Investigating the magnetic and mechanical properties of NdCeFe$_{14}$B permanent magnets: ab initio study

Mphamela Baloyi[1*]*, Ramogohlo* Diale[2]*, Phuti* Ngoepe[1] and *Hasani* Chauke[1]

[1]Material Modelling Centre, University of Limpopo, South Africa
[2]Advanced Materials Division, Mintek, South Africa

Abstract. Neodymium-based permanent magnets (Nd$_2$Fe$_{14}$B) are the potential permanent magnets for various applications in high-efficiency power conversion machines and devices such as wind turbines and electric vehicles. This is due to their high magnetic field strength and demagnetization resistance. However, they suffer from low Curie temperatures below 585 K and poor mechanical properties. In this study, we investigate the effect of the rare earth element, Cerium (Ce) on the Nd$_2$Fe$_{14}$B permanent magnets. The structural, mechanical, and magnetic properties of the magnets are calculated using the ab initio density functional theory approach. Partial substitution of Nd with Ce improves the stability and the magnetic strength of Nd$_2$Fe$_{14}$B permanent magnets. Future research on permanent magnetic compounds with improved magnetic properties will benefit from the findings.

1 Introduction

The permanent magnet (PM) is a material that retains its magnetic properties due to its intrinsic structure [1]. PM includes a variety of magnets including alnico, ferrite, ceramic, and rare earth magnets. Rare earth (RE) magnets are the strongest permanent magnets available and possess unique magnetic properties such as high magnetic field strength, and resistance to demagnetization. Amongst RE$_2$Fe$_{14}$B magnets, Nd$_2$Fe$_{14}$B magnets are the most powerful rare earth magnets, with unique magnetic properties such as high magnetic field strength, high values of saturation magnetization, and resistance to demagnetization [2]. These magnets are used in cutting-edge technological applications such as high-performance motors, storage devices, electric vehicles, wind power generators and magnetic fasteners. However, they possess low Curie temperature (Tc = 585 K) [2], and poor mechanical properties displaying brittle behaviour [3].

Due to the drawbacks of Nd$_2$Fe$_{14}$B permanent magnets, partial substitution of Nd with other available rare earth elements is considered. Nd$_2$Fe$_{14}$B magnets alloyed with Dy have been reported to offer the best performance in the hard magnetic material market. However, Dy substitution reduces magnetic moments, compromising the strength of the magnets and NdDyFe$_{14}$B magnets are more expensive than regular Nd$_2$Fe$_{14}$B [4].

---

* Corresponding author: mphamela.baloyi@gmail.com

In this study, we explore the influence of the rare earth element, Cerium (Ce) on the $Nd_2Fe_{14}B$ magnets using the density functional theory (DFT) approach. The substitutional search tool was used to dope Ce in the $Nd_2Fe_{14}B$ structure. Heats of formation, elastic constants and magnetic moments of $NdCeFe_{14}B$ permanent magnet are reported.

2 Computational method

The calculations on structural, magnetic, and elastic properties were carried out employing first principle density functional theory (DFT) [5] using the Vienna ab initio simulation package (VASP) [6]. The Projector Augmented Wave (PAW) was used to illustrate how electrons and ions interacted [7]. The spin-polarized generalized gradient approximation parameterized by Perdew, Burke and Ernzerhof (GGA-PBE) [8] was chosen for the exchange-correlation functional. To converge the total energy of the tetragonal $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets, a 400 eV total energy cut-off was considered appropriate. To consolidate the total energy of the magnet, the appropriate 6x6x3 k-point mesh parameters from Monkhorst and Pack [9] were employed for all the structures. Before the calculations of ground state properties, full geometry optimisation is performed to relax the structure until the change in total energy is less than 0.01 eV/Å for the unit cell. The strain of 0.005 was used to determine the elastic properties of these magnets.

3 Results

3.1 Structural and thermodynamic properties

Table 1 shows the calculated lattice parameters of $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ at 0 K. The *a* and *c* parameters for $Nd_2Fe_{14}B$ magnets were found to be 8.23 Å and 12.12 Å, respectively. These findings are within 6% of the available experimental values [10] in a notable agreement. For $NdCeFe_{14}B$, *a* and *c* parameters are 8.63 Å and 11.95 Å, respectively. The *c* parameter decreases due to Ce having a small atomic radius (1.85 Å) compared to that of Nd (2.05 Å).

Moreover, heats of formation (ΔH_f) is calculated to predict the thermodynamic stability of these magnets. The ΔH_f is calculated using the following equation;

$$\Delta H_f = E_c - \sum_i x_i E_i,$$

where E_c is the calculated total energy of the compound, E_i is the calculated total energy of element i in the compound and x_i is the composition of each element. Note that negative ΔH_f indicates that the material is thermodynamically stable whereas positive ΔH_f suggests the instability of the material. The predicted ΔH_f suggests that both $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ are thermodynamically stable since they possess negative ΔH_f of -6.50 eV/atom and -6.35 eV/atom, respectively.

Table 1. The lattice parameters and the heats of formation (ΔH_f) for $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets.

Structures	Lattice Parameters (Å)		Heats of formation (eV/atom)
	Calculated	Experimental[10]	
$Nd_2Fe_{14}B$	$a = 8.23$ $c = 12.12$	$a = 8.82$ $c = 12.25$	-6.50
$NdCeFe_{14}B$	$a = 8.63$ $c = 11.95$		-6.35

3.2 Elastic properties

The calculated elastic constants for $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets are shown in Table 2. Elastic constants are calculated to determine the mechanical stability of these magnets. For the magnets to be deemed stable, they must meet the tetragonal stability criteria; otherwise, they would be considered mechanically unstable. The following is the condition of tetragonal crystal stability:

$$C_{44} > 0, C_{66} > 0, C_{11} > |C_{12}| \ and \ C_{11} + C_{12} - \frac{2C_{13}^2}{C_{33}} > 0.$$

Since the computed elastic constants meet the requirements for tetragonal stability, the $Nd_2Fe_{14}B$ magnet is mechanically stable. It is found that C_{11} (298.13 GPa) is larger than the C_{12}, 159.30 GPa ($C_{11} > |C_{12}|$). C_{44} have values of 65.77 GPa and C_{66} to be 119.56 GPa, which are non-zero. In the case of $NdCeFe_{14}B$ magnet, C_{11} (216.10 GPa) greater than the absolute of C_{12} (51.10 GPa), C_{44} (24.14 GPa) and C_{66} (116.45 GPa) are both non-zero. This indicates that the magnet is mechanically stable since satisfying the tetragonal stability criteria.

Table 2. Elastic constants of the $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets.

Elastic Constants (C_{ij})	$Nd_2Fe_{14}B$ (GPa)	$NdCeFe_{14}B$ (GPa)
C_{11}	298.13	216.10
C_{12}	159.30	51.10
C_{13}	65.59	22.86
C_{33}	94.12	159.50
C_{44}	65.77	24.14
C_{66}	119.56	116.45
C'	69.42	82.50

In order to determine the brittleness or ductility of $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets, Pugh's and Poisson's ratios are calculated. Pugh's (B/G) and Poisson's (v) ratio of the $Nd_2Fe_{14}B$ and

$NdCeFe_{14}B$ permanent magnets are shown in Table 3. According to Pugh, a material is deemed brittle when the B/G value is less than 1.75 and ductile when it is more than 1.75 [11]. The calculated B/G of $Nd_2Fe_{14}B$ is 1.13, indicating brittleness in the magnet. We observe that when Nd is replaced with Ce, the B/G of $Nd_2Fe_{14}B$ increases to 1.58. Since B/G is smaller than 1.75, this suggests that the $NdCeFe_{14}B$ permanent magnet is brittle. Additionally, the Poisson's ratio for these magnets is computed and shown in Table 3. A material can be classified as ductile if Poisson's ratio value is higher than 0.26; if not, it can be classified as brittle [12]. $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets are both brittle, due to their computed Poisson's ratios being less than 0.26 (values 0.20 and 0.24, respectively). These results are consistent with the theoretical results that have been previously reported and indicate that the $Nd_2Fe_{14}B$ magnet is brittle, with a Poisson's ratio of 0.22 [13].

Table 3. Pugh's (B/G) and Poisson's (v) ratio of the $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets.

Structures	B/G	v	
		Calculated	Theoretical[13]
$Nd_2Fe_{14}B$	1.13	0.20	0.22
$NdCeFe_{14}B$	1.58	0.24	

3.3. Magnetic properties

Table 4 shows the computed magnetic moments of $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets. Note that the magnetic moments are determined to deduce the magnetic strength of these magnets. The magnet with high magnetic moments possesses high magnetic strength. A magnetic moment of 35.63 μ_B is found for $Nd_2Fe_{14}B$ magnets, which is within 2 % of the experimental results of 35.00 μ_B [2]. It is worthwhile to observe that the magnetic moment decreases when Nd is substituted with Ce. $NdCeFe_{14}B$ magnet has calculated magnetic moments of 26.05 μ_B. The experimental results show that Ce has an insignificant impact on the magnetic moments of the material compared to Nd, with $Ce_2Fe_{14}B$ having a magnetic moment of 22.70 µB [2].

Table 4. Magnetic moments of the $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets.

Structures	*Magnetic Moments* (μ_B)
$Nd_2Fe_{14}B$	35.63
$NdCeFe_{14}B$	26.05

4 Conclusions

The structural, thermodynamic, elastic and magnetic properties of $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ permanent magnets were calculated using ab initio DFT. The calculated lattice parameters agree well with the experimental values, with Ce decreasing the lattice parameters due to the small atomic radius. It was observed that $Nd_2Fe_{14}B$ and $NdCeFe_{14}B$ possess negative heats of formation which indicates that both magnets are thermodynamically stable. $NdCeFe_{14}B$ magnet was found to be mechanically stable satisfying the tetragonal stability criteria. Moreover, these magnets exhibit brittle behaviour. Partial substitution of Nd with Ce reduces

the magnetic moments of $Nd_2Fe_{14}B$ to 26.05 μ_B, which is still considerably high. Future research on permanent magnetic compounds will benefit from the findings.

References

1. Coey J M D. Scripta Materialia, **67**, (2012)
2. Sagawa M, Fujimura S, Yamamoto H, Mutsuura Y. IEEE Transactions on Magnetics. **20,** (1984)
3. Yin X, Yue M, Lu Q, Wang F, Qiu Y, Liu W, Zuo T, Zha T, Li X, Yi X. Eng. **6 (**2020)
4. Coey J M D. IEEE Transactions on Magnetics. **47**, (2011)
5. Kohn W, Sham L J. Phys. Rev. B. **140**, (1965)
6. Kresse G, Hafner J. Phys. Rev. B. **49**, (1994)
7. Kresse G, Joubert D. Phys. Rev. B. **59**, (1999)
8. Perdew J P, Burke K, Enzerhof M. Phys. Rev. Lett. **77**, (1996)
9. Monkhorst H J, Pack J D. Phys. Rev. B. **13**, (1976)
10. Watson R E, Bennett L H. Phys. Rev. Lett. **43**, (1979)
11. Pugh S F. J. Scie. **45**, (1954)
12. Sin'ko G V, Smirnov N A. J. Phys. Condens. Matter. **14**, (2002)
13. Yu J, Jiang S, Sun D, Lin P, Zhang Y. J. Mater. Technol. **18**, (2022)