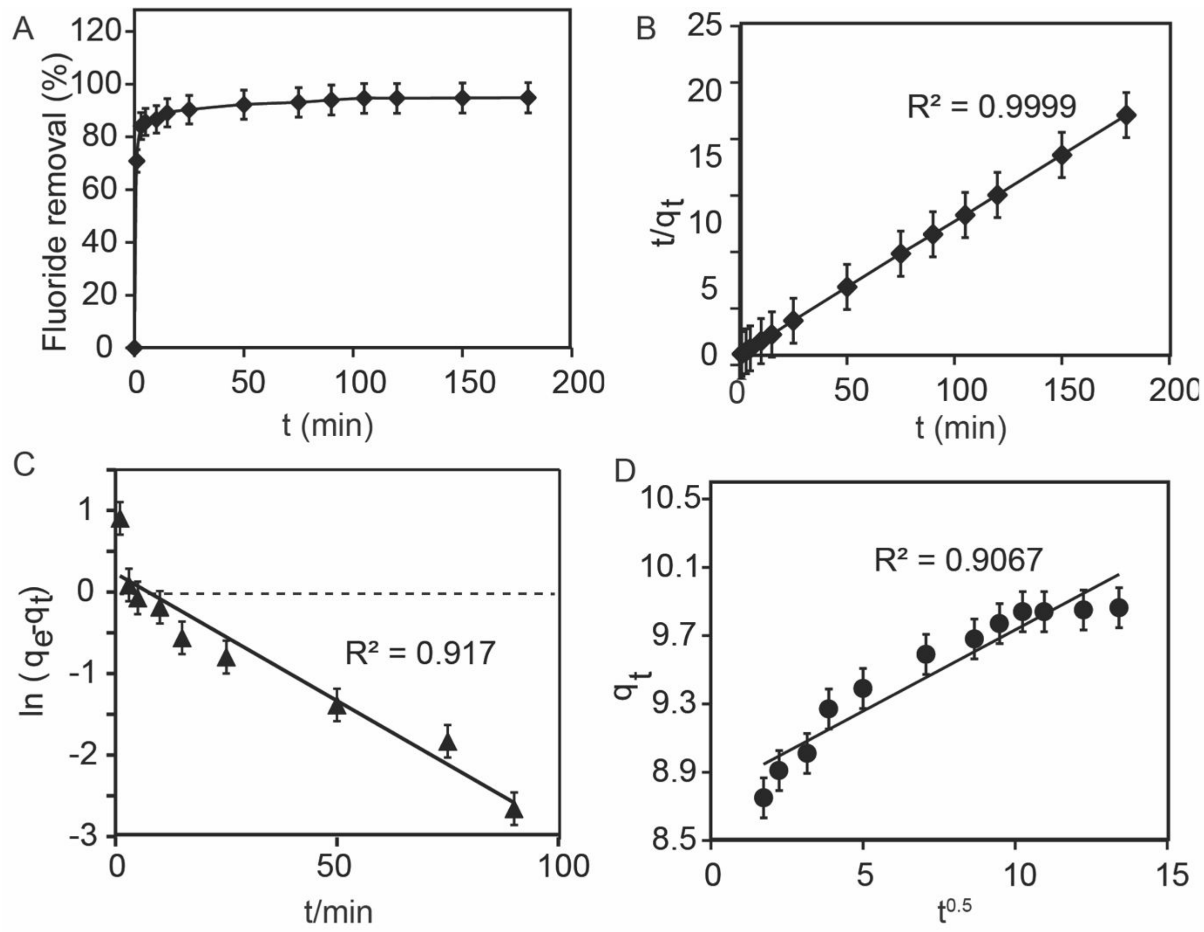


Figure S8. Adsorption kinetics data showing the effect of contact time (A) and adsorption kinetics models: pseudo-second order (B), pseudo-first order (C) and intraparticle diffusion (D).

EQUATIONS:

Equation S1. The maximum uptake of F^- (q_e) was calculated using the equation given by:

$$q_e = \frac{(C_0 - C_e)V}{w}$$

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 fluoride in the bulk solution (mg/L), C_0 is the initial fluoride concentration (mg/L), V is the volume of solution (L) and w is the mass of the adsorbent (g).

Equation S2. Percentage of fluoride removal:

$$\%\ Removal = \left(\frac{C_0 - C_e}{C_0}\right) 100\%$$

Equation S3. The linear form of Freundlich adsorption isotherm is given as

$$ln\ q_e = ln\ k + \frac{1}{n}\ ln\ C_e$$

where k is the adsorption capacity and n is the adsorption intensity

Equation S4. The linear form of Langmuir adsorption isotherm is given by equation

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

where 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 S5. Separation factor R_L, which is calculated from the following equation:

$$R_L = \frac{1}{1 + bC_o}$$

Equation S6. The Temkin isotherm model is represented in the form:

$$q_e = \frac{RT}{k} ln\, A + \left(\frac{RT}{b}\right) ln\, C_e$$

where *k, A* and *b* are Temkin constants.

Equation S7. Dubinin-Radushkevich isotherm model is given by the equation:

$$ln\, q_e = ln\, q_o - k\varepsilon^2$$

Equation S8. The parameter ε in Equation S7 is obtained from the formula:

$$\varepsilon = RT\, ln\left(1 + \frac{1}{C_e}\right)$$

where *R* and *T* are the gas constant and absolute temperature, respectively.

Equation S9. Lagergren pseudo-first order model is given by the equation below:

$$ln\, (q_e - q_t) = ln\, q_e - k_1 t$$

where k_1 is the rate constant and q_t is the adsorption capacity at any given time, *t*.

Equation S10. Ho & Mckay pseudo-second order model is given by the equation below:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e}$$

where k_2 is the pseudo-second order rate constant.

Equation S11. Weber-Morris intraparticle diffusion kinetic model

$$q_t = k_i \sqrt{t}$$

where k_i is the rate constant.

Equation S12. Mass intensity = $\frac{\text{Mass of all materials used excluding water}}{\text{Mass of product}}$ kg/kg product

Equation S13. Water intensity = $\frac{\text{Mass of all water used}}{\text{Mass of product}}$ kg/kg product

Equation S14. $$\text{Reaction mass efficiency} = \frac{\text{Mass of product}}{\text{Mass of all reactants}} \times 100\%$$

Equation S15. $$\text{Energy intensity} = \frac{\text{Amount of non renewable energy uses}}{\text{Mass of product}} \ \text{kW.h/kg}$$

Equation S16. $$\text{E factor} = \frac{[\text{kg (raw materials)} - \text{kg (desired product)}]}{\text{kg (total product including water)}}$$

TABLES:

Table S1. Characteristics of TW before and after spiking with F^-, and after contact with CAlFeC.

Property of TW	Before spiking with F^-	After spiking with F^-	After contact with CAlFeC
Electrical conductivity (µS/cm)	760.3	810	620.5
pH	7.2	8.1	6.7
Fluoride (mg/L)	0.03	10.4	0.6