MULTI-CRITERIA FRAMEWORK FOR REMEDIATION OF CHROMITE ORE PROCESSING RESIDUE (COPR) OVERBURDEN AT CONTAMINATED SITES

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by

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Submitted

In the fulfilment of the requirements of the degree of Doctor of Philosophy

to the

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Dedicated to my parents and to

all the people who supported me

throughout my education.

कर्मण्येवाधिकारस्ते मा फलेषु कदाचन।

मा कर्मफलहेतुर्भूर्मा ते सङ्गोऽस्त्वकर्मणि॥

(Bhagavad Gita, Chapter 2, Verse: 47)

CERTIFICATE

This is to certify that the thesis entitled "**Multi-Criteria Framework for Remediation of Chromite Ore Processing Residue (COPR) Overburden at Contaminated Sites**" is being submitted by **Mr. Gummadivalli Shiva Kumar** to the **Indian Institute of Technology Delhi** for the award of the degree of '**Doctor of Philosophy**' in Department of Civil Engineering and is a record of the bonafide research work carried out by him under my supervision. He has fulfilled the requirements of the Indian Institute of Technology Delhi for the submission of this thesis, which to the best of my knowledge has reached the requisite standard.

The research material and results contained in the thesis have not been submitted in part or full to any other University or Institute for the award of any other degree or diploma.

(Prof. Arvind K. Nema)

Research Supervisor

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(GUMMADIVALLI SHIVA KUMAR)

ABSTRACT

Chromite ore processing residue (COPR) is a hazardous industrial waste derived from the chromate (CrO_4^{2-}) extraction roasting process. It is one of the major sources of environmental contamination as it contains high amounts of chromium (Cr) in, both, the hexavalent [Cr(VI)] and the trivalent [Cr(III)] valence states. COPR has been disposed of in unregulated surface (unlined) landfills in several nations across the world, including India, without consideration to its (eco)toxicological potential. It is among the most environmentally hazardous industrial chromium wastes, posing a threat to both human health and the environment. The leaching of Cr(VI), resulting in the contamination of groundwater, is a key environmental risk arising from these COPR dumps. Therefore, the remediation of COPR is of utmost importance, especially at contaminated sites where it was disposed of in an unscientific manner.

COPR from two of the legacy waste-contaminated sites [Ranipet (TCCL, Tamil Nadu, India) and Rania (near Khan Chandpur village, Kanpur Dehat, Uttar Pradesh, India)] are collected and characterized. Metal content analysis and TCLP-based leaching analysis were performed to arrive at detailed waste properties. In addition to the physicochemical characterization of COPR waste from both sites, the mineralogical composition was also evaluated. The complexity of chromium chemistry, the high exposure risk potential posed by COPR, coupled with the quantity of wastes at these sites advocate for an urgent need to remediate them, and demands the development of a remediation framework that can address the difficulties posed when handling multiple criteria.

National-level clean-up standards (or remediation targets or intervention values or guidelines) associated with Cr-contaminated sites are established only for a few countries. Upon detailed literature review, two sets of standards (Canadian and Dutch) were considered. Considering the two selected COPR sites and two remediation standards, four combinative cases were formulated.

The most appropriate technology alternative for COPR remediation in each of these four cases would be estimated using a multi-criteria framework.

Various aspects regarding the remediation of Cr-contaminated sites that have COPR as the major source of Cr are studied in existing academic literature and databases. A panel of experts (with 4 stakeholder groups) was queried in a 3-phase (4-stage) Delphi process about such aspects of COPR remediation that were not gathered in the literature survey. Their opinions are used to — ascertain and shortlist relevant criteria, create a 'pair-wise comparison matrix' among selected criteria, identify candidate treatment technologies for COPR remediation, and generate an 'information evaluation matrix' between remediation technology and criteria.

A novel resourceful multicriteria analysis framework, PROSPER-COPR [Prioritization of Remedial Options through Systematic Parametric Evaluation and Ranking for contaminated sites having Chromium Ore Processing Residue (COPR) overburden], was developed to arrive at the weights for finalized criteria and to determine the ranking between alternative technologies. Though POSPER can be applied to any remediation of any waste-contaminated site, PROSPER-COPR is directed towards addressing legacy COPR overburden at contaminated sites. As part of the PROSPER-COPR framework, a variety of multicriteria decision-making (MCDM) methods and weighting methods were employed to calculate criteria weights (both, weights applicable to any site and case-specific weights), as well as to evaluate the rankings between the considered remediation technology alternatives. Fuzzy-AHP, Entropy, and CRITIC methods were used for the calculation of weights; and, TOPSIS and PROMETHEE-II were applied to determine the ranks among various shortlisted remediation technologies. Upon arriving at the case-specific optimal solutions, a novel 'Technology Improvement Need Analysis (TINA)' framework has been developed to perform the efficiency analysis of all the remediation technology alternatives or decision-making units (DMUs) for all cases. The aim of measuring the efficiency of DMUs is to quantify the magnitude by which each of the suboptimal alternatives must be improved for every criterion in order for them to become the optimal candidate of choice.

To measure the efficiency of DMUs, two non-parametric deterministic mathematical multi-objective decision-making (MODM) tools — the Data Envelopment Analysis (DEA) method and the Free Disposal Hull (FDH) method, are used. Two well-known basic DEA models i.e., Charnes, Cooper, and Rhodes (CCR) model and Banker, Chames, and Cooper (BCC) model, are considered, both, with input orientation. Appropriate solutions are proposed for physically improving the criteria-wise performance of the technologies before their field implementations. The research outcomes are expected to provide an all-inclusive framework to decision-makers across the world for effective remediation of COPR at existing sites, which can be easily replicable to any toxic and hazardous waste.

Keywords: Heavy metal contamination, Chromite Ore Processing Residue, COPR, Contaminated sites, Remediation, Delphi method, MCDA.

सारांश

क्रोमाइट अयस्क प्रसंस्करण अवशेष (COPR) क्रोमेट (CrO_4^{2-}) निष्कर्षण रोस्टिंग प्रक्रिया से प्राप्त एक विषाक्त औद्योगिक अपशिष्ट है। यह पर्यावरण प्रदूषण के प्रमुख स्रोतों में से एक है क्योंकि इसमें हेक्सावलेंट [Cr(VI)] और ट्राईवलेंट [Cr(III)] वैलेंस अवस्थाओं दोनों में क्रोमियम (Cr) की उच्च मात्रा होती है। सीओपीआर को भारत सहित दुनिया भर के कई देशों में अनियमित सतह (अरेखित) लैंडफिल में इसकी (पर्यावरण) विषैली क्षमता पर विचार किए बिना निपटाया गया है। यह पर्यावरण की दृष्टि से सबसे खतरनाक औद्योगिक क्रोमियम कचरे में से एक है, जो मानव स्वास्थ्य और पर्यावरण दोनों के लिए खतरा पैदा करता है। सीआर(VI) का रिसाव, जिसके परिणामस्वरूप भूजल प्रदूषित होता है, इन COPR डंपों से उत्पन्न होने वाला एक प्रमुख पर्यावरणीय जोखिम है। इसलिए, सीओपीआर का निवारण अत्यंत महत्वपूर्ण है, विशेषकर दूषित स्थलों पर जहां इसका अवैज्ञानिक तरीके से निपटान किया गया था।

दो विरासती अपशिष्ट दूषित स्थलों [रानीपेट (टीसीसीएल, तमिलनाडु, भारत) और रानिया (खान चांदपुर गांव के पास, कानपुर देहात, उत्तर प्रदेश, भारत)] से सीओपीआर एकत्र किया गया है और भौतिक-रासायनिक स्वभाव बताया गया है। विस्तृत अपशिष्ट गुणों पर पहुंचने के लिए धातु सामग्री विश्लेषण और टीसीएलपी-आधारित लीचिंग विश्लेषण किया गया। दोनों स्थलों से सीओपीआर अपशिष्ट के भौतिक-रासायनिक लक्षण वर्णन के अलावा, खनिज संरचना का भी मूल्यांकन किया गया था। क्रोमियम रसायन विज्ञान की जटिलता, सीओपीआर द्वारा उत्पन्न उच्च ऐक्सपोज़र रिस्क क्षमता, इन साइटों पर कचरे की मात्रा के साथ मिलकर उन्हें ठीक करने की तत्काल आवश्यकता की वकालत करती है, और एक उपचार ढांचे के विकास की मांग करती है जो कई मानदंडों को संभालने के दौरान उत्पन्न होने वाली कठिनाइयों का समाधान कर सके।

Cr-दूषित साइटों से जुड़े राष्ट्रीय स्तर के सफाई मानक (या उपचार लक्ष्य या हस्तक्षेप मूल्य या दिशानिर्देश) केवल कुछ देशों के लिए स्थापित किए गए हैं। विस्तृत साहित्य समीक्षा के बाद, मानकों के दो सेट (कनाडाई और डच) पर विचार किया गया। दो चयनित सीओपीआर साइटों और दो उपचार मानकों को ध्यान में रखते हुए, चार संयोजन मामले तैयार किए गए थे। इन चार मामलों में से प्रत्येक में सीओपीआर निवारण के लिए सबसे उपयुक्त प्रौद्योगिकी विकल्प का अनुमान बहु-मानदंड ढांचे का उपयोग करके लगाया जाएगा।

Cr-दूषित साइटों के निवारण के संबंध में विभिन्न पहलुओं, जिनमें सीओपीआर, सीआर का प्रमुख स्रोत है, का मौजूदा शैक्षणिक साहित्य और डेटाबेस में अध्ययन किया गया है। सीओपीआर उपचार के ऐसे पहलुओं के बारे में 3-अवस्था (4-चरण) वाली डेल्फ़ी प्रक्रिया में विशेषज्ञों के एक पैनल (4 विशेषज्ञ समूहों के साथ) से पूछताछ की गई जो साहित्य सर्वेक्षण में एकत्र नहीं किए गए थे। उनकी राय का उपयोग प्रासंगिक मानदंडों का पता लगाने और शॉर्टलिस्ट करने, चयनित मानदंडों के बीच 'जोड़ी-अनुसार तुलना मैट्रिक्स' बनाने, सीओपीआर उपचार के लिए उम्मीदवार उपचार प्रौद्योगिकियों की पहचान करने, उपचार प्रौद्योगिकी और मानदंडों के बीच 'सूचना मूल्यांकन मैट्रिक्स' उत्पन्न करने के लिए किया जाता है।

अंतिम मानदंडों के लिए वजन पर पहुंचने और वैकल्पिक प्रौद्योगिकियों के बीच रैंकिंग निर्धारित करने के लिए एक नया संसाधनपूर्ण बहुमानदंड विश्लेषण ढांचा, PROSPER-COPR [क्रोमियम अयस्क प्रसंस्करण अवशेष (सीओपीआर) ओवरबर्डन वाले दूषित साइटों के लिए व्यवस्थित पैरामीट्रिक मूल्यांकन और रैंकिंग के माध्यम से उपचारात्मक विकल्पों को प्राथमिकता देना] विकसित किया गया था। हालाँकि PROSPER को किसी भी अपशिष्ट-दूषित साइट के उपचार के लिए लागू किया जा सकता है, PROSPER-COPR का उद्देश्य दूषित साइटों पर पुराने COPR ओवरबर्डन को संबोधित करना है। प्रॉस्पर-सीओपीआर ढांचे के हिस्से के रूप में, मानदंड भार (दोनों, किसी भी साइट पर लागू वजन और केस-विशिष्ट वजन) की गणना करने के लिए, साथ ही विचार किए गए उपचारात्मक प्रौद्योगिकी विकल्पों के बीच रैंकिंग का मूल्यांकन करने के लिए

विभिन्न प्रकार के बहुमानदंड निर्णय लेने (एमसीडीएम) के तरीकों और वजन तरीकों को नियोजित किया गया था। वज़न की गणना के लिए फ़ज़ी-एएचपी, एन्ट्रॉपी और क्रिटिक विधियों का उपयोग किया गया था; और, TOPSIS और PROMETHEE-II को विभिन्न शॉर्टलिस्टेड उपचार प्रौद्योगिकियों के बीच रैंक निर्धारित करने के लिए लागू किया गया है। मामले-विशिष्ट इष्टतम समाधानों पर पहुंचने पर, सभी मामलों के लिए सभी उपचारात्मक प्रौद्योगिकी विकल्पों या निर्णय लेने वाली इकाइयों (डीएमयू) की दक्षता विश्लेषण करने के लिए एक नवीन 'प्रौद्योगिकी सुधार आवश्यकता विश्लेषण (TINA)' विकसित किया गया है। डीएमयू की दक्षता को मापने का उद्देश्य उस परिमाण को मापना है जिसके द्वारा प्रत्येक मानदंड के लिए प्रत्येक उप-इष्टतम विकल्पों में सुधार किया जाना चाहिए ताकि वे पसंद के इष्टतम उम्मीदवार बन सकें।

डीएमयू की दक्षता को मापने के लिए, दो गैर-पैरामीट्रिक नियतात्मक गणितीय बहुउद्देश्यीय निर्णय लेने (एमओडीएम) के उपकरण - डेटा एनवेलपमेंट विश्लेषण (डीईए) विधि और फ्री डिस्पोजल हल (एफडीएच) विधि का उपयोग किया जाता है। दो प्रसिद्ध बुनियादी डीईए मॉडल यानी, चार्नेस, कूपर और रोड्स (सीसीआर) मॉडल और बैंकर, चेम्स और कूपर (बीसीसी) मॉडल, दोनों को इनपुट ओरिएंटेशन के साथ माना जाता है। प्रौद्योगिकियों के क्षेत्र में कार्यान्वयन से पहले उनके मानदंड- अनुसार प्रदर्शन को भौतिक रूप से सुधारने के लिए उचित समाधान प्रस्तावित हैं। शोध के परिणामों से मौजूदा साइटों पर सीओपीआर के प्रभावी निवारण के लिए निर्णय निर्माताओं को एक सर्व-समावेशी ढांचा प्रदान करने की उम्मीद है, जिसे आसानी से किसी भी रासायनिक कचरे के लिए दोहराया जा सकता है।

कीवर्ड: भारी धातु संदूषण, क्रोमाइट अयस्क प्रसंस्करण अवशेष, सीओपीआर, दूषित साइट, उपचार, डेल्फ़ी विधि, एमसीडीए।

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ARAS	Additive RAtio Assessment
A+F	Aesthetics + Future use of the site (Post Remediation)
APHA	American Public Health Association
ASTM	American Standard Test Methods
AHP	Analytic Hierarchy Process
ANP	Analytic Network Process
AHP-AM	Analytical Hierarchy Process Arithmetic Mean Method
AHP-GM	Analytical Hierarchy Process Geometric Mean Method
BCC	Banker, Chames and Cooper model
BCS	Basic Chromium Sulphate
BDL	Below Detection Limit
Be	Beneficial
$HCrO_4^{2-}$	Bichromate
B	Brownmillerite
Br	Brucite
C’	Calcite
Ca	Calcium
CAC	Calcium Aluminum Chromium Oxide Hydrate
CLRI	Central Leather Research Institute
CPCB	Central Pollution Control Board
CCR	Charnes, Cooper and Rhodes model
CrO_4^{2-}	Chromate
Cr_2O_3	Chromic Oxide
COPR	Chromite Ore Processing Residue
Cr(0)	Chromium element
$Cr(OH)_3$	Chromium Hydroxide
COPRAS	COmplex PRoportional ASsessment
CR	Consistency Ratio
C	Cost
CSIR	Council of Scientific and Industrial Research
CW-III	Criteria Weights (Combined)
CW-I	Criteria Weights (For any site)
CW-II	Criteria Weights (Site-specific)
DEA	Data Envelopment Analysis
DSS	Decision Support Systems
DEMATEL	DEcision-MAking Trial and Evaluation Laboratory
DMUs	Decision-Making Units
DPR	Detailed Project Report
Ph.D.	Doctor of Philosophy
D-models	Dynamic Models
EI	Ease of Implementation (Post Remediation)

ELECTRE	ÉLimination Et Choix Traduisant la REalité (ELimination Et Choice Translating REality)
EPA	Environmental Protection Agency
ER/R	Environmental Risk (Post Remediation)
EEC	Estimated environmental contaminant concentration
E+OFS1	Excavation + Off-site Treatment-1
E+OFS2	Excavation + Off-site Treatment-2
E+ONS1	Excavation + On-site Treatment-1
E+ONS2	Excavation + On-site Treatment-2
E-R	Exposure-Risk
SW-E	Ex-situ soil washing
$FeSO_4$	Ferrous Sulphate
FDH	Free Disposal Hull model
F-AHP	Fuzzy Arithmetic Hierarchy Process
GA	Genetic Algorithm
GM	Geometric Mean
GP	Goal Programming
GB COPR	Grey Black COPR
G	Grimaldiite
GW	Ground water
GJ	Gujarat
HB COPR	Hard Brown COPR
HQ	Hazard Quotient
Cr(VI)	Hexavalent Chromium
HPSO	Hybrid Particle Swarm Optimization
IIT	Indian Institute of Technology
IITR	Indian Institute of Toxicology Research
ICP-MS	Inductively Coupled Plasma Mass Spectrometer
IM	Information Matrix
SW-I	In-situ soil washing
Fe	Iron
K	Katoite
L	Larnite
LP	Linear Programming
LP	Long-term Performance (Post Remediation)
Cr	Magnesiochromite
Mg	Magnesium
Mn	Manganese
M.Tech	Master of Technology
MLT	Membrane Layer Treatment
MIP	Mixed Integer Programming
MDL	Modified Digital Logic
MODM	Multi Objective Decision-Making

MADM	Multi-Attribute Decision Making
MAV(U)T	Multi-Attribute Value (Utility) Theory
MCDA	Multi-Criteria Decision Analysis
MCDM	Multi-Criteria Decision-Making
MOORA	Multi-Objective Optimization on the basis of Ratio Analysis
NEERI	National Environmental Engineering Research Institute
NIT	National Institute of Technology
NPL	National Priorities List
NILP	New Iterative Linear Programming
NJ	New Jersey
NA	No Action
NOAEL	NO Adverse Effects Level
NBe	Non-beneficial
NSGA	Non-dominated Sorting Genetic Algorithm
NGO	Non-Governmental Organization
K_d	Partition Coefficient
P	Periclase
Pt	Portlandite
PROMETHEE	Preference Ranking Organization METHod for Enrichment Evaluation
QFD	Quality Function Deployment
Q	Quartz
R&D	Research and Development
SP	Short-term Performance (Post Remediation)
SAW	Simple Additive Weighting
SA	Simulated Annealing
SODM	Single Objective Decision-Making
S	Sjogrenite
SA	Social acceptance and impacts
Na_2CrO_4	Sodium Chromate
$Na_2Cr_2O_7$	Sodium Dichromate
SC	Soil Cover
SIPCOT	State Industries Promotion Corporation Of Tamil Nadu
SPCB	State Pollution Control Board
SWARA	Stepwise Weight Assessment Ratio Analysis
SU	Sustainability during the implementation of the remediation technology
TS	Tabu Search
TN	Tamil Nadu
TCCL	Tamil Nadu Chromates and Chemicals Limited
TNPCB	Tamil Nadu Pollution Control Board
TSP	Technical success potential of the remediation technology
TOPSIS	Technique for Order Performance by Similarity to Ideal Solution
TINA	Technology Improvement Need Analysis
T	Time needed to implement the remediation technology

TCLP	Toxicity Characteristic Leaching Procedure
TR	Toxicity reduction (Post Remediation)
Cr(III)	Trivalent Chromium
US EPA	United States Environment Protection Agency
USA	United States of America
UP	Uttar Pradesh
UPPCB	Uttar Pradesh Pollution Control Board
VIKOR	VIse Kriterijumska Optimizacija I Kompromisno Resenje (Multi-criteria optimization and compromise solution)
V	Voltaite
WB	West Bengal
XRD	X-Ray Diffraction
PROSPER	Prioritization of Remedial Options through Systematic Parametric Evaluation and Ranking for contaminated sites having Chromium Ore Processing Residue (COPR) overburden
MAUT	Multi-Attribute Utility Theory
MAVT	Multi-Attribute Value Theory
MACBETH	Measuring Attractiveness by a Categorical-Based Evaluation Technique
CODAS	COmbinative Distance-based ASsessment
MCA	Multi-Criteria Assessment