A RELIABLE POTABLE WATER PURIFICATION SYSTEM USING NANOMATERIAL-INCORPORATED MATRIX FOR HOUSEHOLDS IN CKDu PREVALENT AREAS

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DECLARATION

I declare that this is my own work, and this thesis does not incorporate without acknowledgement any material previously submitted for a Degree or Diploma in any other university or institute of higher learning and to the best of my knowledge and belief, it does not contain any material previously published or written by another person except where the acknowledgement is made in the text.

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The above candidate has carried out research for PhD thesis under my supervision.

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ABSTRACT

Chronic kidney disease of unknown aetiology (CKDu) in Sri Lanka is a national concerning health hazard as those affected face high mortality rates per year. One hypothesis on the disease pathogenesis is long-term exposure to fluoride, hardness, and cadmium in drinking water and their synergic effects, which causes nephrotoxic health hazards. Removal of fluoride, hardness, and cadmium is paramount in providing safe drinking water to the community in CKDu areas. However, available water treatment technologies in such areas do not offer an appropriate solution to drinking water issues. Hence, there are prerequisite to developing a reliable water purification unit to provide safe drinking water. This study investigated the best combination of materials to remove fluoride, hardness, cadmium, and faecal coliform in water to develop a reliable water purification unit to protect the community health and enhance their well-being.

Firstly, nephrotoxic risk factors in drinking water, their threshold levels, and the level of components required to remove complying with the required drinking water guideline values were evaluated. Water samples collected reported hardness in the range of 111.73 ± 1.41 – 680.33 ± 1.53 mg/L as $CaCO_3$ and fluoride 0.72 ± 0.03 mg/L and 2.84 ± 0.05 mg/L. The cadmium concentrations reported below the detection limit of 0.025 mg/L. Literature reported that fluoride (0.1–13.7 mg/L) and hardness (63.6–1921.0 mg/L) concentrations in water are very high. Fluoride concentrations in most CKDu prevalent areas exceed the drinking water guideline value (1.5 mg/L). The World Health Organisation does not declare a health concern permissible value to hardness in water. The cadmium level was reported in trace level in potable water less than the permissible drinking water guideline value (0.003 mg/L). Nephrotoxic drinking water guideline values should be declared for CKDu prevalent areas to control the spreading of nephrotoxic health hazards. In non-CKDu prevalent areas, potable water hardness values were often reported below the level of 120.0 mg/L and fluoride around 0.2 mg/L. Hence, potable water consumption with a fluoride level of around 0.2 mg/L, hardness 120.0 mg/L and cadmium 0.003 mg/L will control the occurrence of CKDu.

Available water treatment technologies introduced in CKDu prevalent areas were evaluated to identify their effectiveness in removing fluoride, hardness, and cadmium. Reverse osmosis, two-layer and seven-layer filter units have been introduced, treating potable water as a short-term

solution for the disease. The reverse osmosis unit removes most of the ions in water, retaining beneficial ions less in hardness 4.0–20.0 mg/L, high in fluoride 0.29–5.5 mg/L for human intake. The other two filters (two-layer and seven-layer filter units) do not remove fluoride and hardness effectively and add more ions into treated water due to the leachability in some minerals in the media. Treated water does not meet the required drinking water guideline values, highlighting a new requirement for water treatment units.

The risk assessment for RO treated water was conducted to identify non-carcinogenic health effects in long term consumption. Hazard's quotient values of different age categories did not exceed the value one (1 > HQ) for a short duration of water consumption. Children (Age category 1-9 years) are highly vulnerable to non-carcinogenic health hazards, and their HQ value exceeded one (HQ > 1) within a short period for fluoride (80 days), calcium (1,440 days), magnesium (2,160 days), and cadmium (360 days) before other age categories. HI mean values with higher concentrations elaborated that multicomponent concentration combinations bring adverse health effects on females in 1–9 and 10–19 years of age categories and males after 20 years of age. With mixture of component, age category 1–9 years exceeded HI>1 within 2 weeks for higher concentrations of the mixture, age category 10–19 years within one month, age category 2–90 years withing three months. The higher concentration value of components makes people vulnerable for adverse health hazard within short period of exposure. Long-run consumption of RO water causes non-carcinogenic health effects. Hence, developing a new water treatment unit is of utmost importance to provide safe drinking water.

The modified fly ash (Zeolite) (ZEOL), MgO loaded alumina (MOMA), silver oxide nanoparticle + graphene oxide composite (SONPs + GO) proposes the best combination of materials to remove hardness, fluoride, and faecal coliform in potable water after conducting batch and fixed-bed column studies. The fluoride (Q = 18.76 mg/g) and hardness (Q = 263.16 mg/g) experimental data aligned with the Langmuir model for batch studies. The fluoride and hardness data corroborated with the Thomas model for fixed-bed column studies. The length of unused bed values was calculated as 1.62 cm, 1.00 cm, and 0.81 cm for ZEOL, MOMA, and SONPs + GO when each material's breakthrough points were considered the maximum allowable concentration. The height of the ZEOL bed required to remove hardness for three months of service period was calculated as 29.09 cm with the mass of adsorbent 2.63 Kg, 18.86 cm adsorbent bed height including the mass of 1.37 Kg of MOMA, and 6.48 cm with the mass

of 1.09 Kg of SONPs + GO. The cost of 1.0 L of treated water production was approximately Rs. 8.80 and the total cost for 10.0 L of water (daily consumption of a family) was estimated at Rs. 88.00. If a family of five household members consumes water for three months, the cost of treated water production was calculated as Rs. 7,920.00 (monthly cost Rs. 2,640.00). The best combination of multi-layer materials is a promising water treatment unit to remove fluoride, hardness, and faecal coliform in drinking water. Therefore, the fabrication of a multi-layer home filter unit using ZEOL, MOMA, and SONPs + GO is recommended to provide safe, clean potable water for the community in CKDu prevalent areas.

Keywords: Adsorption, Faecal coliform, Fluoride, hardness, isotherms, kinetics, nanomaterial, risk assessment

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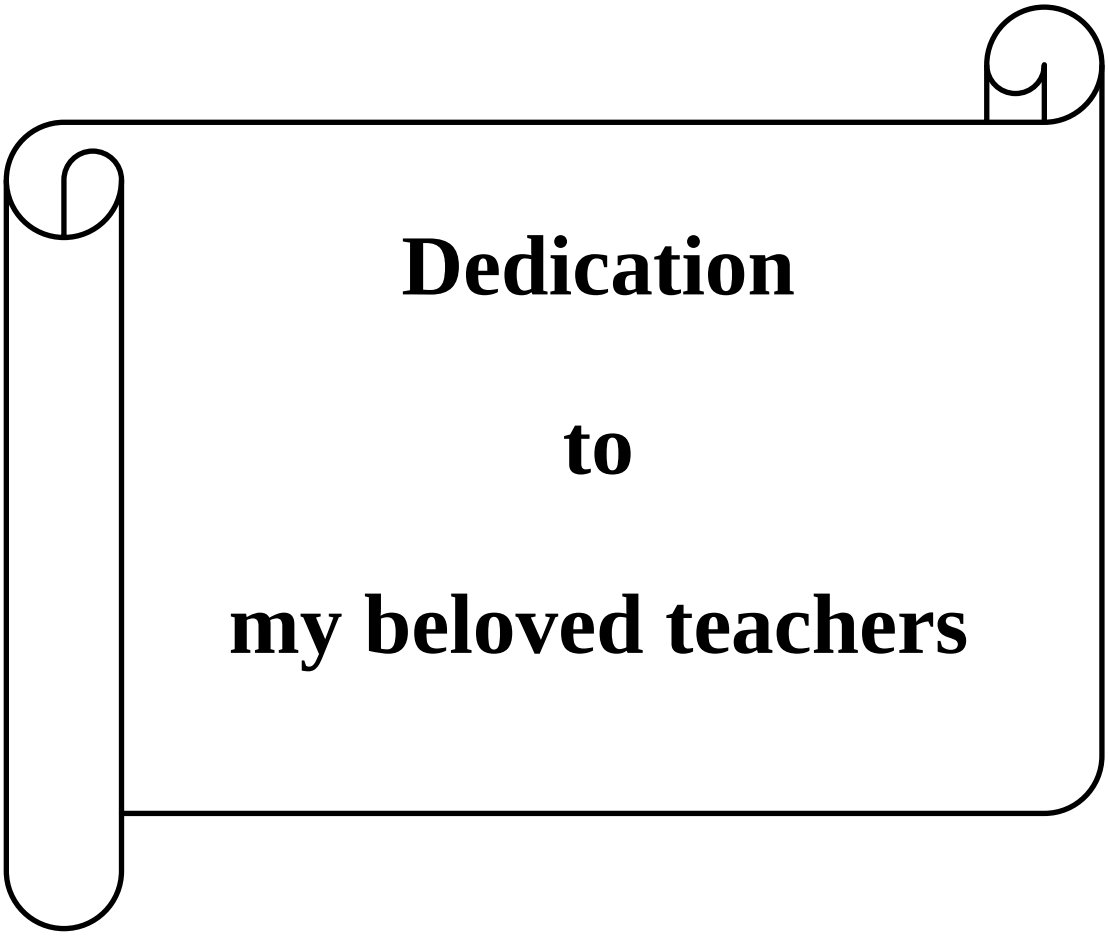
Dedication
to
my beloved teachers

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LIST OF ABBREVIATION

CKDu	- Chronic kidney disease of unknown aetiology
CKD	- Chronic Kidney Disease
WHO	- World health organisation
RO	- Reverse Osmosis
HQ	- Hazard quotient
MEN	- Mesoamerican nephropathy
BEN	- Balkan endemic nephropathy
CIN	- Chronic interstitial nephropathy
GFR	- Glomerular filtration rate
NCP	- North central province
CAN	- Chronic agrochemical nephrology
CINAC	- Chronic interstitial nephritis of agricultural communities
CKDmfo	- CKD of multifactorial origin
NSAIDs	- Nonsteroidal anti-inflammatory drugs
DOC	- Dissolved organic carbon
Cd	- Cadmium
ROS	- Reactive oxygen species
O_2^-	- Superoxide anion
$ROO^\cdot$	- Peroxyl radicals
H_2O_2	- Hydrogen peroxide
$R\text{-}NO^\cdot$	- Peroxynitrite
$OH^\cdot$	- Hydroxyl radicals
$O^\cdot$	- Singlet oxygen
$(CH_3)_2AsOO^\cdot$	- Dimethyl peroxyl radicals

$(CH_3)_2As^{\cdot}$ - Dimethyl arsenic radicals

Ca - Calcium

Mg - Magnesium

F - Fluoride

SLS - Sri Lanka Standards

US EPA - United States Environmental Protection Agency

NPDWRs - National Secondary Drinking Water Regulations

MDP - Meta-phenylenediamine

TMC - Trimesoyl Chloride

nZVI - Nano zero valent iron

CFU - Colony forming unit

MTZ - Mass transfer zone

XRD - X-ray powder diffraction

ESEM - Environmental scanning electron microscope

EDAX - Energy Dispersive Spectroscopy

FT–IR - Fourier-transform infrared spectroscopy

USGS - United States Geological Survey

SMART - Specific, measurable, attainable, relevant, and time-bound

$CaCO_3$ - Calcium carbonate

ADD - Average daily dose

HI - Hazard index

C - Concentration

IR - Intake rate

ED - Exposure duration

BW - Body weight

AT	- Average time
ZEOL	- Zeolite
GO	- Graphene oxide
FeONs	- Iron oxide nanoparticles
MESA	- Mesoporous alumina
Al_2O_3	- Alumina
COMA	- Calcium oxide loaded mesoporous alumina
MOMA	- Magnesium oxide loaded mesoporous alumina
TiO_2	- Titanium dioxide
SONPs	- Silver oxide nanoparticles
MO	- Microorganism
ACS	- American chemical society
PEG	- Polyethylene glycol
DI	- Deionized
BET	- Brunauer-Emmett-Teller
IAST	- Ideal adsorption solution theory
EDTA	- Ethylenediaminetetraacetic acid
ICP-MS	- Inductively coupled plasma mass spectrometry
USA	- United states of America
MPN	- Most probable number
E. coli	- *Escherichia coli*
EOM	- Electronic optical microscopy
EBCT	- Empty bed contact time
BV	- Bed volume

H_{MTZ}	- Length of mass transfer zone
ANOVA	- Analysis of variance
O & M	- Operation and maintenance
NGO	- Non-governmental organization
NaF	- Sodium fluoride
$CaCO_3$	- Calcium carbonate
$MgCO_3$	- Magnesium carbonate
IS	- Inner sphere
OS	- Outer sphere
DNA	- Deoxyribonucleic acid
RNA	- Ribonucleic acid
NAOH	- Sodium hydroxide
HCl	- Hydrochloric acid
EDTA	- Ethylenediaminetetraacetic acid
BTC	- Breakthrough curve