***Supplementary data***

**Controlled removal of fluoride by ZIF-8, ZIF-67, and Ni-MOF of different morphologies**

**1. Response Formulas:**

The process efficiency in term of F- removal percentage (µ), adsorption capacity at any time(qt), and at the equilibrium (qe) were calculated by Eq.1 to Eq.3

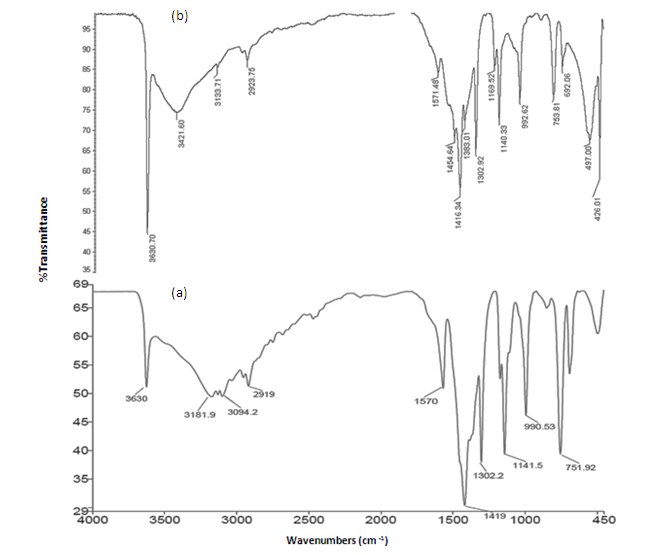
where, Ci, Ct, and Cf are initial F-1 concentration(mg/L), F-1 concentration at any time(mg/L), and under the equilibrium (mg/L), respectively. W and V are MOF mass(g), and solution volume(L), respectively.

|  |  |  |
| --- | --- | --- |
| ZIF-67-NO3 | ZIF-67-Cl | ZIF-67-SO4 |
| ZIF-8- Leaf | ZIF-8-Octahedron | ZIF-67-OAC |
| Ni-MOF | ZIF-8-Cube | ZIF-8-Cuboid |

**Fig. S1.** FE-SEM images of as-synthesized MOFs

**2.FTIR spectra of ZIF-67-NO3**

FTIR spectra of ZIF-67-NO3 before and after fluoride adsorption are shown in Fig S2. The representative ZIF-67 (Co(Hmim)2 ), Hmim (2-methylimidazole) consists of cobalt ion (Co2+) and 2-methylimidazolate anions. As are seen from Figure1a, the bands at about 3400 -3600 cm–1 are related to the O–H stretching vibration of the hydroxyl groups. The bands at about 600– 1500 cm–1 are attributed to the bending and stretching modes of the imidazole ring [1, 2]. The bands at 2400 and 3000 cm–1 are assigned to the vibration bending of the N–H in Hmim. The band at about 1570 cm–1 is attributed to the C=N stretching vibration [3-5]. Besides, the band observed at about 1420 cm–1 is related to the C=C bond in the imidazole ring[3, 6]. Furthermore, the band observed at about 429 cm–1 in the spectrum of based ZIF-67-NO3 is attributed to the Co–N bonds. Similarity of FT-IR pattern for ZIF-67-NO3 before and after adsorption of fluoride (Figure S1. (a,b)), demonstrate the adsorption of fluoride in the pores of adsorbent.



**Fig. S2.** FTIR spectra of the ZIF-67-NO3 before (a), and after (b) fluoride adsorption

**Table S1.** Statistical parameters estimated by the polynomial model for F- adsorption

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Source | Sum of Squares | df | Mean Square | F Value | p-value Prob > F |
| Model | 4774.93 | 14.00 | 341.07 | 51.40 | 0.00 |
| A-time | 774.41 | 1.00 | 774.41 | 116.72 | 0.00 |
| B-MOF | 188.02 | 1.00 | 188.02 | 28.34 | 0.00 |
| C-pH | 2557.92 | 1.00 | 2557.92 | 385.52 | 0.00 |
| D- F-1 | 157.69 | 1.00 | 157.69 | 23.77 | 0.00 |
| AB | 68.06 | 1.00 | 68.06 | 10.26 | 0.01 |
| AC | 103.02 | 1.00 | 103.02 | 15.53 | 0.00 |
| AD | 6.76 | 1.00 | 6.76 | 1.02 | 0.33 |
| BC | 10.89 | 1.00 | 10.89 | 1.64 | 0.22 |
| BD | 12.96 | 1.00 | 12.96 | 1.95 | 0.18 |
| CD | 93.12 | 1.00 | 93.12 | 14.04 | 0.00 |
| A2 | 233.84 | 1.00 | 233.84 | 35.24 | 0.00 |
| B2 | 86.02 | 1.00 | 86.02 | 12.97 | 0.00 |
| C2 | 636.28 | 1.00 | 636.28 | 95.90 | 0.00 |
| D2 | 171.48 | 1.00 | 171.48 | 25.85 | 0.00 |
| Residual | 92.89 | 14.00 | 6.63 |  |  |
| Lack of Fit | 76.29 | 10.00 | 7.63 | 1.84 | 0.29 |
| Pure Error | 16.60 | 4.00 | 4.15 |  |  |
| Cor Total | 4867.82 | 28.00 |  |  |  |
| R2 | 0.98 | Adeq Precision | | 27.14 |  |
| R2 adj | 0.96 | R2 pred | | 0.90 |  |

# Table S2. The models and statistical parameters used in this study [7, 8]

|  |  |  |
| --- | --- | --- |
| **Type of equation** | **Nonlinear form** | **Parameters** |
| **Isotherm models** |  |  |
| Langmuir |  | Ce= adsorbate equilibrium concentration (mg/L)  qe = adsorption capacity at equilibrium (mg/g)  Qm= monolayer coverage capacity (mg/g)  KL= Langmuir isotherm constant (L/mg). |
| Freundlich |  | Kf = Freundlich isotherm constant(mg1-(1/n) L1/n g-1)  n = adsorption intensity |
| Sips |  | = Sips maximum adsorption capacity (mg/g)  KS = Sips equilibrium constant  mS= Sips model exponent |
| Temkin |  | AT =Temkin isotherm equilibrium binding constant (L/g)  bT = Temkin isotherm constant  R= universal gas constant (8.314 |
| Redlich-Peterson |  | KRP = Redlich-Peterson isotherm constant(L/g)  aRP=Redlich-Peterson model constant(mg/L)-g  g = Redlich-Peterson model exponent |
| Khan |  | bK= is the Khan model constant  aK= Khan model exponent |
| Toth |  | KT= Toth equilibrium constant  TT= Toth model exponent |
| **Kinetic models** |  |  |
| Pseudo first-order |  | qe = adsorption capacity at equilibrium (mg/g)  qt = adsorption capacity at any time (mg/g)  = Rate constant min−1 |
| Pseudo second-order |  | = Rate constant (g/(mg min)) |
| Intraparticle diffusion |  | = Rate constant (mg/(g min)) |
| Elovich |  | qt = adsorption capacity at any time (mg/g)  = Rate constants |
| **Statistical parameter** |  |  |
| Coefficient of determination (R2) |  | =The predicted value by the model  yi=The observed value.  =Mean of observed value.  =The number of variables in the model |
| Adjusted R-square(R2adj) |  |
| Sum of squares error (SSE) |  |

**Table S3.** Parameters for fitting nonlinear isotherm models used for adsorption of F- [9]

|  |  |  |
| --- | --- | --- |
| **Values** | **Parameters** | **Isotherm** |
| 0.84 | b (L mg‒1) | Langmuir |
| 25.9 | Qmax (mg/g) |  |
| 0.85 |  |  |
| 0.81 | R2 |  |
| 36.4 | RSS |  |
| 15.6 | Kf (mg g‒1) (mg‒1)1/n | Freundlich |
| 6.9 | n |  |
| 0.99 |  |  |
| 0.99 | R2 |  |
| 2.1 | RSS |  |
| 112.6 | AT (L/mg) | Temkin |
| 3.1 | bT |  |
| - | B (J/mol) |  |
| 0.96 |  |  |
| 0.94 | R2 |  |
| 10.3 | RSS |  |
| 42.66 | qms (mg g‒1) | Sips |
| 0.48 | KS (L mg‒1)ms |  |
| 0.35 | ms |  |
| 0.96 |  |  |
| 0.98 | R2 |  |
| 5.65 | RSS |  |
| 14.34 | KT | Toth |
| 0.01 | AT |  |
| 1.22 | TT |  |
| 0.94 |  |  |
| 0.96 | R2 |  |
| 8.29 | RSS |  |
| 543.16 | kRP (L g‒1) | Redlich-Peterson |
| 37.44 | aRP (mg L‒1)‒bRP |  |
| 0.82 | bRP |  |
| 0.94 |  |  |
| 0.96 | R2 |  |
| 8.09 | RSS |  |
| 6.86 | qs (mg g‒1) | Khan |
| 57.95 | bK |  |
| 0.81 | aK |  |
| 0.94 |  |  |
| 0.96 | R2 |  |
| 8.29 | RSS |  |

**Table S4.** Parameters for fitting nonlinear kinetic models used for adsorption of F-

|  |  |  |  |
| --- | --- | --- | --- |
| Concentration (mg/L) | 10 |  |  |
| qe, exp (mg/g) | 12.57 |  |  |
| **Pseudo-first order** | | **Interparticle diffusion** | |
| qe (mg/g) | 12.57 | k3 | 1.62 |
| k1 (min-1) | 0.04 | C | -0.05 |
|  | 0.99 |  | 0.99 |
| R2 | 0.99 | R2 | 0.99 |
| RSS | 0.4 | RSS | 0.8 |
| **Pseudo-second order** | | **Elovich** | |
| qe (mg/g) | 16.75 | A | 1.6 |
| k2 (g/mg min) | 0.00 | B | 0.27 |
|  | 0.99 |  | 0.99 |
| R2 | 0.99 | R2 | 0.99 |
| RSS | 0.19 | RSS | 0.59 |

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