

## Article

# Recovery of Rare Earth Elements Present in Mobile Phone Magnets with the Use of Organic Acids

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**Abstract:** Currently, the recovery of materials from secondary sources is increasingly necessary because of the scarcity of materials. Significant amounts of rare earth elements (REE) are found in permanent neodymium-iron-boron (NdFeB) magnets, used in various electrical and electronic equipments, such as mobile phones. However, the estimated recycling rate for REEs is only 1%. Hydrometallurgical routes are the most commonly used for REE recovery from secondary sources. This route usually uses inorganic acids, which are expensive and toxic. Thus, in this work the leaching efficiency of organic acids (acetic and citric) in leaching the REE (neodymium and praseodymium) present in magnets of obsolete or defective mobile phones was evaluated. Different concentrations of acids, solid/liquid relations, times and leaching techniques (microwave, ultrasound and conventional) are also evaluated. The results indicate that acetic and citric acids have the potential to leach Nd and Pr. Microwave leaching was the most effective method, compared to ultrasound and conventional methods. In microwaves, citric acid at 0.5 M (ratio s/l 1/100) leached 57% of Nd and 58% of Pr. Acetic acid at 0.5 M (s/l ratio—1/100) leached 48% of Nd and 65% of Pr, in 15 min. Furthermore, both citric acid and acetic acid also leached high percentages of iron (51% and 72%, respectively).

**Keywords:** rare earths; NdFeB magnets; organic acids; mobile phones; recovery



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## 1. Introduction

Rare earth elements (REE) are essential for the development of modern industry, especially in the applications of permanent magnets [1]. REE are used in the electrical and electronics industry, the development of clean technologies (hybrid vehicles, wind and solar energies), oil cracking, communications, and defense applications, among others [2–4]. The global demand for these elements has increased exponentially in the last decades [5–7]. However, the availability of REE has been decreasing each year [8].

The REE production process is extremely complex and expensive. According to Jordens et al. (2013), the recovery rates of these elements in ores are very low, 10% on average. This situation is aggravated by the low concentration of oxides in the minerals extracted (approximately 6%), which results in a net recovery rate of 0.6% [9]. In addition, REE extraction consumes high volumes of resources (energy, water, chemical reagents) and generates a large quantity of waste, that can be toxic and cause damage to the ecosystem [10–13].

REE are considered to be critical raw materials for the development of new technologies, by research institutes and government agencies in the United States and Europe [14,15]. In addition to their technological and economic importance, another factor for criticality is the high risk of supply disruption for these metals [16]. Currently, China provides approximately 85% the world's REE. Therefore, increased research into developing replacements for REE, or reducing the amount of material needed, is required, in addition to improving recycling processes.

In 2020, the global production of REE was 240,000 tons, of which about 23% was used in the production of neodymium-iron-boron (NdFeB) permanent magnets [17]. The estimated global market for these magnets in 2020 was 190,000 tons, with projections of reaching 450,000 tons by 2030 [18,19]. Currently, these magnets are responsible for 76% and 100% of neodymium and dysprosium demands, respectively [20,21]. They are used in computer hard disk drives (HDD), mobile phones, medical equipment (mainly magnetic resonance), power hybrids and electric vehicles, wind turbines, loudspeakers, robots, factory automation equipment, and aircraft controls, etc. [4].

NdFeB magnets contain about 30% of REE. Neodymium (Nd) has a superior magnetization of iron, making it possible to use it in the manufacture of smaller and stronger magnets [22]. Elements such as dysprosium (Dy) and terbium (Tb), known as heavy rare earths (HRE), are conventionally added to NdFeB magnets in small amounts, to increase the coercivity and high temperature stability [22–24]. On the other hand, neodymium and praseodymium (Pr) are elements that are so chemically similar, they are difficult to separate and they have comparable hard magnetic properties [25]. Furthermore, the presence of praseodymium does not affect the quality of the final product. Therefore, in NdFeB magnets the praseodymium amount can reach 20–30% of the neodymium amount [24,26,27].

These magnets are considered to be essential for modern mobile phone devices. They are used in speakers, receivers, vibration mode motors, notification feedback motors, and camera auto-focus mechanisms [27]. Currently, 67% of the world's population makes use of some type of mobile phone device, which corresponds to 5.3 billion people. Estimates indicate that by 2025, the percentage of the population that makes use of this kind of device may reach 70% [28].

However, the development of current technological makes products such as mobile phones obsolete very quickly. The time a person remains with the same device depends on factors such as: purchasing power, cultural behavior and product sales strategies. Estimates indicate that each user remains with the same mobile phone device for 2 years on average [29,30]. In 2019, the volume of waste of electrical and electronic equipment (WEEE), including mobile phone devices, discarded in the world reached 53.3 million tons. Projections indicate that this volume will reach 74.6 million tons by 2030 [31]. Due to the high metal content found in some WEEE, these materials are called 'urban mines' [32].

For many years, the processes and recycling techniques of WEEE were concentrated on the recovery of precious metals (gold, silver, palladium) or valuable metals present in large volumes, such as copper [33,34]. However, the increase in demand for REE for high-tech industry and the potential risk of supply disruption for these metals has stimulated the mineral industries to seek new sources of supply, prospect new deposits and identify possible recycling alternatives for these elements [35]. In this context, printed circuit boards (PCB), scrap magnets (such as those from mobile phones) and other WEEE components, arise as alternative secondary sources of REE because they contain significant levels of these elements [36].

Hydrometallurgical routes are those most commonly used for REE recovery from secondary sources. For the leaching of NdFeB magnets inorganic acids normally are used, such as nitric acid (HNO<sub>3</sub>), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), hydrochloric acid (HCl) and sodium hydroxide (NaOH) [8,37–41]. Rabatho et al. (2013), for example, used a solution containing nitric acid (HNO<sub>3</sub>) and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) to dissolve neodymium (Nd) and dysprosium (Dy) from a magnetic residue sludge generated in the manufacturing process of NdFeB(Dy) magnets. The authors dissolved up to 98% Nd and 81% Dy, keeping the Fe dissolution below 15%. Subsequently, they used oxalic acid (H<sub>2</sub>C<sub>2</sub>O<sub>4</sub>) to precipitate these elements, recovered 91.5% of Nd and 81.8% of Dy from the solution [42]. Ni'am et al. (2020), compared the effectiveness of nitric, sulfuric and hydrochloric acid in removing REEs (neodymium, praseodymium and dysprosium) from NdFeB permanent magnets. The authors concluded that nitric acid was more efficient, removing 58.62% of Nd, 98.46% of Dy and 85.59% of Pr [40].

Studies using organic acids (acetic, citrus, oxalic, ascorbic and formic) in REE leaching are more recent. These acids can be considered promising and efficient, minimizing emissions caused by the use of mineral inorganic acids [39,43–46]. Behera and Parhi (2016), studied the efficiency of acetic acid for the leaching of NdFeB magnets, obtaining 99.9% of Nd and Fe extraction after 240 min [36]. Lai et al. (2018), for example, used magnesium sulfate and ascorbic acid to leach the REE present in ores, obtaining an efficiency of up to 85.7% [43]. Behera et al. (2019), studied the use of organic reagents (ascorbic, citric, acetic, formic and tartaric acid) in the dissolution of neodymium (Nd) from WEEE magnets. The authors concluded that acetic acid was more effective than other organic acids [44]. Oxalic acid has usually been used to precipitate REEs dissolved by other acids. Makarova et al. (2020), studied the electrochemical leaching of Fe and rare earths from NdFeB magnets, using sulfuric acid as a leaching agent and oxalic acid to precipitate the REE. The authors concluded that the addition of oxalic acid reduces energy consumption and, at the same time, improves the recovery of solid oxalates from magnets [45]. Liu et al. (2020), studied the leaching of NdFeB residues using oxalic acid to precipitate REEs, economically and environmentally improving the process. The authors obtained rare earth oxalate precipitation rates of 93.17% [46].

However, more studies are still needed to analyse the effectiveness of organic acids in leaching REE from secondary sources. Thus, the present work seeks to elucidate some gaps present in the literature, analyzing the efficiency of citric and acetic acids in the recovery of REE from NdFeB magnets obtained from obsolete mobile phones without roasting or oxidative roasting. In this study several factors such as concentration, s/l ratio and time are evaluated. Furthermore, the efficiencies of the conventional leaching process with the lixiviation assisted by ultra-sound and microwave, in the leaching of these magnets with organic acids also were compared.

## 2. Materials and Methods

In this study, NdFeB magnets from obsolete or defective mobile phones were used, from several brands, models and years of manufacture.

Initially, 239 mobile phone devices were randomly selected and divided into six categories (according to the year of manufacture). These devices, and the magnets contained within them were weighed, so that the relationship between the masses of the NdFeB magnets contained in the devices and the mass of the same devices could be established.

The magnets were then subjected to processes of demagnetization, grinding (without previous roasting or oxidative roasting), acid digestion and chemical characterization. For the grinding of the NdFeB magnets, a pestle and mortar were used (Figure 1) and the material was ground to a particle size that would allow it to pass through a 325-mesh sieve.



**Figure 1.** Grinding of NdFeB magnets from mobile phone devices with a mortar.

The digestion of the samples followed methodology 3051A USEPA, using microwave-assisted nitric acid ( $\text{HNO}_3$ , P.A) [47]. The samples were analyzed by plasma optical emission spectroscopy (ICP-OES), using equipment 5110, from Agilent Technologies. For this study, only the concentrations of the metals of greatest interest were determined, in this case iron, boron, neodymium, praseodymium, dysprosium and terbium. Other elements used in

the anti-corrosion coating layer of magnets, such as nickel, aluminum and copper, were not determined.

After characterization, the NdFeB magnets collected from other mobile phones were subjected to demagnetization and grinding processes, to proceed with the studies aimed at verifying the efficiency of organic acids in the leaching of REE.

### 2.1. Leaching NdFeB Magnets with Organic Acids

In the first stage of the work, organic acids were studied through the use of the microwave-assisted leaching technique. Subsequently, considering that microwave-assisted leaching can be costly and difficult to apply on an industrial scale, the same acids were studied using ultrasound-assisted and conventional leaching techniques. All experiments were performed in triplicate.

Thus, in this work the following variables were studied:

- Organic acids: acetic acid ( $\text{CH}_3\text{COOH}$ ) and citric acid ( $\text{C}_6\text{H}_8\text{O}_7$ );
- Acid concentrations: 0.25, 0.50 and 1.0 mol/L;
- Solid/liquid ratio: 1/100, 1/50 and 1/10;
- Leaching time;
- Leaching technique: microwave-assisted, ultrasound-assisted, and conventional leaching.

The solutions obtained in the leaching were filtered in filter paper and brought to a constant volume with Milli-Q<sup>®</sup> water. Aliquots of these solutions were removed for analysis by ICP-OES (5110, Agilent Technologies, Santa Clara, CA, USA). For this study, concentrations of iron, neodymium and praseodymium (elements that are relatively abundant in these magnets) were determined. In total, 621 leachings were carried out, using 1.2 kg of crushed NdFeB magnets from mobile phones.

Leaching rates were calculated according to the equation below:

$$E (\%) = (w_{1,i} / w_{i,i}) \times 100$$

where,  $E$  is the efficiency (%),  $w_{1,i}$  is the amount (mg/kg) after the leaching process for rare earth  $i$ ; and  $w_{i,i}$  is the digested sample amount (mg/kg) for rare earth  $i$ .

#### 2.1.1. Microwave-Assisted Leaching

The leaching tests were performed in microwaves (Multiwave, Anton Paar, São Paulo, SP, Brazil), using a methodology similar to the conditions described in methodology 3051A USEPA (a 0.5 g sample, digested at a temperature of 175 °C and pressure of 20 bar) with organic acids replacing nitric acid. Variations in the leaching times of 15, 30 and 60 min were studied.

#### 2.1.2. Ultrasound-Assisted Leaching and Conventional Leaching

In ultrasound-assisted leaching a 2.8 L, piece of Quimis equipment (Model Q335D), with a frequency 50/60 Hz, was used. Conventional leaching was performed in a 150 mL glass, under stirring with an electric motor (Fisatom, 25 W power), without heating. The stirring speed was set at 30 rpm.

Considering that these techniques are less 'aggressive' when compared to microwaves, in this stage, variations of leaching with longer times (30, 60, and 120 min) were tested. All tests were performed at room temperature.

### 3. Results and Discussion

#### 3.1. NdFeB Magnets/Mobile Phones Ratio

Initially, the masses of randomly selected mobile phones and their magnets were measured. As can be observed (Table 1), in terms of average weight, the ratio of the NdFeB magnet in relation to the total mass of mobile phone device increased from 0.47% to 1.13% of mobile phone mass.

**Table 1.** Ratio mass of NdFeB magnets/mass of mobile phones over the years, as a percentage.

Year	% (Average)
1996–2000	0.47
2001–2005	0.64
2006–2010	1.08
2011–2015	1.07
2016–2018	1.11
2019–2020	1.13

Although the size of NdFeB magnets is getting smaller and smaller [26], the increase in the mass proportion of these magnets is explained by the amount of magnets used per device. Mobile phones have evolved from models containing only one of these magnets (in the speaker), until the current models, containing 14 small NdFeB magnets [27]. In general, it can be said that the models manufactured before 2011 were largely made up of three NdFeB magnets, located in the central speaker, the headphone input and the vibration mechanism. Those manufactured after 2011 are made up of four or more magnets, located in the central speaker, the headphone input, the vibration mechanism and the photographic cameras. Furthermore, in the most modern devices, each of these mentioned parts may contain two or more magnets. The increase in the quantity of magnets is related to the entry of smartphones on the market; devices with better technology and better media quality.

The increases in the number of permanent magnets used in more modern mobile phones, and the consequent increase in the mass of the rare earth elements used in these devices, make these materials increasingly attractive for recycling, in order to recover these elements.

#### 3.2. Demagnetisation and Grinding of NdFeB Magnets

The thermal demagnetization process consists of heating the magnets above their Curie temperature (312 °C in this case), to distort the parallel alignment of the magnetic moments, causing a complete and definitive loss of its magnetism, until it is remagnetized again [48]. Thus, the magnets removed from mobile phones were demagnetized in an electric furnace (brand Sanchis, Porto Alegre, Brazil), at a temperature of 320 °C for 60 min. During the demagnetization process, an approximate mass loss of 3.40% was observed. According to Schultz (1999), it is common to have a loss of mass associated with this step. This mass loss can be associated with the pulverization of oxidized surface components from magnets [49].

In this study, a pestle and a mortar were used for manually grinding the NdFeB magnets. Initially, a knife mill was tested for grinding the magnets taken from mobile phone devices. However, the knife mill generated high material loss (over 30%) and low grinding efficiency. This probably occurred due to the small size of the magnets (mainly the magnets used in the cameras of the devices). By manual grinding the material loss was only 4%.

### 3.3. Chemical Composition of NdFeB Magnets Used in Mobile Phones

Initially, NdFeB magnets were characterized separately by year (period) of mobile phone manufacture, as previously categorized. The results obtained (Figure 2) show a reduction in the quantity of dysprosium used (from 1.9% to 0.7%) and an increase in the quantity of neodymium and praseodymium (from 22.0% to 28.3% and 2.9% to 4.7%, respectively), over the years.

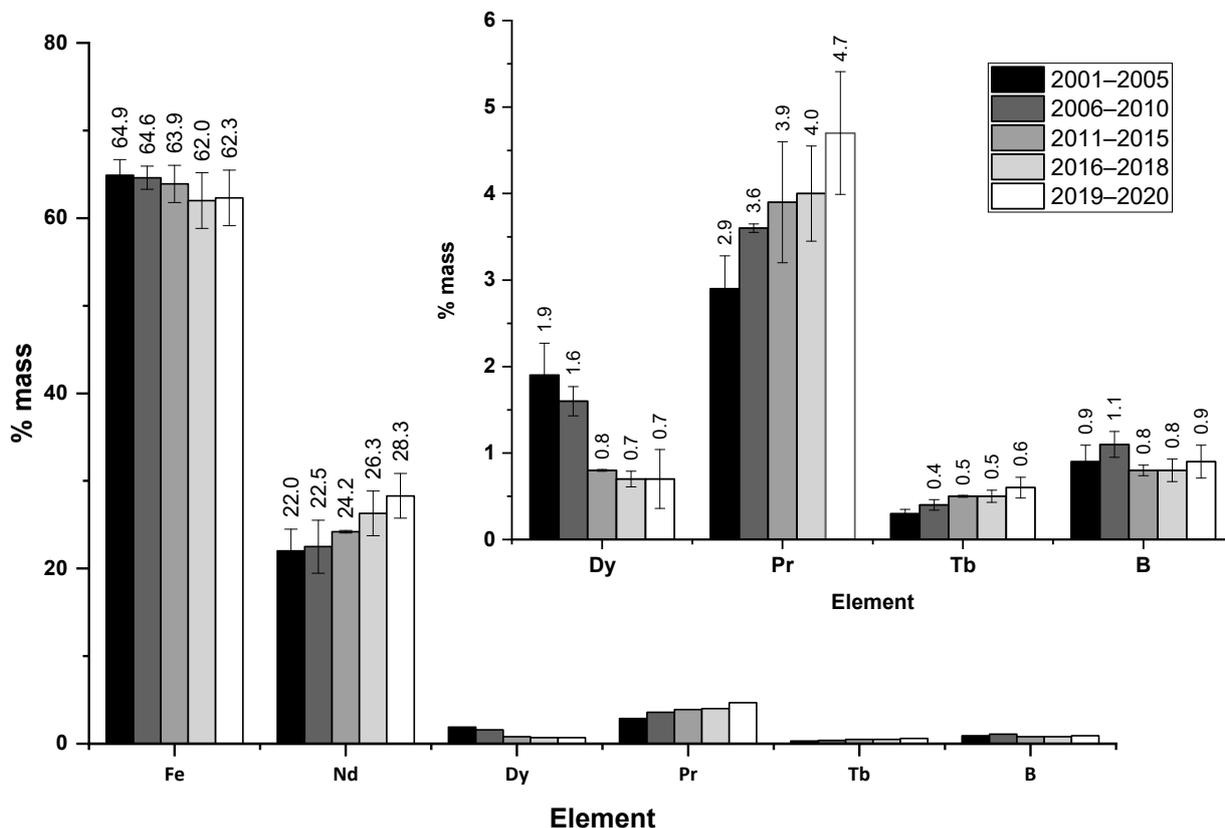


Figure 2. Chemical composition (average) of magnets, over time.

As previously mentioned, practically 100% of Dy production is used in the manufacture of NdFeB magnets, making the product scarce and extremely expensive. Thus, new technologies were created with the aim of reducing the amounts of this element necessary for the manufacture of these magnets [20]. One of these technologies was the Grain Boundary Diffusion process (GBDP). By this process, HRE elements (Dy and Tb) are deposited only at the grain boundary, significantly reducing the HRE amount required to achieve the desired coercivities, with the remanence remaining almost unchanged. This not only generates a performance increase, but also uses less dysprosium, making them more price stable than magnets produced by the traditional method [50]. The use of this process, and other technological advances, can explain the reduction in dysprosium content, one element that had its price highly impacted by Chinese restrictions.

The increase in Neodymium content is linked to the need for miniaturization of NdFeB magnets for use in mobile phones, considering that larger amounts of neodymium allow the increase of coercivity and magnetic fields in small magnets [51]. Praseodymium (Pr) is usually added to replace neodymium (usually in a 1:5 ratio), at a lower cost and without loss of performance [52]. Furthermore, as mentioned earlier, neodymium and praseodymium are so difficult to separate that they are used in NdFeB magnets in the form of a Pr/Nd alloy. Thus, the variation in the contents of the two elements is closely linked.

After this first characterization, a second characterization step was performed. In this step, the average amounts of the elements contained in the magnets of mobile phones

were considered, without considering their manufacturing periods. The results obtained in this analysis show (Table 2) that the analyzed REE (Nd, Pr and Dy) represent 28.8% of the sample mass (on average), while Fe represents an average of 61.9% of the mass of the samples.

**Table 2.** Average chemical composition of NdFeB magnets used in mobile phones.

Element	Content %
Fe	61.9%
Nd	23.9%
Pr	3.8 %
Dy	1.1%
others	9.3%

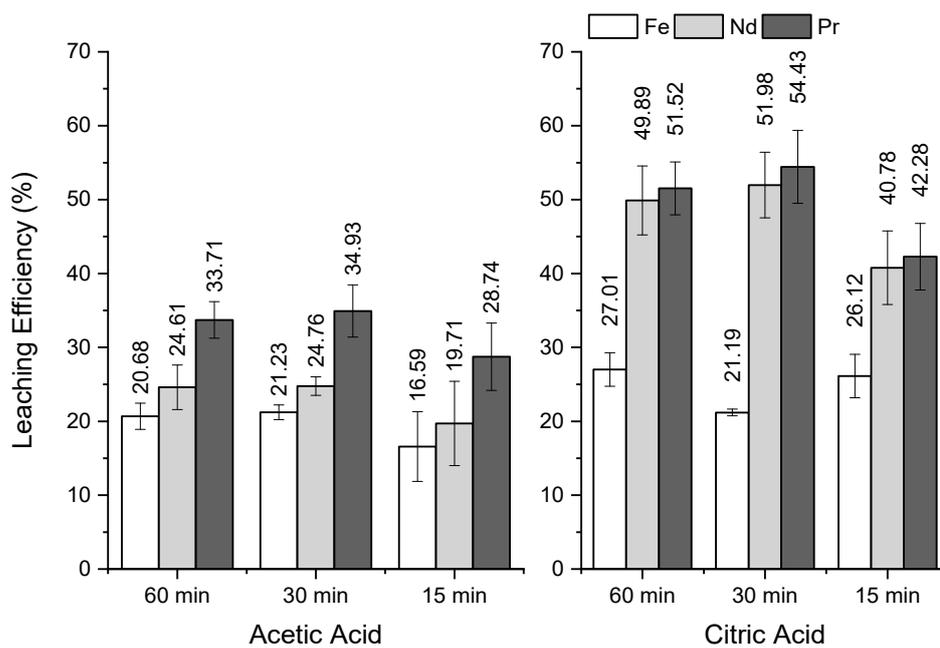
The values presented in Table 2 were used as a reference in the leaching of the REE with the aid of organic acids. The REE contents found in the characterization is in line with other studies which also analyzed the feasibility of recycling NdFeB magnets [53–57]. It is important to point out that the exact composition of the NdFeB magnets depends on the grade used and the application for which it is intended. Therefore, variations in composition are frequent [54,58].

The characterization of NdFeB magnets from mobile phones indicated the presence of REE at higher levels than those found in ores [58], which makes their recycling very attractive, both financially and environmentally.

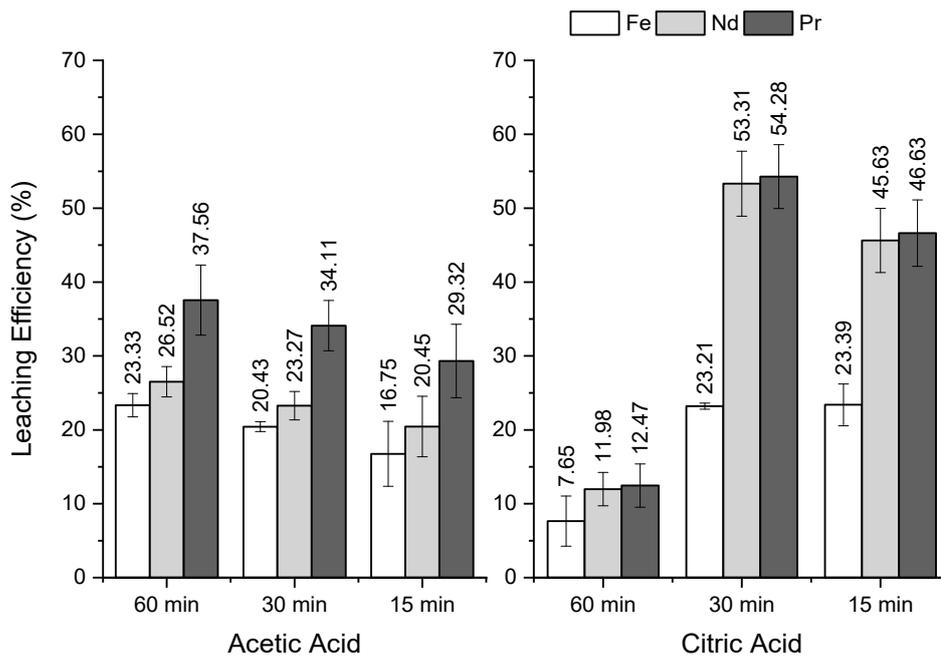
### 3.4. Leaching of REE with Organic Acids

#### 3.4.1. Microwave Leaching

The results obtained in the microwave leaching tests showed that citric acid was more efficient than acetic acid at a concentration of 1 M. Already acetic acid proved to be more efficient at concentrations of 0.50 M and 0.25 M. In addition, it is possible to see that for solid/liquid ratio 1/50 and 1/100 the results do not show significant variations, while in the solid/liquid ratio 1/10 the leaching efficiency decreases, a fact also observed by Behera et al. (2019) [44]. Regarding the leaching times, it is observed that in the case of acetic acid the variations were not significant. For citric acid, in some cases, it was possible to observe a reduction in efficiency with increasing the time to 60 min, an unexpected fact that needs further investigation. For 1 M acetic acid (Figure 3), the best results were obtained in the solid/liquid ratio 1/50, in 60 min, where the leaching efficiency was 26% for Nd, 37% for Pr and 23% for iron. For the solid/liquid ratio 1/10, in 15 min, the leaching efficiency was 27% for Nd and 37% for Pr, however, the percentage of leached iron was greater than 37%, which can make it difficult to recover Nd and Pr. In the case of 1 M citric acid, very similar results were obtained for the solid/liquid ratios 1/50 and 1/100, in 30 min, with a leaching efficiency close to 53% for Nd and 54% for Pr, maintaining the iron leaching close to 22%. At this concentration, the leaching efficiency of REE metals in the solid/liquid ratio 1/10 was below 40%, in all cases; this is probably related to the smaller amount of acid available to leach the solids.

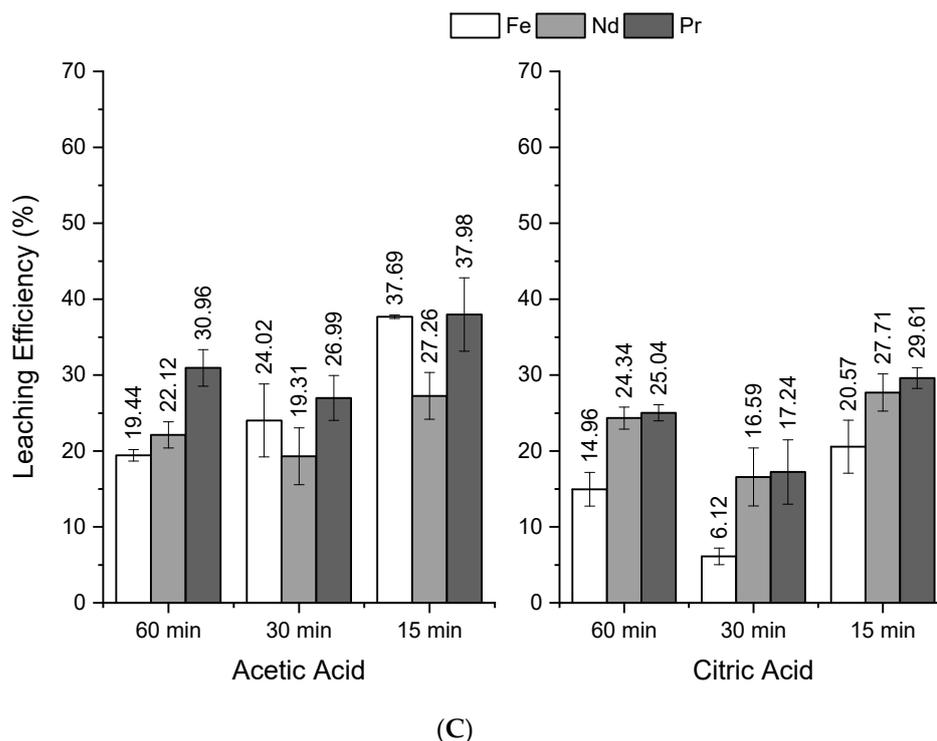


(A)



(B)

Figure 3. Cont.

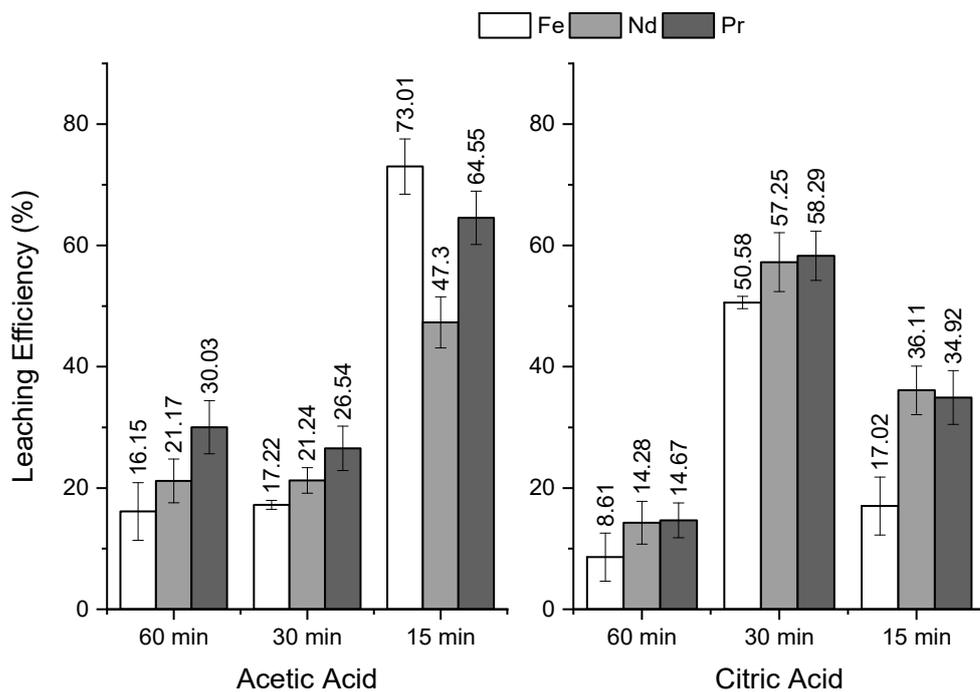


**Figure 3.** Comparison of the leaching efficiency of NdFeB magnets in acetic and citric acids, concentration 1 M, ratio s/l (A) 1/100; (B) 1/50 and (C) 1/10.

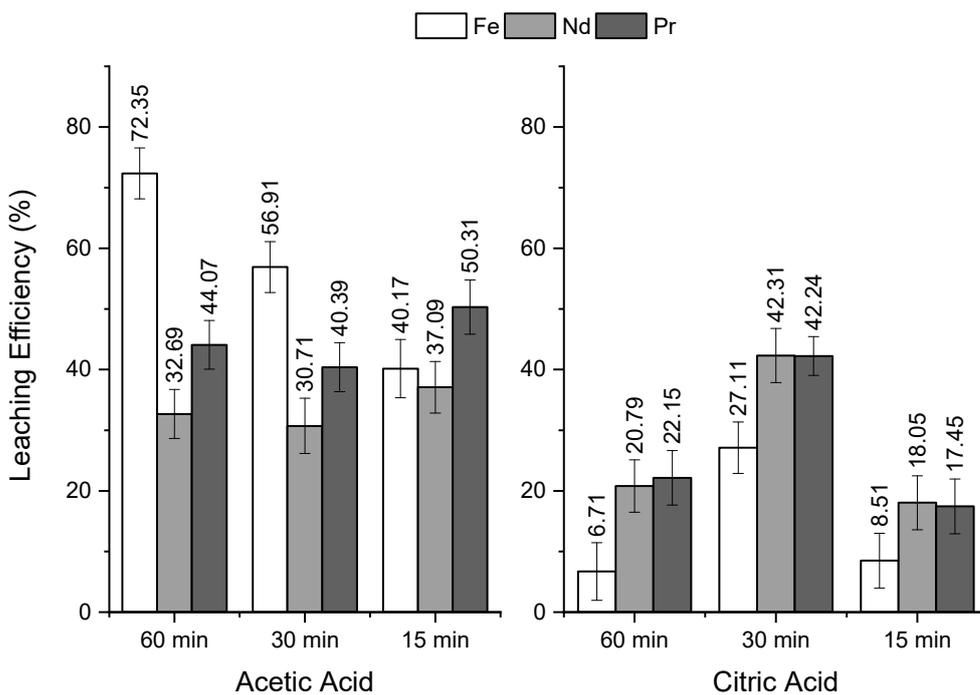
For acids at a concentration 0.5 M (Figure 4), the best results were obtained with acetic acid, in the solid/liquid ratio 1/100 and a time of 15 min, with a leaching efficiency of 48% for Nd and 65% for Pr. However, it is noteworthy that 0.5 M acetic acid leached iron contents higher than neodymium and praseodymium, contents in most cases, which makes the recovery of these metals difficult.

For citric 0.5 M acid, the best results were obtained in the solid/liquid ratio 1/100, in 30 min, with a leaching efficiency of 57% for Nd and 58% for Pr. Under the same conditions, more than 50% of the iron was leached. It is also observed that 0.5 M citric acid presented very low leaching efficiency for the solid/liquid ratio 1/10.

For acids at a concentration of 0.25 M (Figure 5), the leaching efficiency of both metals (Nd and Pr) was below 50% in all cases. In general terms, the concentration of 0.25 molar was less efficient than the other concentrations (1.00 M and 0.50 M) for both acids. The best results were obtained with 0.25 M acetic acid, at a solid/liquid ratio of 1/100, where the leaching efficiency was close to 32% for Nd and 47% for Pr. At this concentration, citric acid proved to be inefficient.

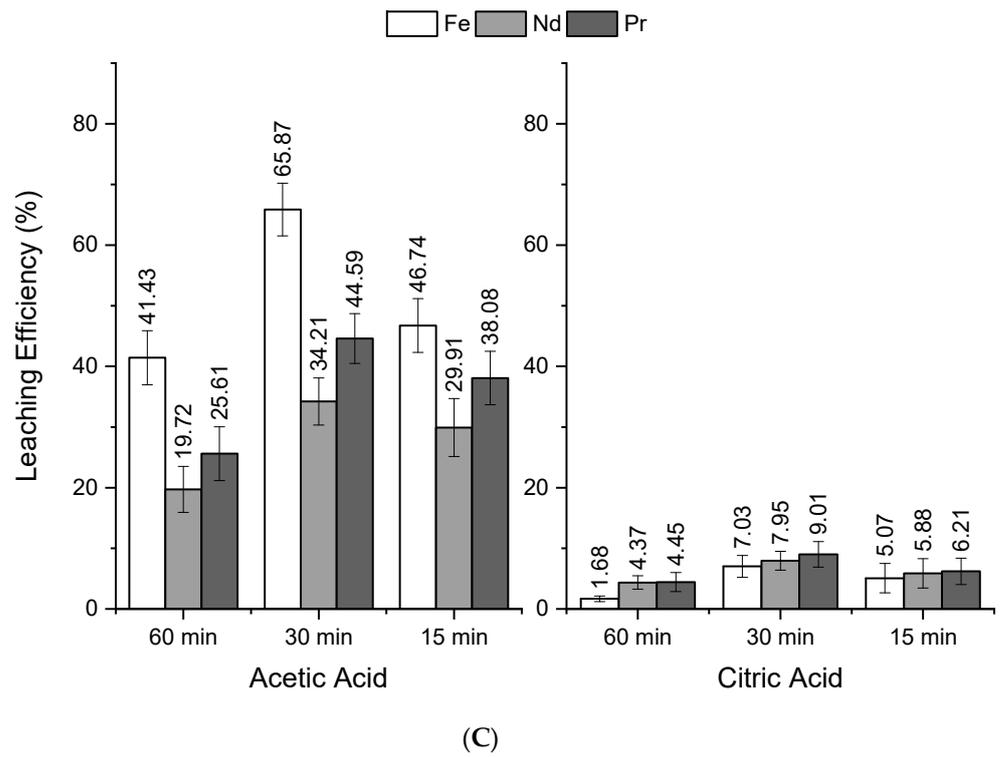


(A)

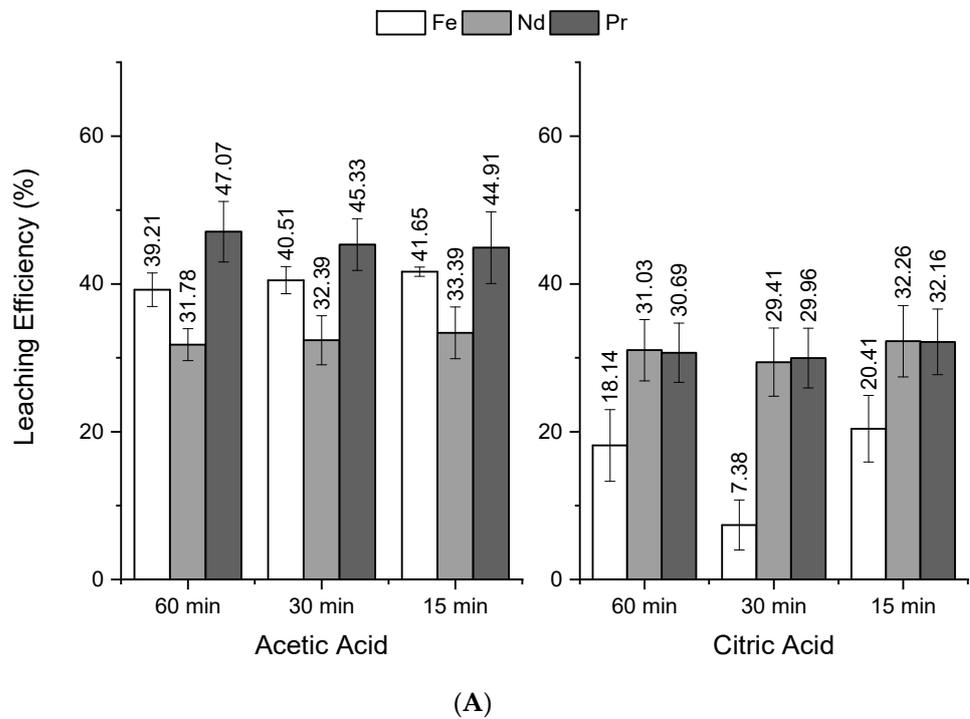


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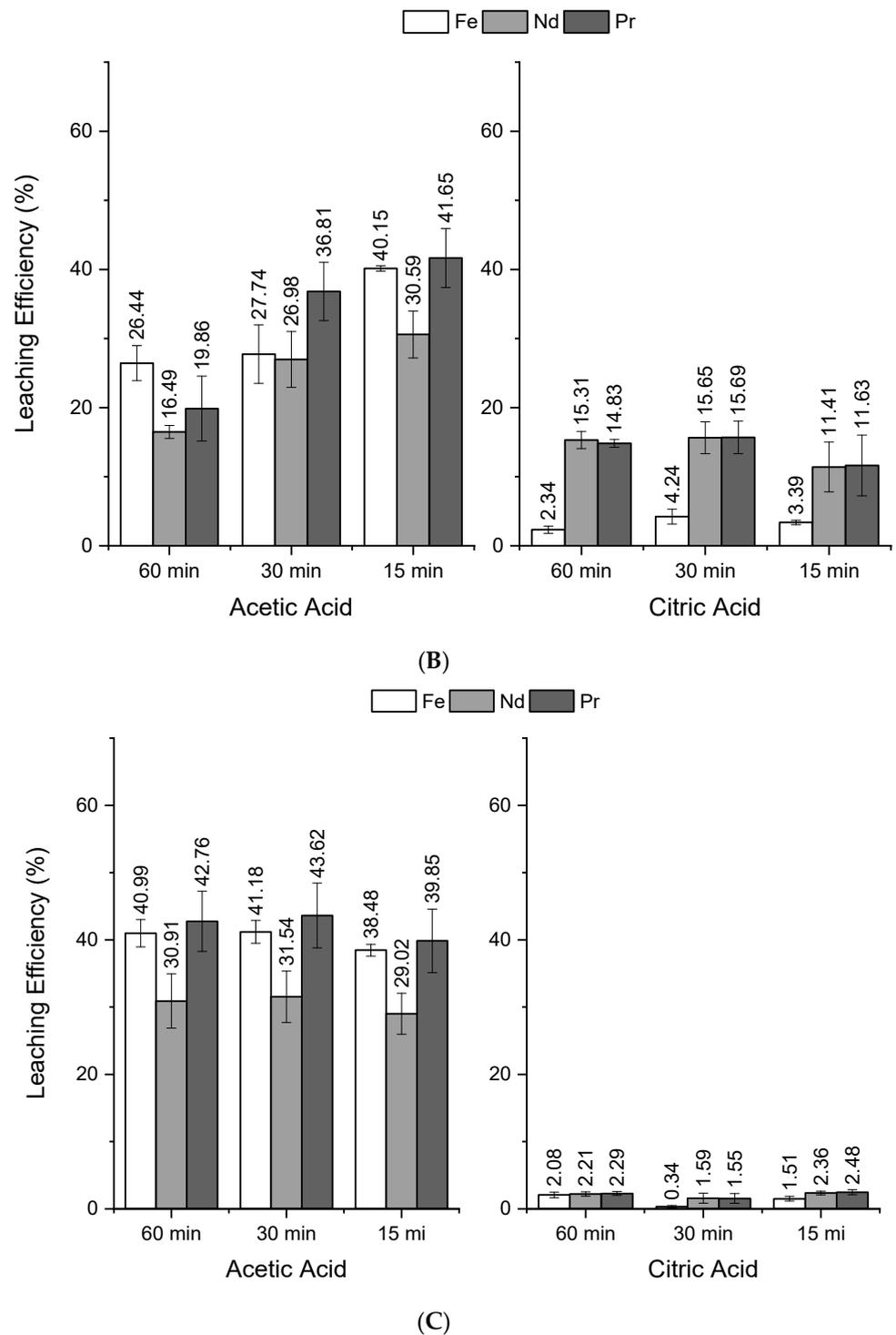
Figure 4. Cont.



**Figure 4.** Comparison of the leaching efficiency of NdFeB magnets in acetic and citric acids, concentration 0.5 M, ratio s/l (A) 1/100; (B) 1/50 and (C) 1/10.



**Figure 5.** Cont.



**Figure 5.** Comparison of the leaching efficiency of NdFeB magnets in acetic and citric acids, concentration 0.25 M, ratio s/l (A) 1/100; (B) 1/50 and (C) 1/10.

### 3.4.2. Ultrasound and Conventional Leaching

At this stage of the study, the leaching efficiency of REE with organic acids (acetic and citric) was analyzed, using conventional leaching and leaching with ultrasound. Acids were tested at concentrations of 1.0 M and 0.5 M, with leaching times of 120, 60 and 30 min.

### 3.4.3. Ultrasound Leaching

The results obtained in the tests using ultrasound (Figures 6 and 7), even using longer leaching times, were worse than the results obtained by the same acids using the microwave technique. It is also possible to observe that, in this case, citric acid showed better results than acetic acid.

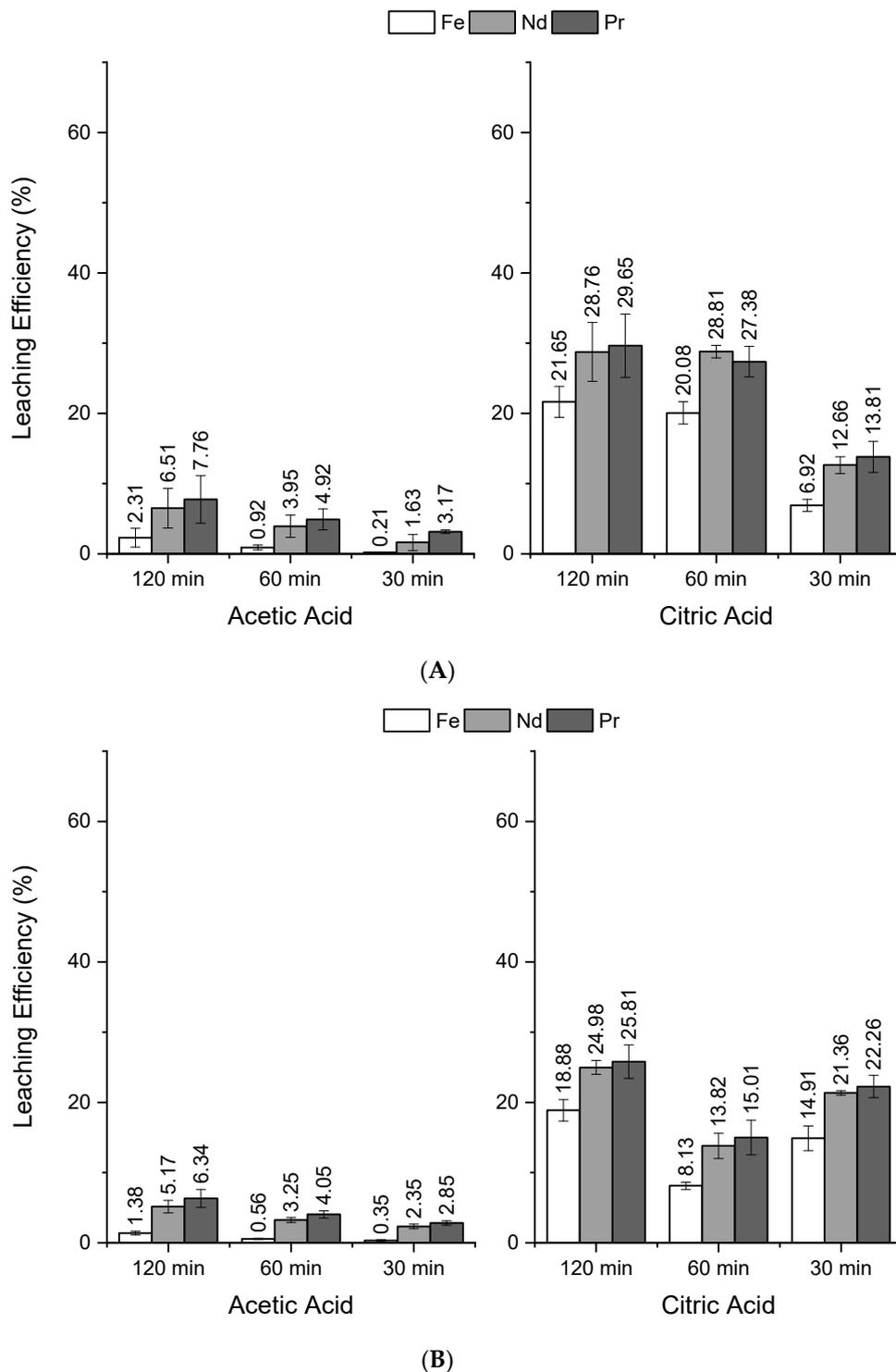


Figure 6. Cont.

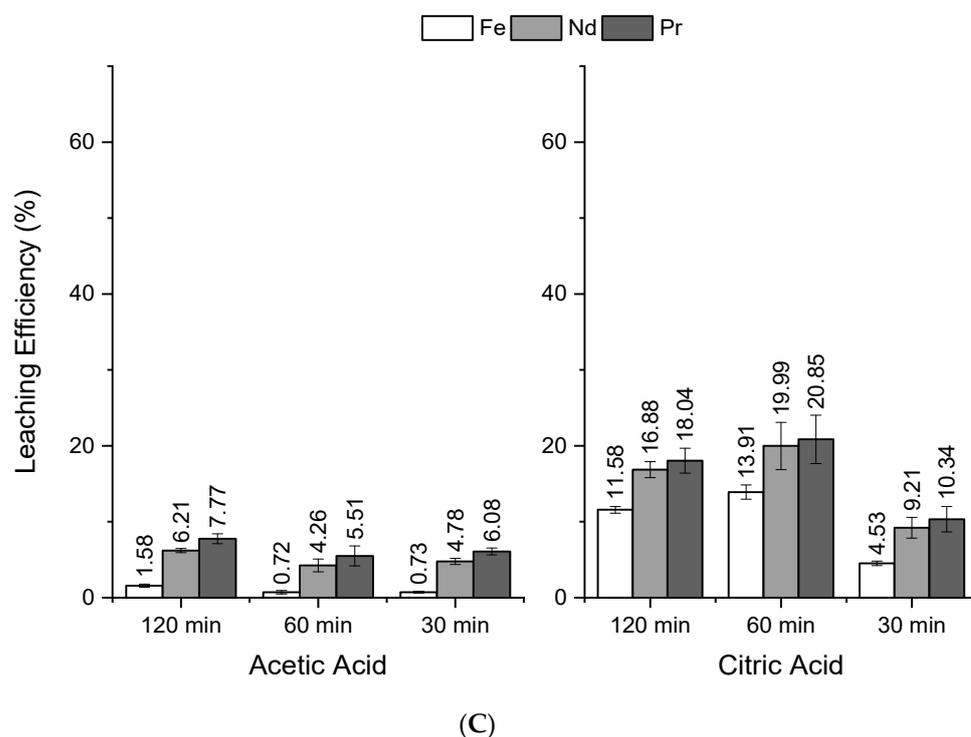


Figure 6. Comparison of the leaching efficiency of NdFeB magnets in acetic and citric acids, concentration 1 M, ratio s/l (A) 1/100; (B) 1/50 and (C) 1/10.

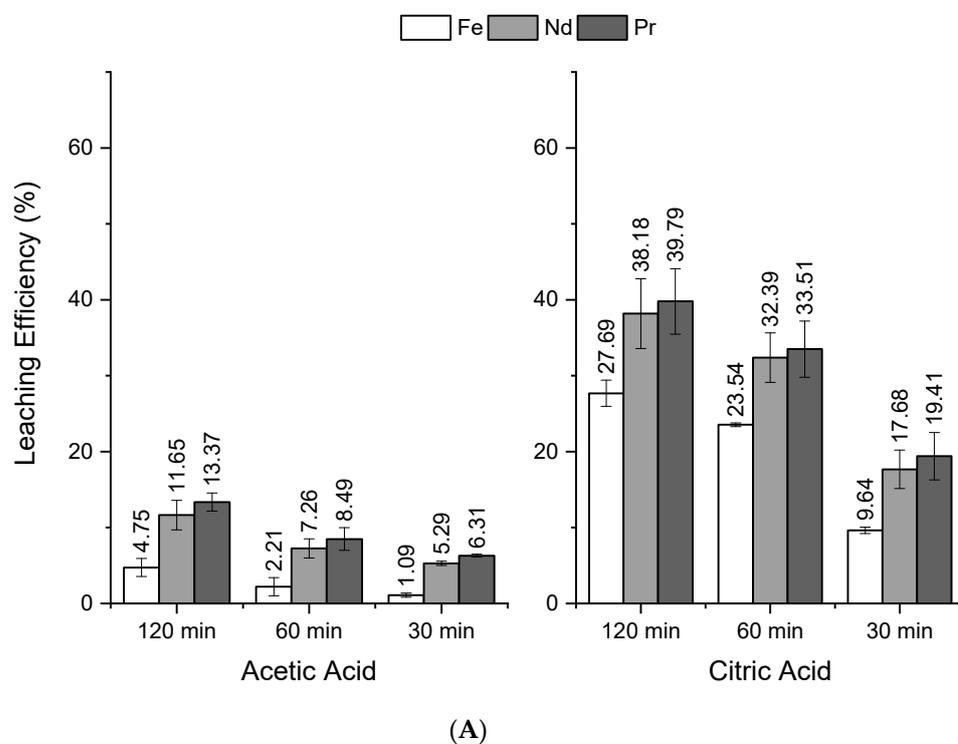
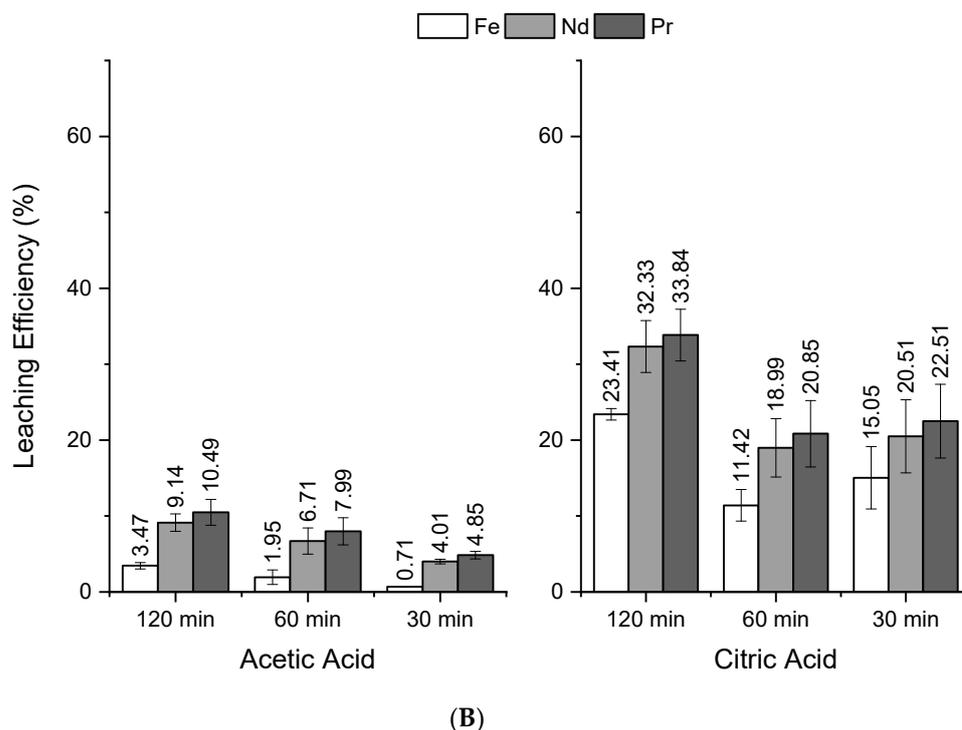


Figure 7. Cont.



**Figure 7.** Comparison of the leaching efficiency of NdFeB magnets in acetic and citric acids, concentration 0.5 M, ratio s/l (A) 1/100 and (B) 1/50.

For the concentration of 1.0 M (Figure 6), the best results were obtained with citric acid at a solid/liquid ratio 1/100, in 120 min, with a leaching efficiency of 29% for Nd and 30% for Pr. For this concentration, the leaching efficiency of acetic acid was below 10%, under all conditions.

In the results obtained in the tests with acids at a concentration of 0.5 M (Figure 7), an increase in leaching efficiency of citric acid in relation to a 1 M concentration was observed. In this case, the leaching efficiency was 38% for Nd and 40% for Pr, in the ratio solid to liquid 1/100, 120 min. The results of tests in which the solid/liquid ratio 1/10 were used were below 10% efficiency and therefore are not presented.

In general terms, it can be seen that under the conditions tested, ultrasound only proved to be slightly efficient, with all results being below 40% efficiency for Nd and Pr.

#### 3.4.4. Conventional Leaching

In conventional leaching (Figures 8 and 9), citric acid presented higher leaching efficiency in relation to acetic acid.

In tests with acids at a concentration 1 M (Figure 8), the best results were obtained with citric acid in a solid/liquid ratio 1/50, at 120 min, with a leaching efficiency of 33% for Nd and 35% for Pr.

The results obtained in the tests with acetic and citric acids at a concentration of 0.5 M (Figure 9), indicate a small reduction in leaching efficiency for citric acid, in relation to a concentration of 1.0 M, for all analyzed times. The best results were obtained with citric acid, in solid/liquid ratio 1/100 and time of 120 min, with a leaching efficiency of 31% for Nd and 32% for Pr. For acetic acid, the results indicate low leaching efficiency under these conditions.

This study aimed to establish the possible effects of concentration, leaching time and solid/liquid ratio on the leaching efficiency of neodymium and praseodymium with organic (acetic and citric) acids. However, as we can see in the results obtained, it was not possible to establish any pattern that could be directly or exclusively related to one of these factors.

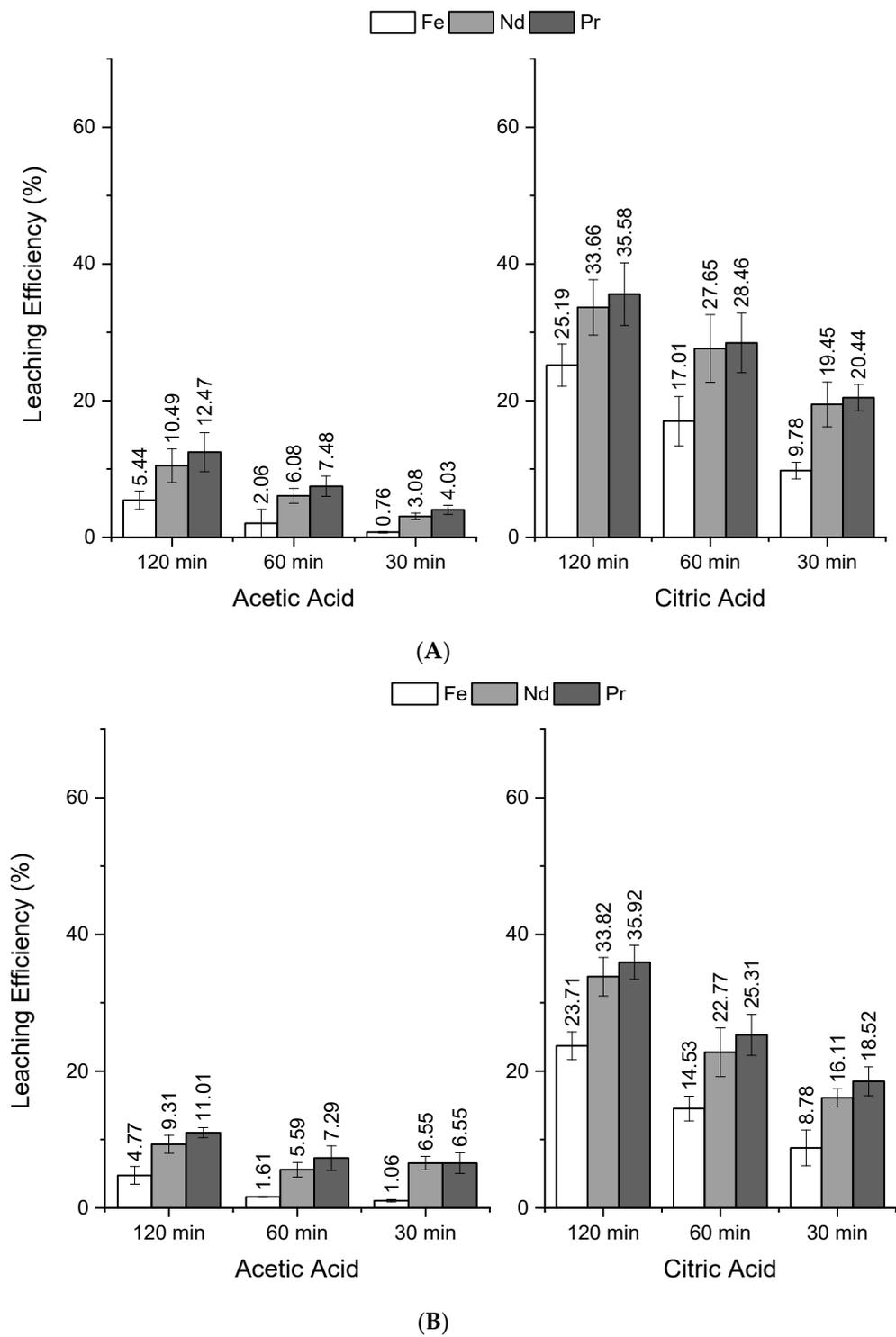


Figure 8. Cont.

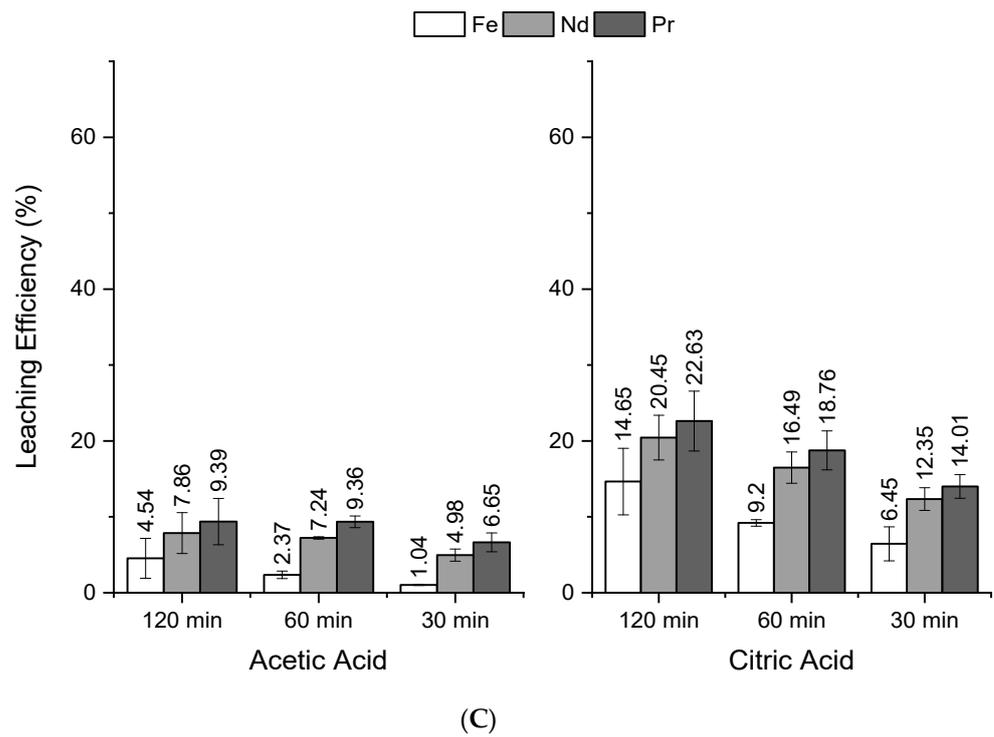


Figure 8. Comparison of the leaching efficiency of NdFeB magnets in acetic and citric acids, concentration 1 M, ratio s/l (A) 1/100; (B) 1/50 and (C) 1/10.

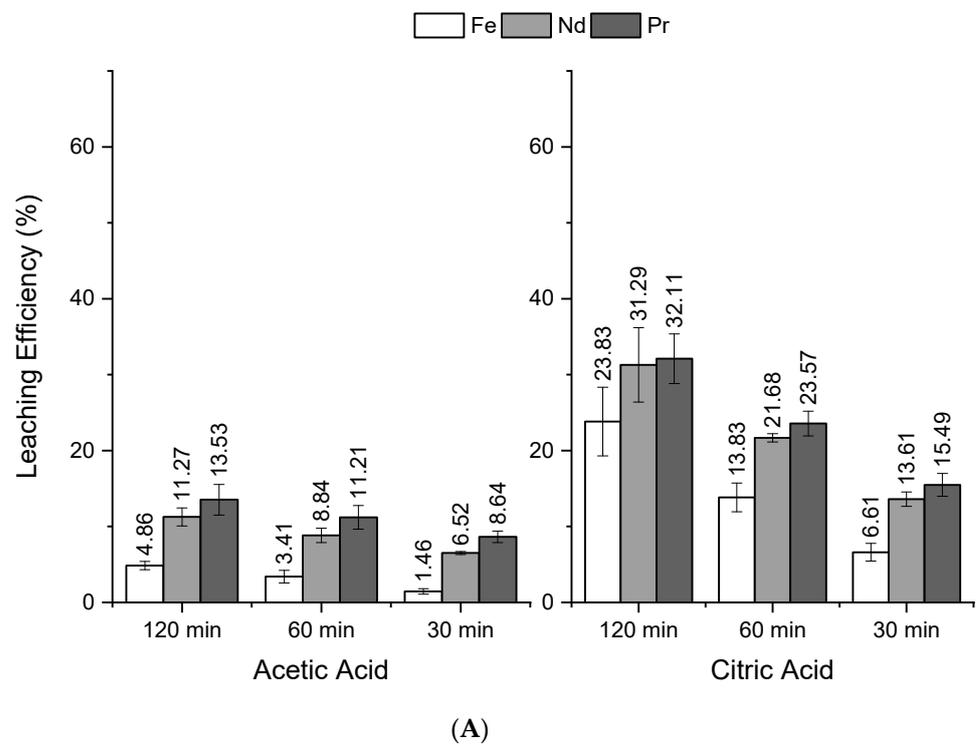
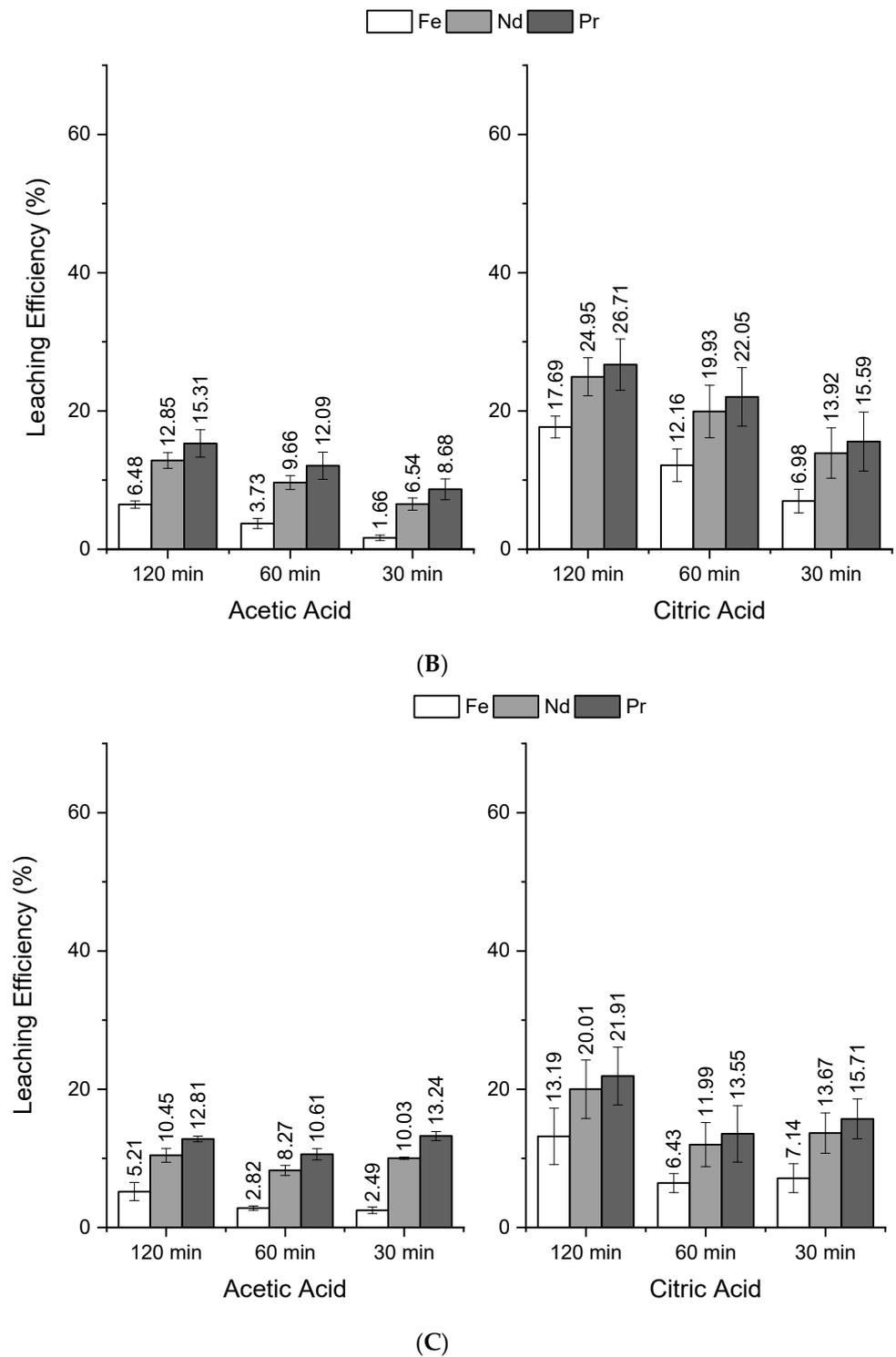


Figure 9. Cont.



**Figure 9.** Comparison of the leaching efficiency of NdFeB magnets in acetic and citric acids, concentration 0.5 M, ratio s/l (A) 1/100; (B) 1/50 and (C) 1/10.

In general terms, ultrasound leaching and conventional leaching showed similar efficiency results and, as expected, lower efficiency compared to the microwave method. The results obtained in microwave leaching tests indicate that organic acids (acetic and citric), have the potential to leach the Nd and Pr present in the NdFeB magnets present in mobile phones.

Although the results obtained in this study are less than the results obtained by other authors who tested the same acids in the leaching of NdFeB magnets, one must take into account the variables studied in each of the works. Behera and Parhi (2016), for example, obtained extraction rates of 99.9% of Nd and Fe in magnet leaching with acetic acid. However, the authors obtained these results using temperatures up to 80 °C and leaching times of 240 min. According to the authors, the acid concentration and the use of temperature seem to be critical in the effective dissolution of Nd and Fe [36]. Behera et al. (2019) investigated the application of ultrasound and microwave-assisted technology on the dissolution of neodymium from NdFeB magnets, with the use of organic acids (ascorbic, citric, acetic, formic and tartaric). According to the authors, acetic acid appears to be more effective in Nd leaching. Furthermore, both ultrasound and microwave assistance improved the dissolution of Nd, in relation to the regular process [44].

Furthermore, in most cases, high percentages of Fe were leached along with Nd and Pr, which demonstrates a low selectivity of these acids. The use of a previous heat treatment (roasting), and other techniques for recovering metals in solution can increase the selectivity in the recovery of Nd [39,59–61].

#### 4. Conclusions

From the results obtained in this research, it is possible to obtain the following conclusions:

- The amount of REE present in mobile phone devices increased from 28.1% (2000–2005 models) to 34.3% (2019–2020 models);
- The studied magnets contained, on average, 28.9% of REE. On average, these magnets had 23.9% of Nd, a higher amount than that found in ores;
- Microwave leaching was the most effective compared to ultrasound and conventional methods;
- The best results for citric acid were obtained in microwaves, using a concentration of 0.5 M, s/l ratio 1/100 and 15 min; in these conditions 57% of Nd and 58% of Pr were leached. For acetic acid the best results were obtained in microwaves, using concentration 0.5 M, s/l ratio—1/100 and 15 min; in these conditions 48% of Nd and 65% of Pr were leached;
- Under the same conditions, both citric acid and acetic acid also leached high percentages of iron (51% and 72%, respectively), demonstrating low selectivity;
- Considering the results obtained, it can be concluded that there is a great potential for using organic acids (acetic and citric) in the leaching of Nd and Pr (rare earths classified as light, with similar chemical properties). However, these acids require the use of more aggressive conditions than those used in this study, such as increased temperature and leaching time.

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**Data Availability Statement:** Publicly available datasets were analyzed in this study. This data can be found here: <https://idealmagnetsolutions.com/knowledge-base/rare-earth-metals-12-things-about-neodymium/>; <https://idealmagnetsolutions.com/knowledge-base/small-ndfeb-magnets-in-smartphones-and-personal-electronics>, accessed on 20 May 2022.

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