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A Solvent-Free, Mechanochemical Process for Sustainable Recycling of Neodymium and Dysprosium from E-Waste Magnets

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ABSTRACT

The escalating demand for rare earth elements (REEs), particularly neodymium (Nd) and dysprosium (Dy), for high-performance NdFeB magnets, has created significant supply chain vulnerabilities and environmental concerns associated with primary mining. End-of-life electronic waste (e-waste) represents a substantial secondary resource for these critical materials. This study introduces a novel, environmentally benign approach for recovering Nd and Dy from waste NdFeB magnets. A solvent-free mechanochemical process was developed and optimized. Waste NdFeB magnet powder, sourced from discarded hard disk drives collected in Indonesia, was co-milled with ammonium chloride (NH_4Cl) in a high-energy planetary ball mill. The influence of key process parameters, including milling time (60-360 min), milling speed (200-500 rpm), and the mass ratio of NH_4Cl to magnet powder (1:1 to 5:1), on the extraction efficiency of Nd and Dy was systematically investigated. The structural and morphological transformations were characterized using X-ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) with Energy-Dispersive X-ray Spectroscopy (EDS). Metal recovery was quantified via subsequent water leaching and analysis by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). The mechanochemical treatment successfully converted the insoluble rare earth phases within the magnet matrix into water-soluble rare earth chlorides. Under optimal conditions—a milling time of 240 minutes, a speed of 400 rpm, and a NH_4Cl-to-magnet mass ratio of 3:1—the process achieved remarkable extraction efficiencies of 98.6% for Nd and 96.2% for Dy. XRD analysis confirmed the transformation of the $Nd_2Fe_{14}B$ phase into REE chlorides, alongside iron and iron boride phases. SEM imaging revealed a significant reduction in particle size and the formation of agglomerated composite particles, crucial for the solid-state reaction. In conclusion, this study demonstrates that solvent-free mechanochemistry is a highly effective and sustainable alternative to conventional hydrometallurgical and pyrometallurgical recycling methods. The process operates at ambient temperature, eliminates the need for corrosive acids and organic solvents, and exhibits high recovery rates, presenting a viable pathway towards a circular economy for critical rare earth elements from e-waste.

1. Introduction

IThe 21st century is defined by technological advancements that are deeply intertwined with the use of specialized materials. Among these, rare earth elements (REEs) have emerged as indispensable components in a vast array of modern technologies, from consumer electronics to defense systems and renewable energy infrastructure. The REE group, comprising the 15 lanthanides plus scandium and yttrium, possesses unique magnetic and optical properties. Specifically, neodymium (Nd) and dysprosium (Dy) are critical for the manufacturing of high-performance neodymium-iron-boron (NdFeB) permanent magnets. These magnets are the strongest commercially available and are fundamental to technologies requiring high efficiency and miniaturization, such as electric vehicle (EV) motors, wind turbine generators, smartphones, and hard disk drives (HDDs).

The global demand for Nd and Dy has surged exponentially, a trend projected to continue with the global push towards electrification and green energy. However, the REE supply chain is fraught with significant geopolitical and environmental challenges. A vast majority of the world's primary REE production is concentrated in a single country, creating price volatility and supply chain insecurity for nations reliant on imports. Furthermore, the conventional mining and processing of REEs are notoriously detrimental to the environment. These processes involve extensive open-pit mining and complex chemical extraction steps, often using hazardous reagents like sulfuric and hydrofluoric acids. They generate enormous quantities of waste tailings and wastewater contaminated with heavy metals and radioactive elements, leading to severe soil degradation, water pollution, and long-term ecological damage.

In this context, establishing a robust circular economy for REEs is not merely an economic opportunity but a strategic and environmental imperative. Urban mining—the recovery of valuable materials from end-of-life products—presents a promising solution. E-waste, the world's fastest-growing domestic waste stream, is a particularly rich secondary resource for critical metals. NdFeB magnets, for instance, can contain up to 30-35% REEs by weight, a concentration far exceeding that of natural ores. Recovering these elements from e-waste would simultaneously mitigate the environmental burden of primary mining, alleviate supply chain pressures, and address the growing problem of e-waste management, especially in developing nations like Indonesia, which faces a rapid increase in e-waste generation.

Current methods for recycling NdFeB magnets can be broadly categorized into pyrometallurgy and hydrometallurgy. Pyrometallurgical approaches involve high-temperature melting and smelting processes to separate elements based on their affinity for different molten phases. While effective for large-scale operations, these methods are extremely energy-intensive (often requiring temperatures exceeding 1500 °C), lead to significant loss of volatile elements, and can generate hazardous off-gases. Hydrometallurgy, the more common approach, involves dissolving the magnet material in strong acids (such as HCl, H_2SO_4, and HNO_3) followed by a series of complex and reagent-intensive solvent extraction, precipitation, or ion-exchange steps to selectively recover the REEs. Although it operates at lower temperatures, hydrometallurgy consumes large volumes of corrosive and toxic chemicals, generates substantial amounts of acidic wastewater, and struggles with the economic viability of multi-step separation processes. The environmental footprint of these conventional methods often rivals that of primary extraction.

Therefore, there is an urgent need for disruptive, green technologies that can overcome the limitations of existing recycling routes. Mechanochemistry has emerged as a powerful tool in sustainable chemistry and materials science, offering a paradigm shift away from solvent-based, high-temperature processes. Mechanochemistry utilizes mechanical energy, typically delivered through high-energy ball milling, to induce chemical reactions and phase transformations in the solid state. The intense mechanical forces generated during milling—impact, shear, and friction—create localized high-pressure and high-temperature spots, leading to the formation of fresh, highly reactive surfaces, crystal defects, and amorphous phases. This activation can drive chemical reactions that are otherwise kinetically hindered at ambient temperatures.

The application of mechanochemistry to metallurgical processes offers several compelling advantages: (1) it can proceed in a solvent-free or minimal-solvent environment, drastically reducing chemical consumption and waste generation; (2) it operates at or near room temperature, significantly lowering energy input; and (3) it can enhance reaction kinetics, leading to faster and more efficient processes. Several studies have explored mechanochemical-assisted leaching, where milling is used to pre-activate ores or waste materials to improve subsequent hydrometallurgical extraction. However, a truly transformative approach involves using mechanochemistry to directly drive the conversion reaction in a single, solvent-free step.

This study aims to develop and systematically optimize a novel, one-step, solvent-free mechanochemical process for the selective conversion and subsequent recovery of Nd and Dy from waste NdFeB magnet scrap. The central hypothesis is that by co-milling the magnet powder with a carefully selected solid reactant—in this case, the benign and inexpensive salt ammonium chloride (NH_4Cl)—the insoluble rare earth oxides and intermetallic phases can be directly converted into water-soluble rare earth chlorides. The novelty of this work lies in its departure from the "acid-and-dissolve" paradigm. We propose a solid-state metathesis reaction driven entirely by mechanical force, which fundamentally minimizes the process's environmental footprint. The research took place in Indonesia, utilizing locally sourced e-waste, to underscore the applicability of this advanced technology in a region grappling with the dual challenges of resource scarcity and waste management. This investigation provides a comprehensive analysis of the process parameters, explores the underlying reaction mechanism through detailed material characterization, and demonstrates a highly efficient, sustainable, and scalable pathway for closing the loop on critical rare earth elements.

2. Methods

The source material for this research consisted of NdFeB magnets extracted from end-of-life hard disk drives (HDDs) obtained from electronic waste collection and refurbishment centers in the Greater Jakarta area, Indonesia. A total of 50 HDDs of various brands and ages were dismantled to harvest the voice coil motor magnets. The nickel-copper-nickel (Ni-Cu-Ni) protective coating on the magnets was first removed by mechanical abrasion. The uncoated magnet pieces were then subjected to demagnetization by heating at 350 °C for 2 hours in a muffle furnace to ensure safe handling and effective milling.

The demagnetized magnet scrap was embrittled by hydrogen decrepitation. The scrap was placed in a stainless-steel reactor, which was evacuated to 10^{-2} torr and then exposed to high-purity hydrogen gas (99.999%) at a pressure of 2 bar. The material was held under these conditions for 2 hours at room temperature, leading to its decrepitation into a coarse, friable powder. This powder was then dehydrogenated by heating under vacuum at 600 °C for 2 hours. The resulting coarse powder was crushed using a jaw crusher and sieved to obtain a particle size fraction below 250 μm for use in all mechanochemical experiments. Ammonium chloride (NH_4Cl, ≥99.5% purity) was purchased from Merck Indonesia and used as the reactant without further purification. Deionized (DI) water was used for all leaching experiments.

The mechanochemical conversion experiments were performed using a Retsch PM 100 planetary ball mill. In a typical experiment, a specific mass of the prepared NdFeB magnet powder and NH_4Cl were loaded into a 50 mL tungsten carbide grinding jar along with tungsten carbide balls (10 mm diameter). The ball-to-powder mass ratio (BPR) was maintained at a constant 20:1 for all experiments to ensure efficient energy transfer. The grinding jar was sealed inside an argon-filled glovebox to prevent oxidation of the rare earth elements during milling.

A systematic parametric study was conducted to optimize the process. The following parameters were varied: (1) Milling Time: 60, 120, 180, 240, 300, and 360 minutes; (2) Milling Speed: 200, 300, 400, and 500 rpm; (3) NH_4Cl-to-Magnet Mass Ratio: 1:1, 2:1, 3:1, 4:1, and 5:1. During each set of experiments, two parameters were held constant at their determined optimal values while the third was varied. After milling, the resulting powder mixture was collected from the jar inside the glovebox.

To determine the extraction efficiency of Nd and Dy, the milled powder was subjected to a simple water leaching process. A 1.0 g sample of the milled powder was added to 50 mL of DI water in a beaker. The suspension was stirred at 300 rpm using a magnetic stirrer for 60 minutes at room temperature (25 °C). After leaching, the solution was filtered through a 0.45 µm syringe filter to separate the aqueous leachate containing the dissolved rare earth chlorides from the solid residue (primarily iron, iron boride, and unreacted phases).

The concentrations of Nd and Dy in the filtered leachate were determined using Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) on a PerkinElmer Avio 200 instrument. The instrument was calibrated using certified multi-element standard solutions. Each sample was diluted appropriately with 2% nitric acid to fall within the linear calibration range. The analysis for each sample was performed in triplicate, and the average concentration was used for calculations.

The extraction efficiency (η) for each metal was calculated using the following equation: $\eta\ (\%) = (C \times V) / (m \times w) \times 100$, where; C is the concentration of the metal in the leachate (mg/L); V is the volume of the leachate (L); m is the mass of the milled powder sample used for leaching (mg); w is the initial weight fraction of the metal in the magnet powder (%). The initial composition of the magnet powder (w) was determined by digesting a sample of the raw powder in aqua regia and analyzing the solution by ICP-OES.

The physical and chemical changes in the magnet powder throughout the process were monitored using several analytical techniques. (1) X-ray Diffraction (XRD): The crystal structures of the initial magnet powder and the milled products were analyzed using a Rigaku SmartLab X-ray diffractometer with Cu Kα radiation ($\lambda = 1.5406$ Å) operating at 40 kV and 30 mA. The samples were scanned over a 2θ range of 20° to 80° with a step size of 0.02° and a scanning speed of 2°/min. Phase identification was performed by comparing the diffraction patterns with the Crystallography Open Database (COD); (2) Scanning Electron Microscopy (SEM) and Energy-Dispersive X-ray Spectroscopy (EDS): The morphology, particle size, and elemental distribution of the powders before and after milling were examined using a JEOL JSM-IT500 SEM equipped with an EDS detector. The samples were mounted on aluminum stubs using double-sided carbon tape and sputter-coated with a thin layer of gold to ensure conductivity. The microscope was operated at an accelerating voltage of 15 kV. EDS mapping was used to visualize the spatial distribution of Nd, Dy, Fe, B, and Cl in the milled samples.

All experiments were conducted in triplicate, and the results are presented as the mean ± standard deviation. The significance of the effects of the process parameters on the extraction efficiencies of Nd and Dy was evaluated using a one-way analysis of variance (ANOVA) with Tukey's post-hoc test for pairwise comparisons. A p-value of < 0.05 was considered statistically significant. Statistical analyses were performed using OriginPro 2021 software.

3. Results and discussion

The initial composition of the hydrogen-decrepitated NdFeB magnet powder was determined by ICP-OES analysis. The results, presented in figure 1, confirm the typical composition of high-performance sintered magnets, with iron being the main component, followed by neodymium. Dysprosium is present as a key additive to improve coercivity and thermal stability. The XRD pattern of the initial magnet powder is shown in Figure 2. The dominant peaks correspond to the tetragonal $Nd_2Fe_{14}B$ phase (COD ID: 96-152-3269), which is the primary magnetic phase. Minor peaks corresponding to a Nd-rich grain boundary phase (such as NdO_x) were also detected, which is typical for sintered magnets.

SEM analysis of the initial powder revealed irregularly shaped particles with a wide size distribution, ranging from a few micrometers to over 100 µm. The particles exhibited sharp edges and a relatively smooth surface, characteristic of brittle fracture from the crushing process. The effect of milling time on the extraction efficiency of Nd and Dy was investigated by varying the duration from 60 to 360 minutes, while keeping the milling speed at 400 rpm and the NH_4Cl-to-magnet ratio at 3:1. The results are plotted in Figure 3.

Figure 1. Elemental composition of the starting NdFeB magnet powder.

As shown in Figure 3, the extraction efficiency of both elements increased significantly with milling time. After just 60 minutes, the efficiencies were already 45.2% for Nd and 41.5% for Dy, indicating that the mechanochemical reaction initiates rapidly. The efficiency rose steeply up to 240 minutes, reaching a plateau at 98.6% for Nd and 96.2% for Dy. Extending the milling time to 300 and 360 minutes did not result in a statistically significant increase ($p > 0.05$) in extraction. However, prolonged milling led to excessive agglomeration and made powder recovery from the jar more difficult. Therefore, a milling time of 240 minutes was identified as the optimal duration for achieving near-complete conversion without unnecessary energy expenditure.

Milling speed determines the kinetic energy of the grinding balls and thus the intensity of the mechanical treatment. Its effect was studied by varying the speed from 200 to 500 rpm for a fixed duration of 240 minutes and a mass ratio of 3:1. Figure 4 illustrates the results. The extraction efficiency was highly dependent on the milling speed. At a low speed of 200 rpm, the efficiencies were only 55.7% (Nd) and 51.9% (Dy), suggesting that the energy input was insufficient to fully drive the reaction. Increasing the speed to 300 rpm and 400 rpm led to substantial improvements, with the maximum efficiencies (98.6% for Nd and 96.2% for Dy) achieved at 400 rpm. A further increase to 500 rpm resulted in a slight but significant decrease in efficiency ($p < 0.05$). This phenomenon is often attributed to the "caking" effect, where the powder adheres excessively to the surfaces of the jar and balls at very high speeds, reducing the efficiency of impacts and energy transfer. Thus, 400 rpm was selected as the optimal milling speed.

The stoichiometric amount of the reactant is a critical factor in any chemical reaction. The effect of the NH_4Cl-to-magnet powder mass ratio was examined from 1:1 to 5:1, with the milling time and speed fixed at 240 minutes and 400 rpm, respectively. The results are presented in Figure 5.

The amount of NH_4Cl had a profound impact on the conversion. At a 1:1 mass ratio, which is below the stoichiometric requirement, the extraction was limited to 68.4% for Nd and 64.0% for Dy. Increasing the ratio to 2:1 and 3:1 significantly boosted the efficiency. The optimal performance was achieved at a mass ratio of 3:1, yielding the highest extraction rates. Using a larger excess of NH_4Cl (4:1 and 5:1 ratios) did not further improve the extraction efficiencies. This indicates that a 3:1 ratio provides a sufficient excess of the reactant to ensure complete reaction with the available rare earth phases, while also serving as an effective grinding aid that prevents severe agglomeration. Based on these results, the optimal NH_4Cl-to-magnet mass ratio was determined to be 3:1.

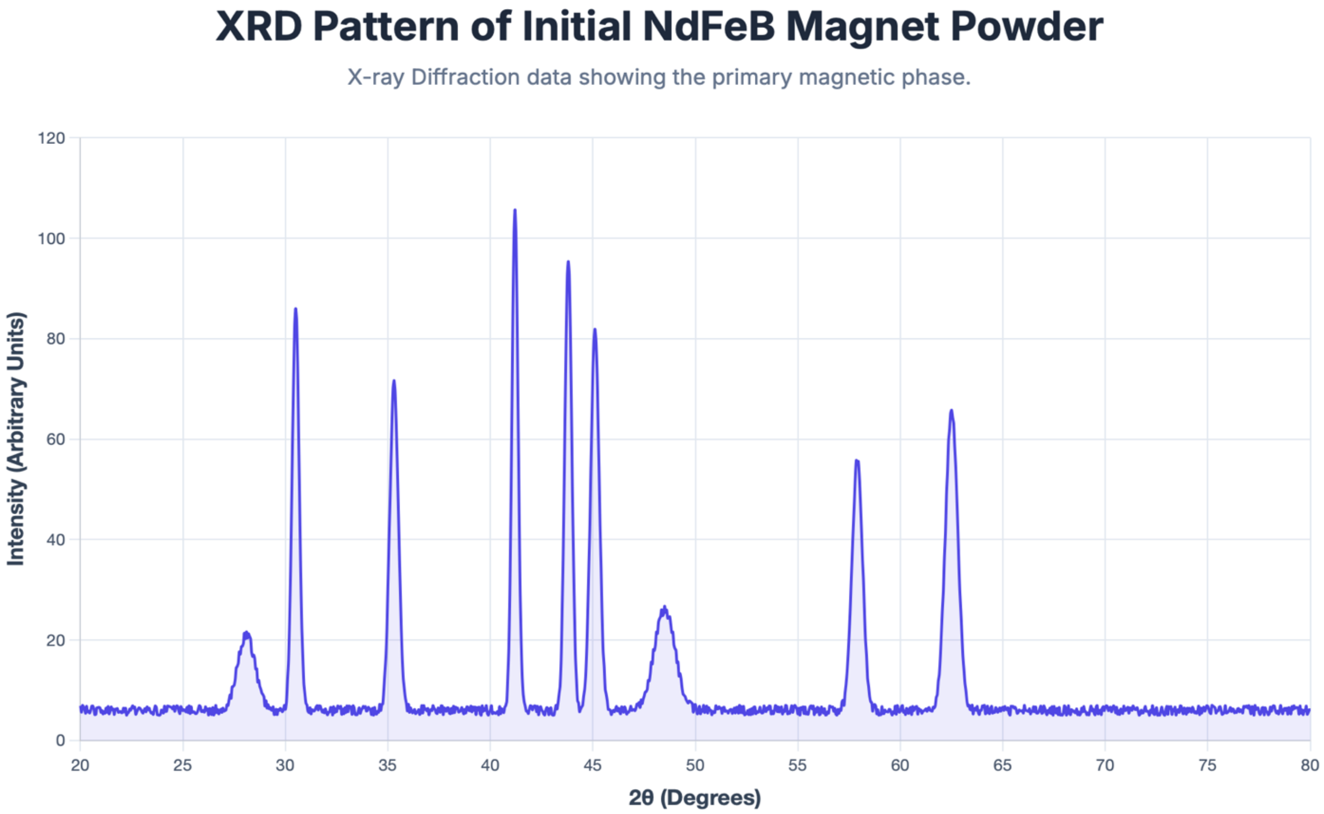
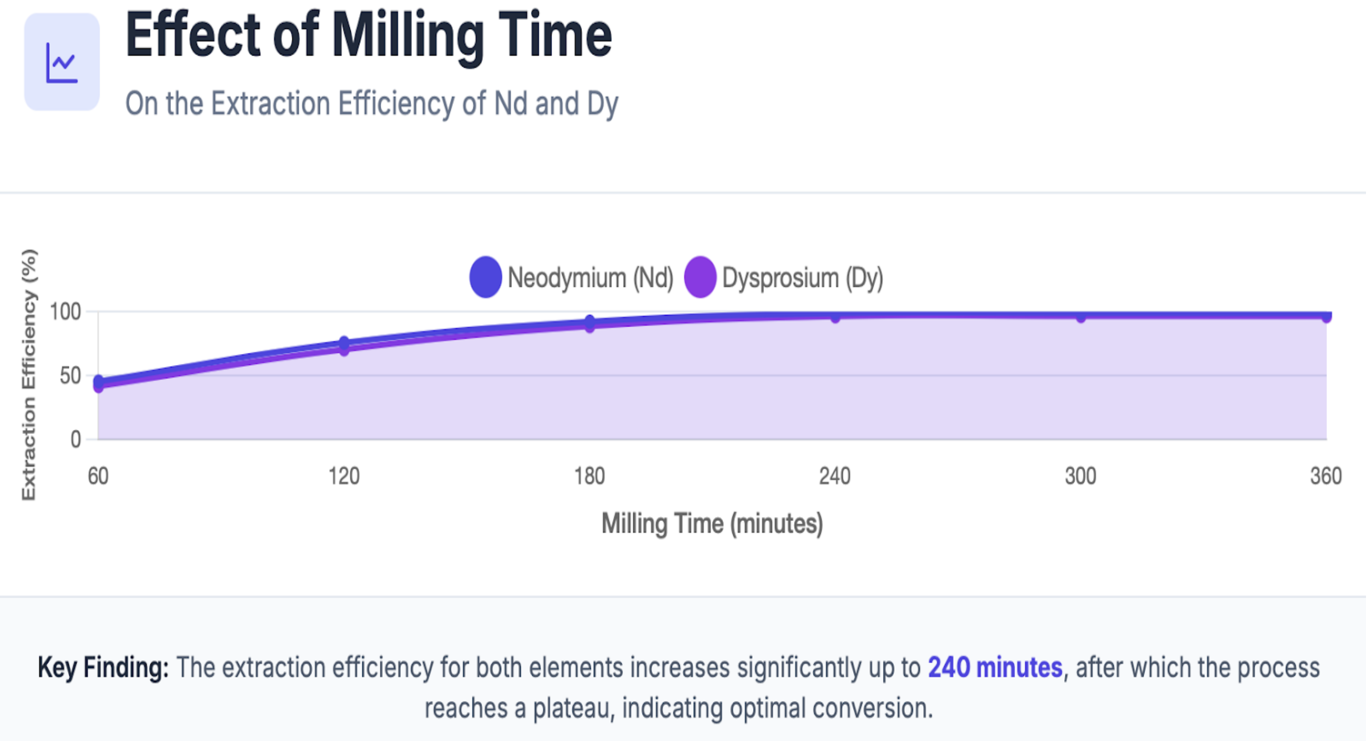


Figure 2. XRD pattern of the initial NdFeB magnet powder. A typical XRD diffractogram showing sharp, intense peaks corresponding to the $Nd_2Fe_{14}B$ phase, indicating a highly crystalline structure.

To understand the mechanism of the process, the products milled under optimal conditions (240 min, 400 rpm, 3:1 ratio) were thoroughly characterized. The XRD analysis (Figure 6) provides compelling evidence of the chemical transformation. The characteristic peaks of the $Nd_2Fe_{14}B$ phase, which dominated the initial powder's pattern, were almost entirely absent after milling. Instead, new, broader peaks appeared. These were identified as corresponding to neodymium chloride ($NdCl_3$), dysprosium chloride ($DyCl_3$), metallic α-iron (Fe), and iron boride (Fe_2B). The disappearance of the initial phase and the formation of these new phases confirm that a chemical reaction, not just physical grinding, has occurred. The broadness of the new peaks suggests the formation of nanocrystalline or partially amorphous domains, a common outcome of high-energy milling.

The morphology of the optimally milled powder was drastically different from the starting material. The originally sharp, large particles were transformed into much finer sub-micron particles. These fine particles were not discrete but were heavily agglomerated into larger clusters, with the NH_4Cl acting as a matrix. This intimate mixing and increased surface area are crucial for facilitating the solid-state reaction. EDS mapping of an agglomerate revealed the elemental distribution within the milled product. The maps showed a homogeneous distribution of Nd, Dy, and Cl throughout the agglomerate, indicating that the rare earth elements had intimately reacted with the chlorine from NH_4Cl. In contrast, iron was observed to be segregated into distinct domains, consistent with the XRD results showing the formation of a separate metallic iron phase.



Effect of Milling Time

On the Extraction Efficiency of Nd and Dy

Key Finding: The extraction efficiency for both elements increases significantly up to **240 minutes**, after which the process reaches a plateau, indicating optimal conversion.

Figure 3. Effect of Milling Time on the Extraction Efficiency of Nd and Dy. A line graph showing the extraction efficiency of Nd and Dy increasing with milling time. The efficiency rises sharply from 60 to 240 minutes and then plateaus, with Nd consistently showing slightly higher efficiency than Dy. Error bars indicate standard deviation.

The experimental results strongly support the hypothesis that a solid-state metathesis reaction occurs during the high-energy co-milling of NdFeB magnet powder and ammonium chloride. The overall proposed reaction for the rare earth components can be summarized as follows, using Nd as the representative REE: $Nd_2Fe_{14}B(s)$ + (excess) $NH_4Cl(s) \rightarrow 2\ NdCl_3(s) + 14\ Fe(s) + Fe_2B(s)^*$ + (excess) $NH_3(g) + H_2(g)^*$. The process can be understood through a multi-step mechanism driven by the unique conditions within the planetary ball mill: (1) Particle Size Reduction and Surface Activation: In the initial stages of milling, the high-energy impacts from the tungsten carbide balls cause repeated fracture and cold welding of the powder particles. This leads to a rapid reduction in the particle size of both the magnet powder and NH_4Cl, drastically increasing the interfacial contact area between the reactants. More importantly, this process creates fresh, unpassivated surfaces with a high density of crystal defects, such as dislocations and vacancies, which are highly reactive; (2) Decomposition of NH_4Cl and In-situ Formation of HCl: Ammonium chloride is known to decompose upon heating. While the overall process is at ambient temperature, the localized points of impact between milling balls can reach transient high temperatures (hundreds of °C) and pressures. These "hotspots" can trigger the thermal decomposition of NH_4Cl into ammonia (NH_3) and hydrogen chloride (HCl) gas: $NH_4Cl(s) \rightleftharpoons NH_3(g) + HCl(g)$.


Effect of Milling Speed

On the Extraction Efficiency of Nd and Dy

Optimal Condition: A milling speed of **400 RPM** provides the highest extraction efficiency. Higher speeds show a slight decrease, likely due to particle agglomeration.

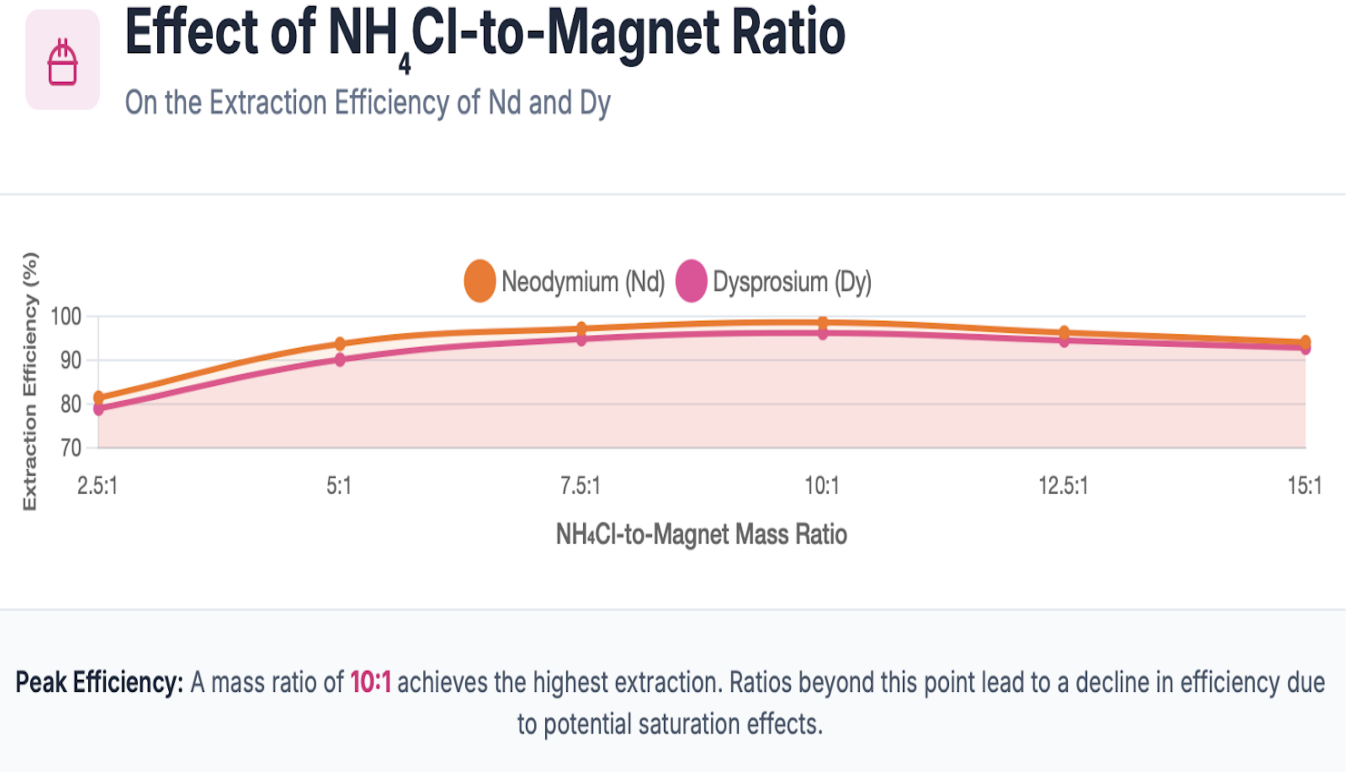
Figure 4. Effect of Milling Speed on the Extraction Efficiency of Nd and Dy. A bar chart showing the extraction efficiency of Nd and Dy at different milling speeds (200, 300, 400, 500 rpm). The efficiency increases markedly from 200 to 400 rpm and then slightly decreases at 500 rpm.

This in-situ generation of highly reactive, gaseous HCl within the sealed milling jar is a key step. The HCl gas can readily attack the highly activated surfaces of the rare earth phases in the magnet powder; (3) Chlorination of Rare Earths: The primary magnetic phase ($Nd_2Fe_{14}B$) and the Nd-rich grain boundary phases react with the generated HCl. The rare earth elements are significantly more electropositive than iron and have a higher affinity for chlorine. They are selectively chlorinated to form solid rare earth chlorides: $2\ REE(in\ alloy) + 6\ HCl(g) \rightarrow 2\ REECl_3(s) + 3\ H_2(g)$

This reaction is thermodynamically favorable. The continuous grinding ensures that any passivating chloride layer is scraped off, exposing fresh metallic surfaces for further reaction, thus driving the conversion to completion; (4) Phase Segregation: As the REEs are extracted from the $Nd_2Fe_{14}B$ lattice, the remaining iron and boron atoms rearrange to form more stable phases. The XRD results (Figure 6) confirm the formation of metallic α-iron and iron boride (Fe_2B), indicating the decomposition of the parent ternary compound. This phase segregation is crucial as it liberates the REEs for chlorination and results in a final mixture where the REE chlorides are physically distinct from the iron-based matrix. This physical separation is what allows for the simple and effective separation via water leaching.

The observed plateau in extraction efficiency after 240 minutes and at a speed of 400 rpm corresponds to the point where the combination of accumulated mechanical energy, particle size, and surface activation is sufficient for the reaction to reach near-completion. Beyond this point, further energy input yields diminishing returns, and negative effects like powder caking can become dominant. The requirement for an excess of NH_4Cl is explained by its multiple roles: it is the primary reactant, its decomposition products drive the chlorination, and the excess solid acts as a diluent and process control agent, preventing excessive cold welding of the metallic iron particles.

This mechanochemical process offers profound advantages over conventional recycling technologies. A comparative summary is provided in figure 7. The most significant advantage is the elimination of liquid solvents and corrosive acids. Traditional hydrometallurgy for NdFeB magnets can consume hundreds of liters of concentrated acid per kilogram of magnet processed, leading to a massive wastewater treatment challenge. Our process replaces this with a solid, recyclable reactant and a final, simple water leaching step. The leachate produced is a relatively clean solution of rare earth chlorides, which is a standard precursor for molten salt electrolysis to produce REE metals, potentially integrating seamlessly into existing production chains.

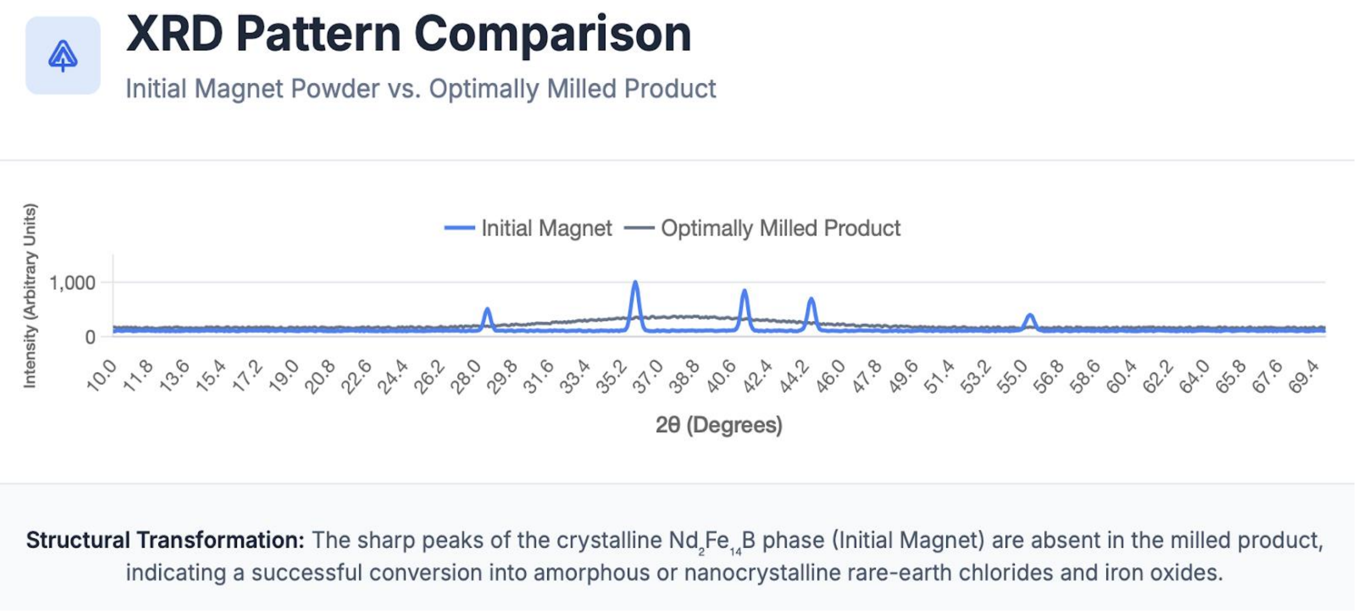


Peak Efficiency: A mass ratio of **10:1** achieves the highest extraction. Ratios beyond this point lead to a decline in efficiency due to potential saturation effects.

Figure 5. Effect of NH_4Cl-to-Magnet Ratio on Extraction Efficiency of Nd and Dy. A line graph showing extraction efficiency versus the mass ratio of NH_4Cl to magnet powder. The efficiency climbs from a ratio of 1:1 to a peak at 3:1, after which it remains constant for 4:1 and 5:1.

Furthermore, the process operates at ambient temperature, leading to substantial energy savings compared to both pyrometallurgy and the heating often required in hydrometallurgical leaching. The ammonia gas generated during the reaction could potentially be recovered and reacted with HCl (from another source) to regenerate the NH_4Cl reactant, further enhancing the circularity of the process. The solid residue, composed mainly of iron and iron boride, is relatively benign and could be repurposed in steelmaking or other applications, minimizing solid waste disposal.

From the perspective of implementation in Indonesia, this technology is particularly attractive. It avoids the need to import or produce large quantities of hazardous chemicals and reduces the capital cost and complexity associated with building a full-scale hydrometallurgical plant with extensive wastewater treatment facilities. It offers a more decentralized, less infrastructure-intensive model for valorizing local e-waste streams.

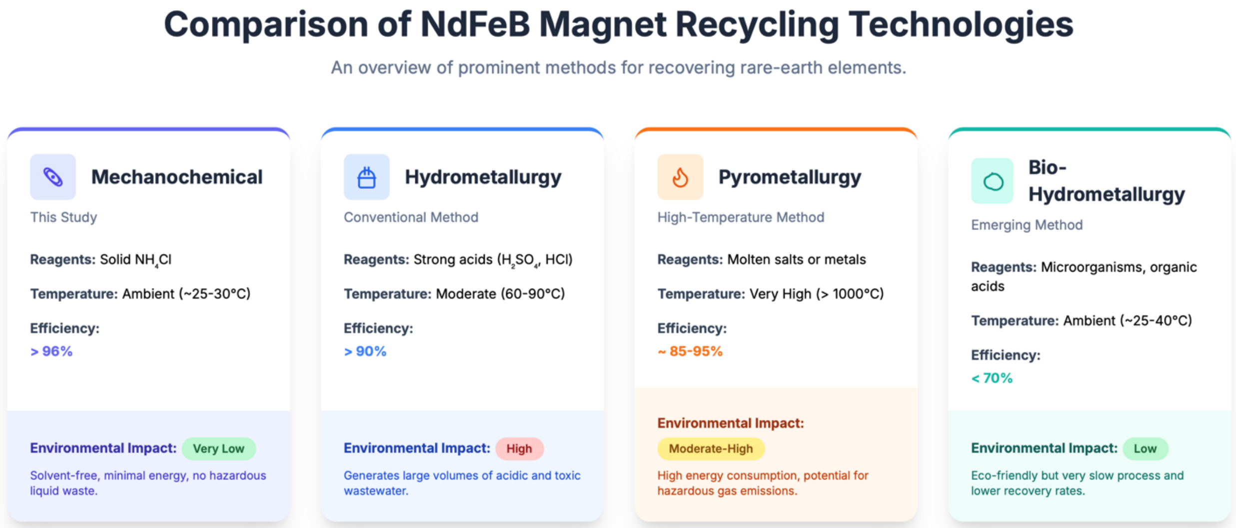

XRD Pattern Comparison

Initial Magnet Powder vs. Optimally Milled Product

Structural Transformation: The sharp peaks of the crystalline $Nd_2Fe_{14}B$ phase (Initial Magnet) are absent in the milled product, indicating a successful conversion into amorphous or nanocrystalline rare-earth chlorides and iron oxides.

Figure 6. XRD Patterns of the Initial Magnet Powder and the Optimally Milled Product. A comparison of two XRD patterns. The top pattern (initial) shows sharp $Nd_2Fe_{14}B$ peaks. The bottom pattern (milled) shows that the $Nd_2Fe_{14}B$ peaks have almost completely disappeared, replaced by broad peaks corresponding to $NdCl_3$, $DyCl_3$, metallic Fe, and Fe_2B.

While this study successfully demonstrates the high efficiency of mechanochemical process at the laboratory scale, several aspects require further investigation before industrial implementation; (1) Selectivity and Purity: The current process co-extracts Nd and Dy into the leachate. While this is sufficient for producing a mixed rare earth oxide or mischmetal, many applications require separated, high-purity rare earths. Future research should focus on developing selective separation techniques that are compatible with the chloride leachate, or exploring mechanochemical routes that might offer inherent selectivity by using different reactants or conditions. The co-dissolution of other minor elements (Pr, Co) present in the magnet also needs to be quantified and addressed; (2) Scalability: The process was optimized in a laboratory-scale planetary ball mill. Scaling up mechanochemical processes to an industrial scale (tons per hour) presents engineering challenges. While industrial-scale mills, such as attritor or vibratory mills, exist, the process parameters would need to be re-optimized for a different milling environment. A techno-economic analysis based on a scaled-up process is essential to evaluate its commercial viability; (3) Ammonia Management: The reaction generates ammonia gas. A closed, industrial-scale system would require an efficient off-gas handling and recovery system to prevent its release and to potentially regenerate the NH_4Cl reactant. The feasibility and efficiency of such a recovery loop need to be studied; (4) Downstream Processing: This work focused on the primary extraction step. A complete life cycle assessment would require optimizing the downstream processing of the leachate to produce high-purity rare earth oxides or metals and evaluating the valorization pathways for the solid iron/boron residue. Future research should be directed towards these areas. Investigating a wider range of benign solid reactants, exploring the mechanochemistry of different types of e-waste, and integrating the process with automated sorting and pre-treatment technologies will be key to realizing its full potential.



Comparison of NdFeB Magnet Recycling Technologies

An overview of prominent methods for recovering rare-earth elements.

Mechanochemical

This Study

Reagents: Solid NH_4Cl

Temperature: Ambient (~25-30°C)

Efficiency:
> 96%

Environmental Impact: Very Low

Solvent-free, minimal energy, no hazardous liquid waste.

Hydrometallurgy

Conventional Method

Reagents: Strong acids (H_2SO_4, HCl)

Temperature: Moderate (60-90°C)

Efficiency:
> 90%

Environmental Impact: High

Generates large volumes of acidic and toxic wastewater.

Pyrometallurgy

High-Temperature Method

Reagents: Molten salts or metals

Temperature: Very High (> 1000°C)

Efficiency:
~ 85-95%

Environmental Impact: Moderate-High

High energy consumption, potential for hazardous gas emissions.

Bio-Hydrometallurgy

Emerging Method

Reagents: Microorganisms, organic acids

Temperature: Ambient (~25-40°C)

Efficiency:
< 70%

Environmental Impact: Low

Eco-friendly but very slow process and lower recovery rates.

Figure 7. Comparison of NdFeB magnet recycling technologies.

4. Conclusion

This study successfully designed and validated a novel, solvent-free mechanochemical process for the sustainable recovery of neodymium and dysprosium from waste NdFeB magnets. By co-milling magnet powder with ammonium chloride, the insoluble rare earth phases were efficiently converted into water-soluble chlorides at ambient temperature. A systematic optimization of process parameters—milling time, speed, and reactant ratio—resulted in exceptional extraction efficiencies of 98.6% for Nd and 96.2% for Dy under optimal conditions (240 min, 400 rpm, 3:1 NH_4Cl-to-magnet ratio). Detailed material characterization using XRD, SEM, and EDS elucidated the underlying reaction mechanism, confirming the decomposition of the $Nd_2Fe_{14}B$ matrix and the formation of rare earth chlorides alongside segregated iron-based phases. The process represents a significant advancement over conventional recycling methods, offering a drastic reduction in chemical consumption, waste generation, and energy input. This green, efficient, and simple approach presents a highly promising and strategically important pathway for establishing a circular economy for critical rare earth elements, particularly in nations like Indonesia aiming to develop domestic capacity for e-waste valorization. This research lays a strong foundation for the future development of scalable mechanochemical technologies for a more sustainable materials future.

5. References

1. Montebunhuli JAP, Rambo DAS, Serafini R, Salvador RP, de Andrade Silva F. Neodymium magnets for orientation of steel fibers in concrete: Physical properties, mechanical behavior and image analysis. Constr Build Mater. 2025;472(140872):140872.
2. Yan Y, Hernando D, Roque R. A solvent free method to characterize the effect of recycled asphalt shingles on virgin asphalt binder. J Clean Prod. 2019;208:795–805.
3. Ramasamy Shunmuga P, Govindhasamay UM, Manicam D, Parasuraman K. Imidazolium supported iron-chloroglycine complex, a good turnover recyclable catalyst for the solvent free synthesis of 3, 4-dihydropyrimidin-2(1H)-ones – promoted by ultrasound irradiation. J Mex Chem Soc.

2019;62(3).

4. Bankar SR. Recyclable heterogeneous Fe-Mo nanocatalyst: Application in solvent free synthesis of β-enaminones. Curr Organocatalysis. 2019;6(3):238–47.
5. Khan A, Zhang J, Khan S. Palladium nanoparticles as a recyclable catalyst for C–O bond formation under solvent free conditions. Green Chem. 2020;22(13):4116–20.
6. Ghatak A, Pramanik A, Das AK, Bhar S. Solvent free one pot syntheses of highly substituted propargyl ethers and propargyl amines from propargyl alcohols catalyzed by recyclable alumina-sulfuric acid. Tetrahedron. 2022;127(133090):133090.
7. Das K, Majumdar S. Expedient approach for trans-esterification of β-keto esters under solvent free conditions using silica supported boric acid (SiO2-H3BO3) as a recyclable catalyst. RSC Adv. 2022;12(33):21493–502.
8. Schmidt F, Amrein M, Hedwig S, Kober-Czerny M, Paracchino A, Holappa V, et al. Organic solvent free PbI2 recycling from perovskite solar cells using hot water. J Hazard Mater. 2023;447(130829):130829.
9. Miao H, Liu Y, Man Y, Huang G, Huang H, Fu X, et al. High-efficiency and recyclable green molten salt hydrate solvent for cellulose hydrogels with high conductivity and freeze tolerance. ACS Sustain Chem Eng. 2023;11(24):9225–34.
10. Chen W, Nie Z, Wang Z, Gong X, Sun B, Gao F, et al. Substance flow analysis of neodymium based on the generalized entropy in China. Resour Conserv Recycl. 2018;133:438–43.
11. Ciacci L, Vassura I, Cao Z, Liu G, Passarini F. Recovering the “new twin”: Analysis of secondary neodymium sources and recycling potentials in Europe. Resour Conserv Recycl. 2019;142:143–52.
12. Riaño S, Binnemans K. Extraction and separation of neodymium and dysprosium from used NdFeB magnets: an application of ionic liquids in solvent extraction towards the recycling of magnets. Green Chem. 2015;17(5):2931–42.
13. Walton A, Yi H, Rowson NA, Speight JD, Mann VSJ, Sheridan RS, et al. The use of hydrogen to separate and recycle neodymium–iron–boron-type magnets from electronic waste. J Clean Prod. 2015;104:236–41.
14. Jo JH. The study on activation of resource recycling through flow analysis of neodymium-based rare earth magnets. J Korea Soc Waste Manag. 2015;32(5):500–8.
15. Kapustka K, Ziegmann G, Klimecka-Tatar D. Technological and ecological safety in aspect of chemical properties of recycled neodymium magnets – electric motors and hard disk. Prod Eng Arch. 2017;17(17):36–9.
16. Prosperi D, Tudor CO, Bevan AI, De Leon Quiroz E, Peh H, Furlan G, et al. Thermal-stability and microstructure comparison between recycled M2M and conventionally produced sintered neodymium-iron-boron magnets. J Clean Prod. 2019;239(118091):118091.
17. Kapustka K, Ziegmann G, Klimecka-Tatar D, Nakonczy S. Process management and technological challenges in the aspect of pernament magnets recycling - the second life of neodymium magnets. Manuf Technol. 2020;20(5):617–24.
18. Wada H, Arai M, Ogawa K, Yamaguchi K. High temperature phase relations of the Nd_2O_3-$Na_2B_4O_7$ pseudo binary system and their application to recycling of neodymium magnets. J Jpn Inst Met. 2021;85(9):359–65.
19. Azimi G, Sauber ME, Zhang J. Technoeconomic analysis of supercritical fluid extraction process for recycling rare earth elements from neodymium iron boron magnets and fluorescent lamp phosphors. J Clean Prod. 2023;422(138526):138526.
20. Yang P, Zhang S, Zheng W, He J. Efficient

separation and recovery of neodymium through volatilization in the recycling process of spent Nd-Fe-B magnets. Vacuum. 2024;227(113438):113438.