A Review of M-Type Ferrites as Rare-Earth-Free Permanent Magnets for Green Energy and Sustainable Transportation

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Abstract:

The emergence of clean energy around the globe has propelled the development of electric vehicles (EVs), which are considered to be environmentally sustainable means of transport compared to those run on fossil fuels. However, the utilization of rare earth elements (REE) containing permanent magnets in the EV traction motors is faced with several problems due to resource limitation, uneven resource distribution, volatile prices and the environmentally unfriendly process of extracting the minerals. Rare-earth-free materials have become a suitable option towards attaining sustainable electric mobility. The M- type hexaferrites such as $BaFe_{12}O_{19}$ & $SrFe_{12}O_{19}$ retain many advantages over other ferrite materials, being low cost, chemically stable, high electrical resistivity and availability of their components. The magnetic properties of hexaferrites can be improved by compositional, size and magnetic anisotropy modification. This paper reviews different dopant elements to increase the permanent magnetic properties of the material. These advances make them more energy efficient for electric motors and other green energy technologies, decrease reliance on critical materials, minimize environmental impact, and contribute to a more circular, low-carbon, and sustainable transportation system.

Keywords: M-type ferrite, Electric Vehicle, Coercivity, Remanence

1. Introduction

Climate change is causing a worldwide shift towards clean energy, particularly in the transport sector, where 49% of conventional energy consumption & emission occurs. Oil resources are predicted to run out by 2038, creating the urgency to find out alternative source of energy. The use of combustion free transport is transforming the world. world is growing towards a zero-carbon transition, that involves eliminating all combustion. The advancement in electric vehicles (EVs) come out as the key option for its replacement [1]. The overall forecast efficiency of electric cars will be between 40% to 70% compared to the 20% - 30% efficiency of petrol or diesel vehicles [2]. Transport is evident with 6.5 million EVs sold worldwide in 2021. 23% of all EVs were sold in China [3] which makes it the world's largest EVs market in less than a decade. Europe is moving in the same direction, with more than 2.3 million EVs sold in 2021, accounting 19% of all sales in 2021 [2]. In recent years, a significant attention has been paid to the following remarkable phenomenon. The two key components of an EVs that have a direct impact on its cost and efficiency are the battery and motors. Across 95% of EVs are using permanent magnets (PM)

Synchronous motors. The permanent magnetic substance used to utilize in these motors is the primary focus [4]. Permanent magnets are an essential component for many others modern devices as well, including computers, mobile phones, generators and magnetic levitation transport [4]. Rare earth element magnets are widely used in EVs application or other application where permanent magnet case arises. However rare earth elements are mined and processed by a few nations, resulting in price fluctuations and geopolitical risks [4]. Mining and processing rare earth elements can also harm the environment by generating radioactive waste, contaminating water and damaging ecosystems. The long-term viability of emerging magnetic energy sector depends on developing new technology that eliminates or reduces the need of rare earth materials [5]. As EVs become more dependent on rare earth elements, therefore the manufacturing of high-performance permanent magnets for traction drive poses several challenges due to the large scale and green development of electric vehicle [6]. While the rare earth elements-based magnets like NdFeB, possess very high magnetic properties, but the extraction of rare earth and processing of the magnets are known to have environmental impact. The major concerns are land degradation, high energy demand, contamination of water and soil which makes the extraction [2] and processing extremely susceptible, both economically and geopolitically for the EV industry [6]. The high and volatile price of rare earth elements, combined with their geographically concentrated supply chains makes this concern more serious. The challenges can lead to higher manufacturing costs, which impact the sustainability and scalability of EV production in the long term [6].

To control the environmental, economic, and geopolitical concerns related to the use of rare earth elements (REE) in PM, there has been an alternative solution to be used as the motor applications of rare earth elements [6]. The first one is to substitute PM with electromagnets; however, electromagnetically excited machines require a constant electricity supply, which leads to increased copper losses, heating, and machine complexity. Despite the absence of the need for rare earth, application of such motors in EVs' traction system is difficult [7]. Another alternative option is to reduce, reuse, and recycle rare earth magnets. The third and promising long-term strategy is the use of REE-free magnetic materials. Figure 1 shows the magnetic properties of different material such as Smco, AlNiCo and ferrite along with NdFeB [8]. The reason behind the use of $BaFe_{12}O_{19}$ and $SrFe_{12}O_{19}$, lies in their cost-effectiveness, excellent chemical and thermal stability, higher electrical resistivity, and availability of raw material. Due to their good electrical and magnetic properties, these compounds could be used as potential materials for producing permanent magnets. Traditional ferrites show poorer saturation magnetization and energy product than NdFeB magnets (as shown in Figure 1) and hence cannot be directly used in high-power-density EV motors. The problem can be mitigated by using different methods for the modification of the ferrites, like compositional variation, controlling grain size, enhancing remanence, coercivity, and magnetic anisotropy, etc. M-type hexaferrite provides a promising way for decreasing reliance on REEs in EV motors[7]. The combined materials and motor design approach can offer a rare-earth-free and sustainable solution for electric mobility [6].

To solve this problem, efforts have been made to enhance the magnetic properties of M-type hexaferrites such as $BaFe_{12}O_{19}$ and $SrFe_{12}O_{19}$ by the use of one or more dopant elements. The crystal structure, magneto crystalline anisotropy, saturation magnetization, remanence, and coercivity of these ferrites can be modified by these dopant elements to meet their lower magnetic performance of as compare to rare-earth magnets. The magnetic properties of metal ions have been changed by substituting different metal ions into different crystallographic sites, in order to change the magnetic behaviour of the overall material. Various substituent

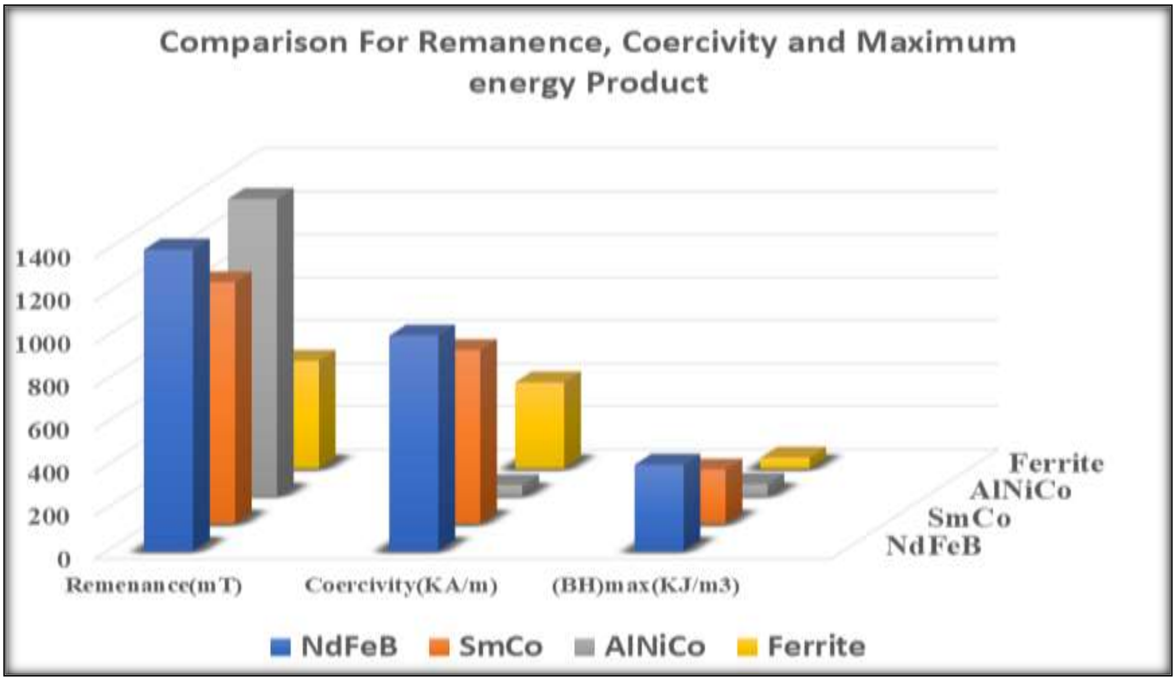

Figure 1: Comparison of magnetic properties such as coercivity, remanence, and $(BH)_{max}$ of different magnetic materials [8]

elements such as Al^{3+} [9], Cr^{3+}[10], Cu^{2+} [11], La^{3+}[12], Nd^{3+}[13] etc. have been studied singly or in combination to obtain the best combination of high values of coercivity, remanence, saturation magnetization, and magnetic anisotropy. Besides compositional modifications, the control of the concentration of the dopant, its ionic distribution, and grain size and microstructural characteristics are important factors that affect the magnetic properties of the resultant material. Hence, the present review is limited to the different types of dopant combinations that have been done on M-type hexaferrites, systematically examining the changes in structural and magnetic properties, highlighting the best doping method for optimizing the performance of hexaferrite permanent magnetic material for use in next-generation electric vehicle traction motors, without the presence of rare earth elements.

2. Synthesis method

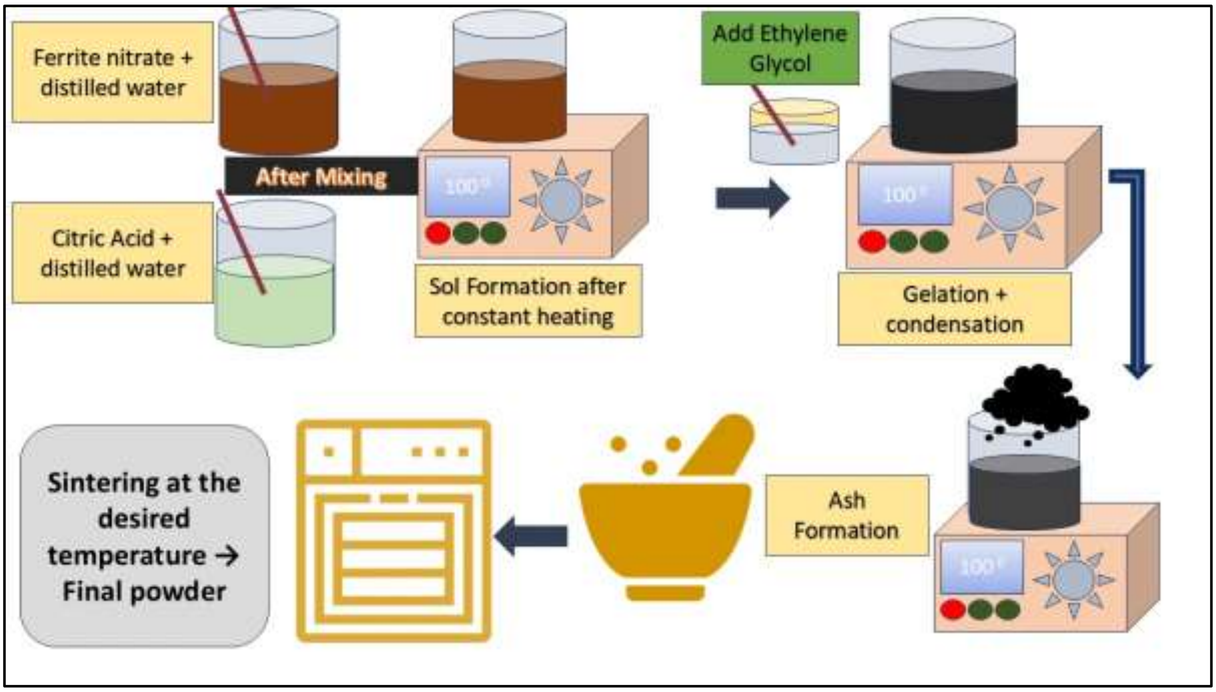

Figure 2: Sol gel method for the preparation of ferrite material

Attempts have been made since 80s to prepare the nano powder accordingly. The M-type hexaferrite is prepared with the help of different synthesis routes for specific applications. It can be prepared via solid-state reaction[14–16], microemulsion [17,18], combustion synthesis [19–24], aerosol pyrolysis [25], glass crystallization [26,27], sol–gel [28,29], spontaneous combustion technique[30], and hydrothermal techniques [31–35]. Among all these routes the low temperature synthesis method is considered to be the most impactful for the preparation of M type hexaferrite material with controlling shape and size. Therefore, sol gel method is the best for the synthesis of M type ferrite with controlled shape and size [36–39]. To improve the magnetic properties via regulating the size and reduce the formation of secondary phase, there has to be control over the several parameters. Nga and coworkers prepared the strontium ferrite nano powder by sol gel method and studied the effects of calcination temperature and the molar ratio of Fe/Sr, influence of pH and time on the magnetic properties of SFO [40–42].

The synthesis of magnetic nano particle is done via sol gel auto combustion method as shown in Figure 2. This synthesis is advantageous because the sample is prepared using simple equipment and a straightforward route, yielding high powder purity, chemical homogeneity, and crystallinity, resulting in the creation of ferrite nanoparticles [43,44]. Highly pure chemicals are used in the experiment, including ferric nitrate nonahydrate Fe $(NO_3)_3.9H_2O$, barium nitrate $Ba(NO_3)_2$, strontium nitrate $Sr(NO_3)_2$, anhydrous citric acid $C_6H_8O_7$, and ethylene glycol $C_2H_6O_2$. To prevent the precipitation of hydroxylated molecules and to form complexes with metal ions, citric acid functions as a chelating agent [45–47].

3. Literature review

Doping	Formula Used	Route of Synthesis	x , y	H_c(KOe)	M_s(emu/g)	M_r(emu/g)	ref.
Al-Nd	Sr1-	Sol-gel	0.00, 0.00 0.05, 0.5	5.5 3.51	64.4 42.93	39.29 28.54	[4]

	$xNd_xAlyFe_{12-y}O_{19}$	auto-combustion method	0.10, 1.00 0.15, 1.50 0.20,2.00	3.57 4.52 8.00	44.57 36.38 23.01	29.55 24.65 15.61	
Al & Pr	$SrFe_{12}O_{19}$, $Sr_{1-x}PrxFe_{10}Al_2O_{19}$	Sol-gel auto-combustion technique	0.00 0.05 0.10 0.15 0.20	5.500 10.500 10.009 10.008 10.009	82.76 29.51 40.85 25.50 24.01	50.11 17.65 23.84 14.16 12.22	[5]
Hard /Soft	(1-x) $SrFe_{10}Al_2O_{19}$/ x $CoFe_2O_4$	Sol-gel auto-ignition method	0.00 0.05 (CM) 0.10 (CM) 0.15(CM) 0.20(CM) 0.05 (PM) 0.10 (PM) 0.15 (PM) 0.20 (PM) 1.00	10.50 3.24 4.45 2.6 2.06 4.42 4.41 2.0 3.2 1.25	29.71 37.94 40.01 42.76 32.06 49.71 53.55 63.64 43.86 70.31	17.65 25.21 27.52 25.61 19.38 33.58 35.53 38.48 29.15 34.61	[48]

Overall, these studies indicate that there is no single dopant that simultaneously maximizes H_c, M_s, and M_r. Instead, the choice of dopant or composite phase must be guided by the desired application. For permanent-magnet and high-coercivity applications, Al–Pr or optimized Al–Nd substitution appears advantageous, whereas for applications requiring a better balance between magnetization and coercivity, hard/soft ferrite composites such as $SrFe_{10}Al_2O_{19}/CoFe_2O_4$ provide a promising strategy.

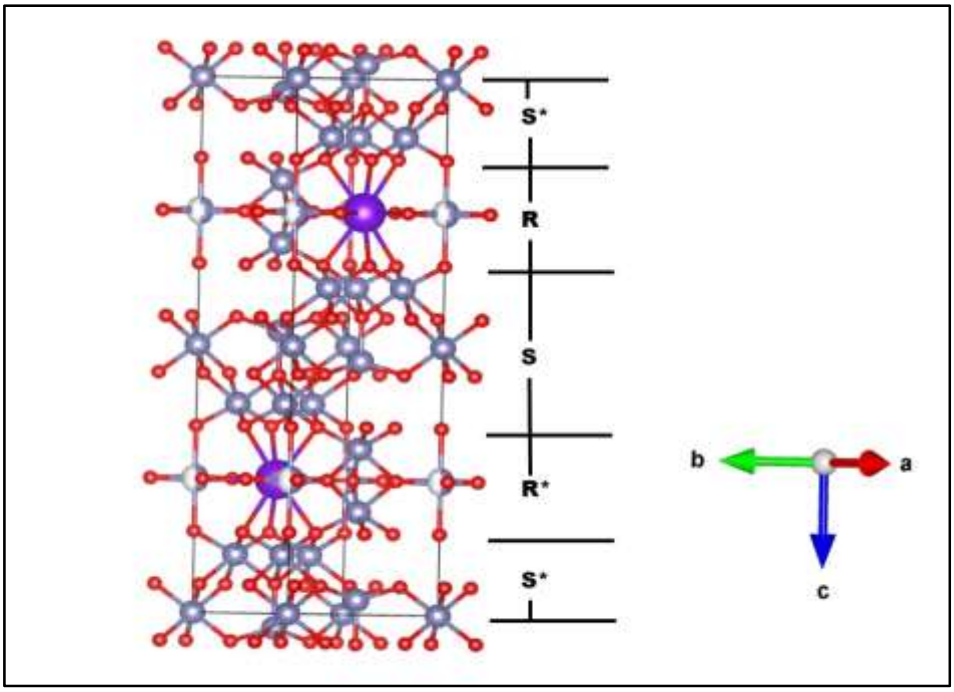

Figure 3: Pictorial representations of the structure of M-type hexaferrite

Sharma et al. have studied the effect of Al-Nd co-doping on SrM hexaferrite. The SrM hexaferrite with the composition of $Sr_{1-x}Nd_xAl_yFe_{12-y}O_{19}$ is synthesized using the sol-gel auto-combustion method, and the combined substitution of Nd^{3+} and Al^{3+} in SrM hexaferrite has been investigated. The values of coercivity (H_c), saturation magnetization (M_s), and remanent magnetization (M_r) of the undoped sample were found to be 5.5kOe, 64.4 emu/g, and 39.29 emu/g, respectively. The magnetic parameters were found to vary nonlinearly with the increase of Nd and Al concentration. At x = 0.05 and y = 0.5, H_c decreased to 3.51 kOe from 5.5 kOe for the pure sample, while M_s and M_r decreased to 42.93 and 28.54 emu/g. A marked increase in the doping level led to the variation in the coercivity value, with an H_c value of 8.0 kOe at x = 0.20, y = 2.0, but M_s continually decreased to 23.01 emu/g, and M_r decreased to 15.61emu/g. The decrease in the magnetization can be related to the substitution of magnetic Fe^{3+} ions by non- equivalent substituted ions, which leads to changes in the super exchange interactions between different crystallographic sites. The value of H_c at higher concentrations of Al and Nd suggests that there are some variations in magnetic hardness due to Al-Nd concentration, which may be attributed to variation in magneto-crystalline anisotropy and microstructural properties

In the next event, Sharma et al have synthesized the M-type ferrite with Al-Pr dopant. The other combination of interest is Pr^{3+} and Al^{3+}, which can be found in $SrFe_{12}O_{19}$ and $Sr_{1-x}Pr_xFe_{10}Al_2O_{19}$. The material has been prepared by the sol-gel auto-combustion method. The H_c, M_s, and M_r of the undoped $SrFe_{12}O_{19}$ were found to be 5.50 kOe, 82.76 emu/g, and 50.11 emu/g, respectively. When Pr^{3+} and Al^{3+} were added, an interesting increase in the coercivity was noticed. The value of Hc for the sample was found to be nearly double that of the undoped sample at x = 0.05. This suggests that the substitution of Pr into Al can greatly improve the magnetic hardness of SrM ferrite. The coercivity however, increased strongly with a simultaneous dramatic decrease of the Ms and Mr reaching 29.51 and 17.65 emu/g respectively, for x = 0.05 as the Pr concentration increased, the Hc values remained near 10kOe, while the Ms and Mr values showed a decreasing trend, reaching value of 24.01 and 12.22 emu/g at x = 0.20 respectively. This behaviour represents the typical coercivity or magnetization trade-off that occurs when substituting rare earth ions and non-magnetic ions. Magnetic anisotropy can be modified by the substitution of Al^{3+} for Fe^{3+}.

Hard/ soft magnetic composite

$SrFe_{10}Al_2O_{19}$ and $CoFe_2O_4$ another way of changing the magnetic properties of SrM ferrite is to create the hard/soft magnetic composite by adding soft magnetic $CoFe_2O_4$ to Al-substituted SrM hexaferrite. The reported system is (1−x) $SrFe_{10}Al_2O_{19}$ / x $CoFe_2O_4$ and was prepared by the sol-gel auto-ignition technique. A chemical mixing as well as a physical mixing (PM) approach were explored. The undoped hard ferrite exhibited Hc = 10.50 kOe, Ms = 29.71 emu/g and Mr = 17.65 emu/g. All these parameters were significantly changed when the soft magnetic phase was added. For the chemically mixed samples, Hc varied from 3.24 to 2.06 kOe for x = 0.05 – 0.20, while Ms reached values between 37.94 and 42.76 emu/g. However, the samples magnetized physically exhibited comparatively high Ms values with an increase to 53.55 emu/g for x = 0.10 and 63.64 emu/g for x = 0.15. Also, the corresponding Mr values showed significant rise in comparison with open hard phase up to x= 0.15 for PM samples, which is 38.48 emu/g. However, the coercivity dropped significantly as all the soft magnetic $CoFe_2O_4$ phase weakens the magnetic hardness of the system. The results show that hard/soft phase coupling is an effective method to tailor the magnetic properties. The high coercivity of the hard magnetic phase SrM is combined with the high magnetization of the soft magnetic phase $CoFe_2O_4$. Thus, the magnetic properties can be tailored for the desired use.

From the above literature, it is evident that dopant selection is a key parameter for controlling the magnetic behaviour of SrM hexaferrites. Rare earth / nonmagnetic co substitution such as Al, Pr and Al, Nd, primarily modifies the magnetic anisotropy and Fe^{3+} O^{2-}, Fe^{3+} super exchange interactions, leading to substantial changes in coercivity and saturation magnetization. In particular Pr, Al substitution is highly effective in enhancing coercivity, making such compositions attractive for hard magnetic applications. On the other hand, the incorporation of $CoFe_2o_4$ as a soft magnetic phase provides a fundamentally different mechanism for property modification. The hard/soft composite approach generally decrease coercivity while increasing saturation and remanent magnetization. The differences between chemical and physical collaboration further demonstrate that phase distribution and interfacial coupling are important factors in determining the final magnetic performance. Overall, these studies indicate that there is no single dopant that simultaneously maximizes H_c, M_s, M_r. Instead, the choice of dopant or composite phase must be guided by the desired application. For permanent- magnet and high coercivity applications, Al, Pr, or optimized Al, Nd substitution appears advantageous, whereas for applications requiring a better balance between magnetization and coercivity. Hard/soft ferrite composites such as $SrFe_{10}Al_2O_{19}$/ $CoFe_2O_4$ provide a promising strategy.

4. Conclusion

M-type hexaferrites like $BaFe_{12}O_{19}$ and $SrFe_{12}O_{19}$ have been considered to be a suitable rare-earth-element-free alternative solution for permanent magnetic applications. The concern about rare-earth magnets is resource availability, high prices, geopolitical dependence, and the environmental impacts of rare-earth resource extraction and processing. The review highlights that the magnetic properties of M-type hexaferrite can be modified effectively by introducing the dopant element, choosing a suitable synthesis route and sintering temperature, and microstructural engineering and hard-soft super-exchange coupling. The use of different dopant elements and hard-soft super-exchange coupling highlights that the

control of dopant parameters would be necessary in order to optimize the magnetic parameters of the material. The composite of $SrFe_{10}Al_2O_{19}$/ $CoFe_2O_4$ showed the enhancement of saturation and remanent magnetization and a decrease of coercivity with the addition of soft magnetic $CoFe_2O_4$ phase.
Thus, it is not absolute that the particular dopant or composition is always superior, but rather the magnetic composition should be chosen depending on the application's needs.

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