Contract No:

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Title: Functionalized Mesoporous Carbon Materials for Effective Recovery of Rare Earth Elements from NdFeB Magnet Scrap

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Objectives: The increasing demand and high dependence on international supplies of rare earth elements (REEs) have prompted the US to explore alternative resources and sustainable technologies for domestic REE production.[1] One potential resource is the secondary electronic and industrial wastes at DoD complexes and across the US. These hazardous wastes require high costs for the maintenance and remediation, but their high-REE contents offer unique opportunities for REE recovery.[1] Current hydrometallurgical technologies for REE extraction are energy intensive and costly, with low REE recovery efficiency and the generation of hazardous secondary wastes. Thus, it is crucial to develop high efficiency and environmental-friendly technologies for REE extraction and recovery from the secondary wastes. This project aims to develop novel functionalized mesoporous carbon materials for effective recovery of REEs from NdFeB magnet scrap. The proposed novel functionalized materials will be applied in a solid-liquid separation platform that can effectively extract, recover and separate REEs from NdFeB magnet scrap (the current focus), but also from other secondary electronic/industrial wastes in the future, minimize the generation of secondary hazardous wastes, and offset the management costs for existing waste streams.

Approach: This whitepaper outlines the plans and significances for continuing and expanding our SERDP SEED project WP22-3463. Data obtained from our SEED project demonstrated the proof-of-concepts on environmental-friendly ligand-assisted REE extraction, synthesis and functionalization of tunable mesoporous carbon fibers (MCFs) for improved REE recovery, and MCF material regeneration for reuse and REE recycling.[2] Our planned approach for future research is to use combined rational material design, theoretical computation and machine learning, advanced molecular-level characterization, and robust engineering processes to establish and optimize this advanced separation system (**Figure 1**). Such combined experimental-computational approach will significantly expedite rational materials design and advance our understanding of the driving factors for efficient REE separation. Building on the initial success of our SEED project, key experimental focus areas will continue to include the synthesis of stable/tunable MCFs and other carbon materials, organic ligand functionalization into pores, and surface reactivity of the new materials for REEs extraction and recovery. In addition, we will pursue computation and machine learning approaches for rational materials design and optimization, as well as upscaling, demonstration, and evaluation of REE recovery from real NdFeB scrap and other waste streams. The specific tasks are discussed below.

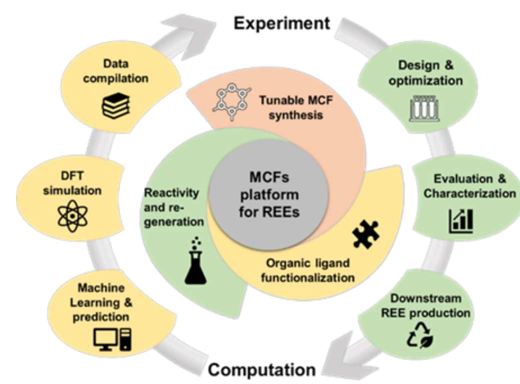

Figure 1. Strategic plan of the future research.

Task 1. Ligand assisted extraction of NdFeB powder and scrap. Inexpensive and biodegradable organic ligands (e.g., citrate) will be used as environmental-friendly ligands for REE extraction. We will study the impacts of ligand type/concentration, solution pH, and reaction time/cycle on REE extraction efficiency. Following extraction, we will evaluate oxalate assisted precipitation to separate REEs from other impurity metals such as Al and Fe, which have high concentrations in the real extraction solutions and can interfere with downstream REE recovery. The resulting REE-oxalate precipitates can achieve near 100% REE recovery with minimal amount of other metal impurities and will be used as a new feedstock for downstream REE purification and separation using carbon materials. The goal of this task is to reduce mineral acid use and secondary waste generation while improving REE extraction efficiency.

Task 2. Synthesis and characterization of tunable MCFs. Polymethyl methacrylate (PMMA)–Polyacrylonitrile (PAN) block co-polymer (PMMA-b-PAN) will be used as a template for the synthesis of

MCFs.[3] The co-polymer solution is electrospun onto a fiber mat. After phase-separation, polymer fibers are oxidized and carbonized into porous carbon fibers. The PMMA-b-PAN block copolymer composition controls the pore size, surface area, and ion diffusion. The MCF materials will be characterized using SEM, N_2 sorption measurements, and other conventional analytical methods. The goal of this task is to make MCFs materials with tunable surface area, pore size, pore structure and surface functionalities for high REE recovery efficiency.

Task 3. Functionalization of MCFs with novel organic ligands. Novel organic ligands (e.g., diglycolamide, diethylenetriaminepentaacetic acid, phosphonate, and polyethyleneimine) will be functionalized onto MCFs using post-synthetic methods. The functionalized materials will be thoroughly characterized and evaluated using batch experiments for REE recovery efficiency, selectivity, regeneration, reuse and REE recycling from simulants and real extractions of NdFeB powder or scrap. The goal of this task is to develop new functionalized MCFs for improved REE recovery selectivity and capacity.

Task 4. Molecular-level mechanisms on REE-ligand interactions. It is important to understand molecular-level mechanisms of REE recovery. State-of-the-art synchrotron X-ray absorption spectroscopy will be used to determine REE chemical speciation and probe REE-ligand interactions at molecular, nano and micro scales. The goal of this task is to help design new functional materials for improved REE recovery efficiency and more robust processes for REE recycling.

Task 5. Computational calculation and machine learning for materials design and optimization. This new strategy will integrate experimental methods with computational modelling and machine learning to further improve the rational design and optimization of new functionalized mesoporous materials. We have established collaboration with computational materials scientists from the University of South Carolina on another project and can readily add this capability to this project.

Task 6. Upscaling and demonstration of REE recovery from NdFeB magnet scrap. Based on the fundamental research on material design and optimization, we will down select high performance materials for upscale demonstration of the overall REE extraction and separation system using NdFeB magnet scrap and other electronic/industrial waste streams such as those from DoD complexes. The proposed MCF materials are facile scalable for their synthesis. They will be built into a solid-liquid separation system to recover REEs from the extraction solutions and recycle REEs. The unique pore shape and structure of MCFs are expected to facilitate the mobility of leachates through the pores, reduce clog and improve life span of the materials.

The new functional MCF materials will bring the following technical benefits for selective REE recovery from NdFeB magnet scrape and other waste sources: **1)** improved selectivity and extraction capacity for REEs, **2)** improved material stability, life span, and reusability, **3)** reduced processing steps and costs for REE recovery and separation, **4)** reduced release of toxic wastes and risks to human health and environment, **5)** high scalability and robustness of the separation technology. This technology will ultimately facilitate the domestic recovery of REE from a variety of electronic and industrial wastes in environmentally friendly manner, while mitigating the environmental risks of the wastes at DoD complexes and across the US. As such, the proposed research will **advance the DoD and US's capability** of *securing the future REE material supplies in environmentally sound manner*s *whiling reducing the management risk and remediation cost of electronic and industrial waste streams.*

Cost Estimate:

	FY23	FY24	FY25
Total funding ($K)	450	450	450

Literature Citations: **(1)** U.S. Department of Energy: *Critical Material Strategy*; U.S. Department of Energy: Washington, D.C., 2010. **(2)** Li, D.; Tang, Y. Z.; Liu, G. L.: *Functionalized Mesoporous Carbon and Silica for Effective Recovery of Rare Earth Elements from Magnet Scrap, SRNL-STI-2023-00049*; Savannah River National Laboratory: Aiken, SC 29808, 2023. **(3)** Zhou, Z. P.; Liu, T. Y.; Khan, A. U.; Liu, G. L. Block copolymer-based porous carbon fibers. *Science Advances* **2019**, *5*, eaau6852.