

Fluoride Removal from Water By Carbonaceous Materials: Review

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Abstract—This study depicts the improvement of carbonaceous adsorbents for removal of fluoride from drinking water. The synthesized adsorbent was categorized and fluoride removal from aqueous solution was studied by batch mode. The impacts of pH of the solution, initial fluoride concentration, and temperature effect of fluoride removal has been verified in point of interest. The optimum pH range for fluoride removal was 5-8 for CSC as against 6-7 for activated alumina. The mechanism of adsorbent may be depicted by ion exchange occurred on the active sites. Generally speaking, the outcomes achieved in this study demonstrate carbonaceous adsorbents have a capability of useful applications for fluoride removal from aqueous solutions.

Keywords—Carbonaceous; Adsorption; Langmuir; First order reaction; Fluoride removal

I. INTRODUCTION

Vicinity of fluoride in drinking water brings out significant enthusiasm because of its vast effect on physiological frameworks of living creatures. Fluoride enters the human body from any source is held somewhere around 35% and 50% and the rest of principally excreted in pee. Drinking water is the single biggest supporter to day by day fluoride consumption, which changes extraordinarily in the middle of areas and people. Contingent upon the fluoride level in the drinking water and day by day water utilization. Among the accessible techniques, adsorption procedure is generally viewed as the most proper, especially for little group water defluoridation. This is because of its adaptability and simplicity of outline, relative comfort of operation and expense adequacy if a good adsorbent is available. Thus, many endeavors has been growing new fluoride adsorbents with good act in modern years. Endeavors have been under taken to review on defluoridation process utilizing adsorption process in light of a wide cluster of minimal effort and specific adsorbents to water for utilization having fluoride in attractive or passable amounts recommended by different national and international benchmarks. The recovery and safe transfer of adsorbents have been additionally review on this part.

II. CARBONACEOUS ADSORBENTS

The analyst's majority utilized activated carbon as adsorbent for removal of an extensive variety of oceanic poisons because of its high surface area (500-1500 m²/g), profoundly created inside the micro-porosity, low cost, prepared accessibility and vicinity of a scope of functional groups. The activation of carbonaceous materials such as a creature, plant or mineral origin was completed by using chemical or gas activation process. The most ordinarily raw materials utilized for the preparation of activated carbon were woods, charcoal, nut shells, fruit pits, lignite, coal, peat, bone, lignin, petroleum residues, synthetic polymers and also used low cost materials like rice-husk, saw dust, coconut shell, tree bark, cotton waste and so on by subjecting them to a high temperature warming with or without chemical activation.

Christoffersen *et al.* (1991) was examined the recovery of bone char by surface coating with a solution containing calcium chloride and sodium di-hydrogen phosphate with pH adjustment to 3.0.

At the point when the solution was left in contact with the bone char for 24 hours, the fluoride concentration was lessened to 0.02 moles/l [1].

Arulanantham *et al.* (1992) have researched defluoridation capability of coconut shell carbon (CSC) and alum impregnated CSC from aqueous solution. The adsorption capacity of modified CSC was more when contrasted with commercial activated carbon however, not exactly the limit of activated alumina (5.5 mg/g). The ideal pH range for fluoride removal was 5-8 for CSC as against 6-7 for activated alumina. The recovery of CSC was done with an aluminium sulfate solution without bringing about obstructing issue and falls in capacity as observed amid recovery of activated alumina [2].

Bhargava and Killedar (1992) contemplated the defluoridation of water by utilizing Fishbone charcoal as a part of a moving media adsorption system. The fluoride removal occurred principally vicinity of calcium phosphate. They found that the proportion of achieving equilibrium sorbate concentration to the initial sorbate concentration and the fluoride removal capacity of the sorbent was found to shift contrarily with the sorbent mass input rate, and changed straightforwardly with the sorbate flow rate and initial sorbate concentration. The proportion of achieving equilibrium sorbate concentration to the initial sorbate concentration was observed to be a sorbate's function sorbent-sorbate mass input rate ratio [3].

Ramos *et al.* (1999) have researched the defluoridation capacity of aluminium impregnated carbon (AIAC) from aqueous solution. The aluminium nitrate solution at settled pH was blended with carbon took after by the calcination to 300°C. The adsorption capacity of AIAC was 3 to 5 times of plain activated carbon (AC) at an equilibrium fluoride concentration of 2-8 mg/l. Impregnated carbon was demonstrated to have a fluoride adsorption capacity of 3 to 5 times that of plain activated carbon. The Langmuir adsorption capacity of AIAC was observed to be 1.07 mg/g at neutral pH when contrasted to plain carbon (0.49 mg/g) [4].

Mckee and Johnston (1999) have utilized minimal effort materials such as rice husk, wheat husks for removal of fluoride from water. The rice husk has removed 83% of fluoride at pH under 3.0 with 6 g/l of dose and accomplished the equilibrium condition within 180 minutes [5].

Dahiya and Kaur (1999) have utilized coconut coir pith carbon (CPC) via carbonization with sulfuric acid and alum impregnated CPC for defluoridation of water. The alum impregnation with 1% and 2% on CPC expanded its defluoridation capacity by 1.5 to 2 times when contrasted with plain CPC. The fluoride removal of 78.8% from 2.5 mg/l of fluoride solution was found by 10 g/l of CPC impregnated with 2% alum. The defluoridation capacity of adsorbent was discovered diminished progressively with expansion in initial fluoride concentration and with diminished in a dose of adsorbent [6].

Sreenivasulu *et al.* (1999) have utilized defluoridation by activated carbon arranged by Umbles of Parangos Pavularia Lindl (PPC) in batch study. The adsorption was quick and finished inside of an hour and a half for a fluoride concentration of 2.8 – 7.6 mg/l. The fluoride removal of 85% was acquired by PPC at pH of 8.75 and a temperature of 330K for 7g/l of fluoride containing solution [7].

Sivasamy *et al.* (2001) have contemplated the defluoridation capacity of coal based adsorbent as a minimal effort adsorbent. The adsorption capacity of lignite, fine coke and bituminous coal were 7.09, 6.90 and 7.44 mg/g respectively. For lignite, the higher defluoridation was happened at pH range 6-12 though the higher defluoridation for fine coke and bituminous coal happens at acidic pH below 4. The fluoride removal efficiencies of three coal based adsorbent were somewhere around 77.0 and 85.0%. The outcomes information fitted well with Langmuir and Freundlich isotherms and

also first-order rate constant and film-pore diffusion coefficients. These coals based adsorbent had higher defluoridation capacities when contrasted to commercial activated carbon [8].

Abe *et al.* (2004) have considered the defluoridation capacity of different carbonaceous materials included six types of activated carbon (AC), carbon block (CB), four kinds of coal charcoals (CC) and bone char (BC) from water. The fluoride removal of different carbons was found in the order of BC (82%) > CC (0.4-3.5%) > CB (10%) > AC (5.4-17%). Among all, the most extreme fluoride removal was finished by BC at pH 4.6. The Freundlich coefficient (r) was 0.998 for BC and adsorption reaction was endothermic. The mechanism of adsorption was an ion exchange between fluoride and phosphate ions available in bone char [9].

Tembhurkar and Dongre (2006) have utilized commercial activated charcoal for defluoridation of water from aqueous solution. The ideal dose of adsorbent was 2.0 g/100 ml and the equilibrium was accomplished in 120 minutes and fluoride removal of 94 % was found at pH of 2.0. The outcome fitted well with Langmuir isotherm ($R^2=0.984$) than Freundlich isotherm ($R^2=0.9634$) [10].

Karthikeyan and Ilango (2007) have investigated the fluoride adsorption capacity of activated carbon by smoldering and carbonization of *Moringa indica* bark. The fluoride removal of 71% was found in the initial fluoride concentration of 2 mg/l at neutral pH. The experimental data took after the Freundlich and Langmuir isotherms and pseudo-second-order kinetic model and the reaction was endothermic in nature [11].

Karthikeyan and Elango (2008) have utilized graphite for defluoridation of aqueous solution. The experimental data fitted well to Freundlich and Langmuir isotherms and took after reversible first order kinetic and the adsorption process was endothermic in nature and physical process according to thermodynamic study. The Lower range of pH and high temperature ranges were found good for ideal fluoride removal. The adsorption capacity of three commercial graphite was 3.13, 0.25 and 0.16 mg/g for G1, G2 and G3 respectively and the higher capacity of G1 was because of higher carbon content and lower particle size [12].

Emmanuel *et al.* (2008) have utilized commercial activated carbon (CAC) and indigenously prepared activated carbons from *Pithecellobium dulce* (PLDC), *Ipomoea batatas* (IBC) and *Peltophorum ferrugineum* (PFC) for removal of fluoride from drinking water. The author was found that the fluoride removal expanded with diminished in the initial fluoride concentration and particle size of adsorbent furthermore expanded with expansion in contact time, amount of adsorbent used and the initial pH of the solution. The experimental data has fitted well with Langmuir and Freundlich isotherm models and in addition first order kinetics. The removal of fluoride was found to be in the order of PLDC (78.96) > IBC (76.62) > PFC (74.48) > CAC (39.20) [13].

Gupta *et al.* (2009) have explored fluoride removal capacity of micronanohierarchical web comprising of activated carbon fibres (ACF) and carbon nanofibers (CNF) impregnated with Al from wastewater and its adsorption capacity of 17 mg/g was observed for fluoride concentration of 0.1- 50 mg/l. The Al-CNF was dealt wastewater without earlier treatment at a pH range of 5.0-8.0 [14].

Pekar (2009) has utilized natural lignite from the South Moravia region of the Czech Republic for defluoridation of water. Binding of fluoride on lignite was tight and just minor part (13%) drained into water. The removal of no less than 90% was accomplished by lignite within 10 minutes. The removal of small concentration of fluoride was additionally done effectively lignite as an adsorbent. The outcomes were best fitted with Langmuir and Freundlich isotherm models. Foreign ions and pH demonstrated minor impact on fluoride removal capacity [15].

Khan and Ansari (2009) have prepared activated charcoal from any carbonaceous materials by physical or chemical activation. The suitable chemical impregnated activated charcoals have more adsorption capacity because of adjustment of surface area or pore structure of charcoal. The adsorption was significantly influenced by the pH of the solution and presence of foreign ions [16].

Kishore and Hanumanthrao (2010) have utilized Acacia Arabica fruit carbon as compelling adsorbent for defluoridation of water. The variable impacts on adsorption process for the chose area was observed in the order of granulometry (G) < agitation time (T) < adsorbent's concentration (A). The significance level of 5% for pH and 1% for different parameters was found within the chose area. The Acacia Arabica was less expensive than Activated carbon and found suitable for defluoridation [17].

Tchomgui-Kamga *et al.* (2010) have utilized calcium chloride impregnated wood charcoal took after via carbonization at 500 °C, 650 °C or 900 °C as adsorbent for defluoridation of water by batch experiment condition. The adsorption capacity of adsorbent was found in the order of Ca650/C (19.05 mg/g) > Ca500/C (17.09 mg/g) > Ca900/C (15.22 mg/g). The fluoride removal of more than 92% was accomplished within 24h at neutral pH for fluoride solution of 10 mg/l. The experimental data has fitted well with Langmuir and Freundlich isotherm models and in addition pseudo-second-order kinetic model. The fluoride removal was not influenced by the vicinity of sulphate and phosphate ions, while bicarbonates and chloride ions somewhat influenced the defluoridation capacity. The charcoal was chemically steady in the solution and only 3.23 mg/l of Ca was disintegrated at neutral pH. Additionally the amount of dissolved Al and Fe was observed to be 0.67 and 1.8 mg/l respectively at neutral pH and the scattering of calcium oxide or calcium carbonate into the carbon framework gave more fluoride removal effectiveness than calcium compounds [18].

Ansari *et al.* (2011) have utilized multi-walled carbon nanotubes (MWCNTs) for defluoridation of drinking water in the experiment. The greatest fluoride removal of around 94% was observed at pH 5 and 18 minutes of contact time, though 85% removal was observed in Kohbanan city drinking water. The MWCNTs have a saturation capacity of 3.5 mg/g and the experimental outcomes best fitted with Freundlich isotherms [19].

Li *et al.* (2011) examined the defluoridation capacity of grapheme from aqueous solution in batch mode condition. During the experiment, the adsorption capacity of 17.65 mg/g was observed with 25 mg/l of initial fluoride concentration at 25°C. The experimental data has fitted well with the Langmuir isotherm model. The adsorption reaction was spontaneous and endothermic in nature [20].

Rao *et al.* (2011) have examined defluoridation capability of three activated carbon prepared such as BKC, BOC and RSC prepared from biomaterials of bergera koenigh (curry leaf seeds), Batavia orange and raphanus sativus (garden radish) respectively. The ideal condition for maximum fluoride removal was accomplished by keeping up a pH of 6.0, 30 minutes of contact time and 1 g/l of optimum dose of 5 mg/l of initial fluoride concentration. The fluoride removal by adsorbents established altogether of BOC > RSC > BKC and the regeneration of spent adsorbents was done by passing 0.1M NaOH solution [21].

Cui *et al.* (2012) have utilized a novel poly (aniline-co-o-aminophenol) (PAOA) adjusted carbon felt electrode reactor in a continuous flow reactor for defluoridation of aqueous solution. The ideal defluoridation efficiency was observed at around 1.2 V and the leap forward capacities were > 10 mg/g over a wide pH range (5-9) with an initial fluoride concentration of 10 mg/l [22].

Mendoza *et al.* (2012) have utilized carbonaceous materials from pyrolysis of sewage sludge for defluoridation of aqueous solution in the experimental condition. The maximum adsorption capacity

of 2.84 ± 0.03 mg/g was observed with 0.4 g/l and with 20 g/l of carbonaceous material, $82.2 \pm 0.5\%$ of fluoride was observed to be removed. The equilibrium was come to following 18 hours of contact time and maximum adsorption was found at pH of 7.06 ± 0.08 . The outcomes took after the Langmuir-Freundlich isotherm, pseudo-second-order and intra-particle mass transfer diffusion models. The adsorption mechanism was essentially administered by chemisorption on a heterogeneous material and desorption of fluoride and recovery of spent adsorbent was completed by a solution of bicarbonate and HCl (pH=1) [23].

Chen *et al.* (2013) have utilized porous composite in view of aluminium sulphate and grapheme hydrogel (BAS@GHG) were prepared through homogeneous precipitation of BAS in GHG and utilized for defluoridation. The adsorption capacity of 33.4 mg/g was acquired at an equilibrium fluoride concentration of 10.7 mg/l and the temperature of 298 K. The experimental data has fitted well with pseudo first order kinetics, the Weber-Morris model and Langmuir equation [24].

Atasoy *et al.* (2013) have utilized commercial activated carbon for defluoridation of water. The maximum fluoride adsorption capacity of 0.50 mg/g and 50% removal of fluoride were accomplished with initial fluoride concentration of 10 mg/l at 25°C. The experimental data has fitted well with Freundlich isotherm (R=0.96) than Langmuir isotherm(R=0.60) [25].

Yu *et al.* (2014) examined the defluoridation capacity of lanthanum altered carbon (LMC) got from Sargassum sp. The ideal pH for maximum fluoride removal was observed to be 3 to 9. The 90% of fluoride removal was observed within 1 hour and equilibrium was come to within 4 hours. The maximum adsorption capacity of LMC was 94.34 mg/g at neutral pH. The adsorption process followed Langmuir isotherm and pseudo-second-order-kinetic model and the fluoride removal was somewhat get influenced by the vicinity of phosphate, bicarbonate and sulfate ions, while unaffected by the vicinity of humic acid [26].

Getachew *et al.* (2015) utilized thermally activated banana peel and coffee husk as bio adsorbents for defluoridation of water. The maximum fluoride removal by banana peel and coffee husk were 85 and 86% respectively for fluoride solution of 10 mg/l. The ideal pH for both the adsorbent were seen to be 2.0. The fluoride removal proficiency of coffee husk was observed more than the banana peel. The adsorption process was fitted well with Langmuir isotherm and pseudo-second-order kinetic model [27].

Brunson *et al.* (2015) was utilized eucalyptus wood char for defluoridation of water. The fluoride adsorption capacity of wood char was more at burning temperature of 500 and 600 °C than bone char. The aluminum oxides changed eucalyptus wood char was expanded adsorption capacity from 0.11 to 0.72 mg/g for initial fluoride concentration of 10 mg/l [28].

Guan *et al.* (2015) examined the altered Mongolian scotch pine tree saw dust char (MMSC) made by utilizing phosphoric acid – microwave method for removal of fluoride from drinking water. The fluoride removal of 82, 71 and 63% were observed for fluoride solution of 5, 10 and 20 mg/l respectively. The optimum dosage of MMSC for successful removal of fluoride was found to be 3.6 g/100mL. The maximum adsorption capacity of MMSC was achieved to be 0.885 mg/g in high fluoride asset water. The adsorption reaction observed to be spontaneous an endothermic in nature [29].

Emmanuel *et al.* (2015) utilized Pithacelobiumdulce carbon (PLDC) as a minimal effort adsorbent for defluoridation of aqueous solution and compared it with commercial activated carbon (CAC). The fluoride removal by PLDC and CAC was 81.60% and 40.20% respectively from aqueous solution. The Langmuir adsorption capacity of PLDC and CAC was 0.07 and 0.11 mg/g respectively

at neutral pH. The optimum condition was accomplished at 3g/l dosage of adsorbent, neutral pH, 40 minutes of contact time and 5 ppm of fluoride solution for both the adsorbents. The experimental data has fitted well with Langmuir isotherm and first order kinetic reaction [30].

Ajisha and Rajagopal (2015) utilized pyrolyzed Delonix regia pod carbon for defluoridation of water. The maximum fluoride removal was found at optimum conditions of 300 minutes of contact time, pH of 2.0 and dosage of 1.5 g/l. The value of Q_0 according to Langmuir isotherm were ranged from 18.52 to 142.86 for different temperature ranged from 303K to 333K respectively. The adsorption process followed Freundlich isotherm and pseudo-second-order kinetic and in addition intraparticle diffusion models. The adsorption reaction was spontaneous, irreversible, steady and endothermic in nature [31].

III. CONCLUSION

The ground water in a couple of countries like India gets dirtied by fluoride and responsible for fluorosis [32-37]. This paper depicts the audit of diverse carbonaceous adsorbents in lieu of excessive business adsorbents for defluoridation of water. The fluoride uptake of particular adsorbent depends on upon test conditions, for instance, pH of aqueous solution, dose of adsorbent, temperature, initial fluoride concentration, agitation speed, contact time, etc. The evacuation limit of carbonaceous adsorbent observed to be increments with the adsorbent's dosage and littler size of adsorbent. The equilibrium data fitted well with Langmuir or Freundlich isotherms and in addition adsorption procedure took after pseudo-first-order or pseudo-second-order dynamic models. A review of carbonaceous adsorbents has wonderful potential for defluoridation of water. The carbonaceous adsorbents are calm and fruitful for fluoride departure and can be exchanged the commercial adsorbents. The future exploration ought to be centered around the assessment of fluoride uptake in term of expense and field suitability to check the section execution to acquire the drinking water with fluoride arrangement under 1.5 mg/l (World Health Organization limit for drinking water).

REFERENCES

- [1] J. Christoffersen, M.R. Christoffersen and R. Larsen, "Regeneration of surface coating of bone char used for defluoridation of water", *Water Res.*, vol. 25, pp. 227-229, 1991.
- [2] A. Arulanantham, T.V. Ramcrishna and N. Balsubramaian, "Studies on fluoride removal by coconut shell carbon", *J. Environ. Protect.*, vol. 12, pp. 531-532, 1992.
- [3] D.S. Bhargava and D.J. Killedar, "Fluoride adsorption on fishbone charcoal through a moving media adsorber", *Water Res.*, vol. 26, pp. 781-788, 1992.
- [4] R.L. Ramos, J. Ovalle-Turrubiartes and M.A. Sanchez-Castillo, "Adsorption of fluoride from aqueous solution on aluminium-impregnated carbon", *Carbon*, vol. 37, pp. 609-617, 1999.
- [5] R. Mckee and W.S. Johnston, "Removal of fluorides from drinking water using low-cost adsorbent", *Ind. J. Environ. Health*, vol. 41, pp. 53-58, 1999.
- [6] S. Dahiya and A. Kaur, "Studies on removal of fluoride by coconut coir pit carbon", *Ind. J. Environ. Protect.*, vol. 19, pp. 811-814, 1999.
- [7] A. Sreenivasulu, E.V. Sumdaram, M.K. Reddy, "Defluoridation studies using activated carbon prepared from umbles of parangos pavularia Lindl", *J. Ind. Pollut. Contr.*, vol. 15, pp. 24-27, 1999.
- [8] A. Sivasamy, K.P. Singh, D. Mohan and M. Maruthamuthu, "Studies on defluoridation of water by coal based adsorbents", *J. Chem. Technol. Biotechnol.*, vol. 76, pp. 717-722, 2001.
- [9] I.K. Abe, S. Iwasaki, T. Tokimoto, N. Kawasaki, T. Nakamura and S. Tanada, "Adsorption of fluoride ions onto carbonaceous materials", *J. Colloid Interf. Sci.*, vol. 275, pp. 35-39, 2004.
- [10] A.R. Tembhurkar and S. Dongre, "Studie on fluoride removal using adsorption process", *J. Environ. Sci. Eng.*, vol. 48, pp. 151-156, 2006.
- [11] G. Karthikeyan and S.S. Ilango, "Fluoride sorption using Moringa Indica-based activated carbon", *Iran. J. Environ. Health Sci. Eng.*, vol. 4, pp. 21-28, 2007.
- [12] M. Karthikeyan and K.P. Elango, "Removal of fluoride from aqueous solution using graphite: A kinetic and thermodynamic study", *Indian J. Chem. Technol.*, vol. 15, pp. 525-532, 2008.
- [13] K.A. Emmanuel, K.A. Ramaraju, G. Rambabu and A. V. Rao, "Removal of fluoride from drinking water with activated carbons prepared from HNO₃ activation—A comparative study", *Rasayan J. Chem.*, vol. 1, pp. 802-818, 2008.

- [14] A.K. Gupta, D. Deva, A. Sharma and N. Verma, “Adsorptive removal of fluoride by micro-nanohierarchical web of activated carbon fibers”, *Ind. Eng. Chem. Res.*, vol. 48, pp. 9697–9707, 2009.
- [15] M. Pekar, Fluoride anion binding by natural lignite (South Moravian Deposit of Vienna Basin)”, *Water Air Soil Pollut.*, vol. 197, pp.303–312, 2009.
- [16] A.M. Khan and R. Ansari, “Activated charcoal: Preparation, characterization and applications: A review article”, *Int. J. Chem. Tech. Res.*, vol.1, 859–864, 2009.
- [17] M. Kishore and Y. Hanumantharao, “Validation of defluoridation method with “Acacia Arabica” plant by product through 2n factorial experimentation-A statistical approach”, *Int. J. Appl. Biol. Pharma. Technol.*, vol. 1, pp.1230–1235, 2010.
- [18] E.T. Kamga, E. Ngameni and A. Darchen, “Evaluation of removal efficiency of fluoride from aqueous solution using new charcoals that contain calcium compounds”, *J. Colloid Interf. Sci.*, vol. 346, pp. 494–499, 2010.
- [19] M. Ansari, M. Kazempour, M. Dehghani and M. Kazempour, “The defluoridation of drinking water using multi-walled carbon nanotubes”, *J. Fluorine Chem.*, vol.132, pp. 516–520, 2011.
- [20] Y. Li, P. Zhang, Q. Du, X. Peng, T. Liu, Z. Wang, Y. Xi, W. Zhang, K. Wang, H. Zhu and D. Wu, “Adsorption of fluoride from aqueous solution by Grapheme”, *J. Colloid Interface Sci.*, vol. 363, pp.348–354, 2011.
- [21] V.S. Rao, C. Chakrapani, C.S. Babu, K.S. Rao, M.N. Rao and D. Sinha, “Studies on sorption of fluoride by prepared activated Kaza’s carbons”, *Der. Pharma. Chemica.*, vol. 3, pp.73–83, 2011.
- [22] H. Cui, Y. Qian, H. An, C. Sun, J. Zhai and Q. Li, “Electrochemical removal of fluoride from water by PAOA modified carbon felt electrodes in a continuous flow reactor”, *Water Res.*, vol.46, pp. 3943–3950, 2012.
- [23] S.M. Mendoza, J.M. Reyes, M.S. Rios and E.G. Segura, “Fluoride removal from aqueous solutions by a carbonaceous material from pyrolysis of sewage sludge”, *Water Air Soil Pollut.*, vol. 223, Issue 5, pp. 1959–1971, 2012.
- [24] Y. Chen, Q. Zhang, L. Chen, H. Bai and L. Li, “Basic Aluminum Sulfate@Graphene Hydrogel Composites: Preparation and Application for Removal of Fluoride”, *J. Mater. Chem. A*, vol. 1, pp. 13101-13110, 2013.
- [25] A.D. Atasoy, M.I. Yesilnacar, and A.Z. Celik, “High Fluoride Level in Ground Water and Fluoride Adsorption on Activated Carbon”, *Digital Proceeding of THE ICOEST*, pp. 377-382, 2013.
- [26] Y. Yu, C. Wang, X. Guo, J.P. Chen., “Modification of carbon derived from Sargassum sp. by Lanthanum for enhanced adsorption of fluoride”, *J. Colloid Interf. Sci.*, vol. 441, pp. 113-120, 2015.
- [27] T. Getachew, A. Hussen and V.M. Rao, ““Defluoridation of water by activated carbon prepared from banana (*Musa paradisiaca*) peel and coffee (*Coffea arabica*) husk””, *Int. J. Environ. Sci. Technol.*, vol. 12, pp.1857–1866, 2015.
- [28] L.R. Brunson, D.A. Sabatini and M. Asce, “Role of Surface Area and Surface Chemistry during an Investigation of Eucalyptus Wood Char for Fluoride Adsorption from Drinking Water”, *J. Environ. Eng.*, vol. 141, pp. 1-8, 2015.
- [29] X. Guan, J. Zhou, N. ma, X. Chen, J. Gao and R. Zhang, “Studies on modified conditions of biochar and the mechanism for fluoride removal”, *Desalination Water Treat.*, vol. 55, pp. 440–447, 2015.
- [30] K.A. Emmanuel, A. Veerabhadrarao, T.V. Nagalakshmi, M.G. Reddy, P.P. Diwakar and C. Sureshbabu, “Factors influencing the removal of fluoride from aqueous solution by *Pithacelobium dulce* Carbon”, *Der Pharma Chemica*, vol. 7, pp. 225-236, 2015.
- [31] M.A.T. Ajisha and K. Rajagopal, “Fluoride removal study using pyrolyzed *Delonix regia* pod, an unconventional adsorbent”, *Int. J. Environ. Sci. Technol.*, vol. 12, pp.223–236, 2015.
- [32] S. S. Waghmare, T. Arfin, N. Manwar, D. H. Lataye, N. Labhsetwar and S. Rayalu, “Preparation and Characterization of *Polyalthia longifolia* Based Alumina as a Novel Adsorbent for Removing Fluoride from Drinking Water”, *Asian J. Adv. Basic Sci.*, vol. 4(1), pp. 12-24, 2015.
- [33] D. Thakre, P. Dixit, S. Waghmare, N. Manwar, N. Labhsetwar and S. S. Rayalu, “Synthesis Optimization and Fluoride Uptake Properties of High Capacity Composite Adsorbent for Defluoridation of Drinking Water”, *Environ. Progr. & Sust. Ener.*, pp. 1-10, 2015.
- [34] S. S. Waghmare and T. Arfin, “Fluoride Removal from Water By Calcium Materials: A State-Of-The-Art Review”, *Int. J. Innov. Res. Sci., Eng. Technol.*, vol. 4(9), pp. 8090-8102, 2015.
- [35] S. S. Waghmare and T. Arfin, “Fluoride Removal From Water By Mixed Metal Oxide Adsorbent Materials: A State-Of-The-Art Review”, *Int. J. Eng. Sci. Res. Technol.*, vol. 4(9), pp. 519-536, 2015.
- [36] S. S. Waghmare and T. Arfin, “Fluoride induced water pollution issue and its health efficacy in India- A review”, *Int. J. Eng. Res. Gen. Sci.*, vol. 3(5), pp. 345-358, 2015.
- [37] S. S. Waghmare and T. Arfin, “Fluoride Removal from Water by various techniques: Review”, *Int. J. Innov. Sci. Eng. Technol.* vol. 2(9), pp. 560-571, 2015.

