POLYACRYLIC ACID - ACRYLAMIDE BASED WATER PURIFICATION SYSTEMS FOR THE REMOVAL OF SELECTED ANIONS AND CATIONS FROM AQUEOUS MEDIA

By

SARNIS KANKANAMGE THILINI THATHSARA

M.Phil 2018

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Thesis submitted to the University of Sri Jayewardenepura for the award of the Degree of Master of Philosophy

Declaration

The work described in this thesis was carried out by me under the supervision of Dr. Thilini D. Gunasekara, Dr. Asitha Cooray, and Dr. Dilru R. Ratnaweera and this has not been submitted in whole or in part to any university or any other institution for another Degree/Diploma

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S. K Thlini Thathsara
2018

Certification of the supervisor

We certify that the above statement made by the candidate is true and that this thesis is suitable for submission to the University for the purpose of evaluation.

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Abbreviation

1/n	Heterogeneity factor
AAS	Atomic Adsorption Spectroscopy
C	Intercept of the plot of intra-particle diffusion model (mg/g)
C_e	Equilibrium concentration of the adsorbate (mg/L)
C_{liquid}	Liquid phase concentration (mg/L)
CMC	Carboxy Methyl Cellulose
C_0	Initial concentration (mg/L)
C_{solid}	Solid phase concentration at equilibrium (mg/L)
DETA	Diethylenetriammine
DSC	Differential Scanning Colorimetry
FTIR	Fourier transfer infrared spectroscopy
%In	Percentage of interference
k_1	Rate constant for pseudo first order adsorption (min^{-1})
k_2	Rate constant for pseudo first order adsorption (g/mg min)
K_F	Freundlich constant (mg/g)
k_{id}	Rate constant for intra-particle diffusion model ($mg\ g^{-1}\ min^{-1/2}$)
KPS	Potassium persulphate

K_L	Langmuir constant
m	Weight of the adsorbent (g)
NBIS	N,N'-methylene-bis-acrylamide
PAA	Polyacrylic acid
PAM	Polyacryalamide
pH_{pzc}	Zeta-Potential
PXRD	Power X-ray diffraction
q_e	Adsorption capacity at equilibrium (mg/g)
q_{max}	Maximum adsorption capacity (mg/g)
qt	Adsorption capacity at time t (mg/g)
q_{wo}	adsorption capacity of fluoride without interfering ions
q_w	adsorption capacity of fluoride with interfering ions
R	Universal gas constant (J mol^{-1} K^{-1})
R_L	Dimensionless adsorption intensity
T	Absolute temperature (K)
t	Time (min)
ΔG_o	Standard Gibbs free energy change
ΔH_o	Standard enthalpy change
ΔS_o	Standard entropy change
SEM	Scanning Electron Microscopy
SEM-EDS	SEM-Energy Dispersive Spectroscopy

TCIP	Trimetallic composite incorporated polyacrylamide
TEMED	Tetraethyl methylenediammine
TGA	Thermal Gravimetric Analysis
V	Volume
XRF	X-Ray Fluorescence

Acknowledgment

I would first like to thank my supervisors Dr. Thilini D. Gunasekara, Dr. Dilru R. Ratnaweera and Dr. Asitha Cooray of the Department of Chemistry, Faculty of Applied Sciences, University of Sri Jayewardenepura with great respect and honor for the guidance and encouragement throughout my research project. The door to Dr. Thilini D. Gunasekara, Dr. Dilru R. Ratnaweera and Dr. Asitha Cooray were always open whenever I ran into a trouble spot or had a question about my research or writing. .

I would also like to extend my special thanks to the University Research Grants (ASP/01/RE/SCI/2015/31, ASP/06/RE/SCI/2014/06 and ASP/01/RE/SCI/2015/29 for financial support, Advanced Material Research Center, Instrument Centre at the Faculty of Applied Sciences, University of Sri Jayewardenepura have facilitated the instrumental analysis for this study and Department of Chemistry, University of Sri Jayewardenepura for providing required facilities to carry out my research successfully.

Finally, I must express my very profound gratitude to my parents to my loving husband and my friends (specially our research assistant family) for providing me with unfailing support and continuous encouragement throughout my years of study and through the process of researching and writing this thesis. This accomplishment would not have been possible without them.

Thank you.

S. K Thilini Thathsara

ABSTRACT

POLYACRYLIC ACID-ACRYLAMIDE BASED WATER PURIFICATION SYSTEMS FOR REMOVAL OF SELECTED ANIONS AND CATIONS FROM AQUEOUS MEDIA

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Access to safe drinking water has declined over the last decades in almost every part of the world. High consumption rates of metals and chemicals in industrial processes have resulted in generation of large quantities of effluents that contain high level of toxic heavy metals. In addition to heavy metals, high concentration of fluorides could also impart toxicity towards humans and animals.

In this endeavor, water purification systems were developed using modified polymeric hydrogels under two approaches. The first approach was to fabricate polyacrylamide (PAM)/metal composite for selective removal of fluoride ions, from aqueous media and the second approach was to modify acrylic acid (AA)/acrylamide (AM) hydrogel using strong metal complexing ligands with the expectation of removing heavy metals from water. Diethylenetriamine (DETA) are the identified ligand in this regards.

Developed novel tri-metal composite of Fe-La-Ce has shown a significant binding efficiency towards fluoride ions in the presence of the other competing anions. The removal efficiency of fluorides was 161.29 mg/g at 28 ^{o}C and pH 7.00. The amount of fluoride adsorbed onto adsorbate is highly pH dependent and the adsorption capacity of 303.03 mg/g was achieved around pH 4.

Further, fabricate Fe-La-Ce tri-metallic composite incorporated polyacrylamide hybrid material was used to remove arsenate and chromate from aqueous media. A novel tri-metal composite incorporated polyacrylamide (TCIP) has shown a significant binding efficiency towards arsenate, chromate and fluoride ions in the presence of the other competing anions. The maximum adsorption capacities (q_{max}) of 43.85, 42.25 and 107.52 mg/g were achieved for arsenate, chromate and fluoride respectively at 300 K and in pH 7. Arsenate, chromate and fluoride adsorbed onto adsorbate is highly pH dependent.

The second approach is to focus more on removing cationic form of the heavy metals from aqueous media. In the second approach, that have been used the crosslinker with urea, that's the novelty of this endeavor. At the second stage, PAA-co-PAM cross-linked DETA modified urea was fabricated. The effect of contact time on Co(II), Ni(II), Mn(II) and Zn(II) onto the PAA-co-PAM-DETA-Urea was established. The adsorption capacity of Ni(II) and Mn(II) were rapidly increased within initial 120 minutes. The adsorption capacity of Mn(II) reached equilibrium state within 60 minutes while Co(II) took 180 min to reach the equilibrium. The adsorption process of Mn(II), Ni(II), Co(II) and Zn(II) onto the PAA-co-PAM-DETA-Urea was highly depend on the pH of the medium. The calculated results clearly indicated that developed PAA-co-PAM-DETA-Urea has ability to regenerate and reuse. Therefore it is great advantage to use PAA-co-PAM-DETA-Urea for removal of Mn(II), Ni(II), Co(II) and Zn(II) from aqueous media.

Keywords: Isotherms, Kinetics, Fluoride, Arsenic, Chromate, Polyacrylamide, Polyacrylic acid, Diethylenetriamine.