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Evaluation of magnetocaloric material optimization and magnetic field design for magnetic refrigeration systems

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Abstract

Magnetic refrigeration has emerged as a promising alternative to vapor-compression systems owing to its refrigerant-free operation and high theoretical efficiency based on the magnetocaloric effect (MCE). Recent research has focused on advancing both material-level and system-level aspects of the active magnetic regenerator (AMR) cycle. On the material side, multilayer AMRs composed of magnetocaloric materials with graded Curie temperatures have proven effective in extending the achievable temperature span. On the system side, various magnetic field configurations, including electromagnet-based, rotating permanent magnet, and reciprocating linear designs, exhibit distinct trade-offs in terms of field strength, controllability, compactness, and energy efficiency. While high magnetic fields maximize the intrinsic magnetocaloric response, system-level performance is increasingly constrained by mechanical losses, parasitic power consumption, and irreversible processes. Accordingly, this review emphasizes that future progress should prioritize efficient magnetic circuit design under moderate magnetic fields and the mitigation of irreversible losses such as heat transfer limitations and flow non-uniformity, rather than solely enhancing the reversible magnetocaloric effect. By synthesizing recent advances from a system-oriented perspective, this work highlights key design trade-offs and outlines research directions critical for the scalable, high-COP commercialization of magnetic refrigeration technologies.

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1. Introduction

According to the Montreal Protocol and its Kigali Amendment, increasingly stringent refrigerant regulations have intensified global interest in heat pump technologies that operate without conventional refrigerants. Among the limited refrigerant-free alternatives, magnetic refrigeration has emerged as a promising option, as it exploits the magnetocaloric effect (MCE) of solid materials to produce cooling. Because the MCE is a reversible thermodynamic process, magnetic refrigeration offers a higher theoretical coefficient of performance (COP) than conventional vapor-compression systems. Accordingly, extensive research has been devoted to improving its cooling capacity and efficiency. This paper reviews and compares recent advances in magnetic refrigeration technologies and discusses key system-level challenges and future directions toward practical commercialization.

2. Recent Advances and Comparative Evaluation

In recent years, significant progress has been made in both material-level and system-level aspects of magnetic refrigeration. The following subsections summarize and compare representative developments in three key areas: (1) optimization of magnetocaloric material structures and (2) magnetic field design.

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2.1. Magnetocaloric material optimization

The magnetocaloric effect (MCE) is the fundamental mechanism of the active magnetic regenerator (AMR) cycle and constitutes a solid-state caloric process that operates without conventional refrigerants. It originates from changes in magnetic entropy induced by the application or removal of an external magnetic field: adiabatic magnetization decreases magnetic entropy and raises temperature, while demagnetization reverses this process to produce cooling. The magnitude of this entropy change is an intrinsic material property and is typically maximized near the Curie temperature due to strong spin–phonon coupling.

The magnetocaloric performance of a material is characterized by two key parameters: the magnetic entropy change (ΔS_m) and the adiabatic temperature change (ΔT_{ad}). ΔS_m represents the variation in spin disorder caused by the application or removal of an external magnetic field, indicating the amount of entropy transferred between the magnetic and lattice subsystems. In contrast, ΔT_{ad} denotes the resulting temperature shift under adiabatic conditions and directly determines the temperature swing achievable during operation. The two quantities are related through the material's heat capacity (C_p) as Eq. (1).

$$\Delta T_{ad} = -\int_0^H \frac{T}{C_p}\left(\frac{\partial M}{\partial T}\right)_H dH \tag{1}$$

A large ΔS_m reflects the material's thermodynamic potential for heat transfer, while ΔT_{ad} governs its practical temperature response. Since the specific cooling power (Q_c) of an active magnetic regenerator depends on the product of ΔS_m, the operating temperature, and the system efficiency, ΔS_m can be regarded as the intrinsic indicator of the potential cooling capacity. However, the actual cooling output is also strongly influenced by factors such as heat capacity, flow rate, porosity, and cycle frequency. Therefore, ΔS_m represents the latent caloric capability, whereas ΔT_{ad} determines the effective temperature span, and both must be optimized simultaneously to maximize the overall cooling performance of magnetic refrigeration systems.

Magnetocaloric materials are commonly classified by their magnetic transition type. First-order transitions exhibit discontinuous magnetization changes accompanied by lattice distortion, yielding large ΔS_m and ΔT_{ad} but suffering from thermal and magnetic hysteresis that degrade cyclic stability. In contrast, second-order transitions, such as in Gd, occur smoothly without structural change, resulting in smaller ΔS_m but excellent reversibility. Owing to their higher cooling potential, first-order materials are often preferred, and extensive research has focused on mitigating hysteresis to preserve their strong magnetocaloric effect. Hysteresis originates from phase coexistence and lattice strain during magnetostructural transitions and can be reduced through intrinsic approaches, such as tuning toward the tricritical point, where the transition character shifts toward second order and volume mismatch between phases decreases (Gutfleisch et al., 2016).

Extrinsically, microstructural refinement, grain-size control, and chemical substitution (e.g., Co, Mn, B, or H doping in La–Fe–Si or Mn–Fe–P–Si alloys) relieve interfacial stress and stabilize the transition. Applying external stimuli such as pressure or combined magnetic and mechanical fields can also narrow hysteresis by tuning the transformation pathway [1].

Table 1. Representative magnetocaloric materials by temperature range (0–300 K)[2-9]

Temperature Range (K)	Representative material	Transition type	$T_{(C)}$ (K)
< 20 K	$Gd_3Ga_5O_{12}$ (GGG), Er_3Ni	SOMT	2–20
20–80 K	$DyNi_2$, $HoCo_2$, $ErCo_2$	FOMT	30–80
80–150 K	$GdNi_2$, $DyCo_2$	FOMT	70–140
150–220 K	$(La,Ce)(Fe,Si)_{13}H_x$, $MnFeP_{1-x}As_x$	FOMT	170–210
220–260 K	$La(Fe,Si)_{13}H_x$, $MnFeP_{1-x}Si_x$, Fe_2P	FOMT	230–260
260–295 K	Gd / $La(Fe,Co,Si)_{13}$, $MnAs_{1-x}Sb_x$	SOMT / FOMT	270–295
> 295K	FeRh, $Gd_5Si_2Ge_2$, $MnFeP_{1-x}As_x$, $Ni_2Mn_{1-x}Sn_x$	FOMT	300–320

Table 1 summarizes representative magnetocaloric materials commonly used across different temperature ranges between 0 K and 300 K. They are representative materials commonly used based on several

considerations such as entropy change, cost, and availability. In most studies, these materials are employed with compositional modifications to optimize their properties.

Each magnetocaloric material has a fixed Curie temperature, which limits the achievable temperature span when using a single material in a magnetic refrigerator. Therefore, a multilayer active magnetic regenerator (AMR) is employed, and its configuration plays a crucial role in determining system performance.

The multilayer arrangement can be designed based on either the magnetic entropy change (ΔS_m) or the adiabatic temperature change (ΔT_{ad}). A ΔS_m-based configuration is advantageous for maintaining thermal balance and continuity of heat exchange within the regenerator, while a ΔT_{ad}-based design maximizes the overall temperature span of the system. In practice, ΔS_m is commonly used as the design criterion because it ensures smooth thermal matching between layers and stable regenerative heat transfer during cyclic operation.

Table 2. Representative studies on multilayer Active Magnetic Regenerators (AMR)

No	References	Type	Materials	Layers	Range (K)	Design criterion
[10]	Yuan et al. (2022)	Numerical	La–Fe–Mn–Si–H, Gd	5	270 – 300	ΔS_m
[11]	Teyber et al. (2018)	Experimental	Gd–Tb	2	280-300	ΔS_m
[12]	A. Rowe, A. Tura (2006)	Experimental	Gd,$Gd_{0.74}Tb_{0.26}$,$Gd_{0.85}Er_{0.15}$	3	265-315	ΔS_m
[13]	Navickaitė et al. (2018)	Experimental + Numerical	$La(Fe,Si,Mn)_{13}H_\gamma$ + 2 wt% epoxy	2/5/9	275 – 310	ΔS_m
[14]	Park et al. (2015)	Numerical	$GdNi_2$ / $Dy_{0.85}Er_{0.15}Al_2$ / $Dy_{0.5}Er_{0.5}Al_2$ / $Gd_{0.1}Dy_{0.9}Ni_2$	4	80 – 20	ΔS_m
[15]	Zhang et al. (2017)	Numerical	La–Fe–Mn–Si–H	16	268– 305	ΔS_m
[16]	Maiorino et al. (2021)	Numerical	$La(Fe,Co,Si)_{13}$	4	273 – 305	ΔT_{ad}
[17]	Aprea et al. (2011)	Numerical	Gd-Dy alloys	6	260-300	ΔT_{ad}

Table 2 summarizes studies employing multilayer regenerators and presents the specific configurations used in each case. While the tunability of the Curie temperature via compositional control is a general characteristic of magnetocaloric materials, La-based compounds are preferred because such tuning can be achieved without a significant degradation of magnetocaloric strength or an excessive increase in hysteresis. In addition, their capability for post-synthesis hydrogenation, controllable first-order transition strength, favorable cost-to-performance ratio, and well-established processing routes make La-based materials particularly suitable for practical AMR systems. From an economic perspective, magnetocaloric materials are generally expensive and scarce; therefore, rather than employing a wide variety of materials, most studies focus on adjusting the composition ratios to achieve the desired performance. In summary, magnetocaloric material optimization requires balancing intrinsic thermodynamic potential (ΔS_m, ΔT_{ad}) with extrinsic manufacturability and economic feasibility. Comparative evaluations among first- and second-order materials, ΔS_m- versus ΔT_{ad}-based layering strategies, and compositionally tunable systems such as La–Fe–Si alloys provide critical insights into designing practical, efficient, and scalable magnetic refrigeration systems.

2.2. Magnetic field design

Magnetic circuit design is a key factor governing the efficiency and scalability of magnetic refrigeration systems. While magnetocaloric materials determine the intrinsic entropy change, the magnetic field configuration dictates how effectively this potential is utilized. Recent developments have therefore focused on improving field strength, uniformity, and energy efficiency, enabling more compact and high-performance AMR prototypes. Magnetic field generation methods are commonly classified into three categories: electromagnet-based systems, rotating permanent magnet systems, and reciprocating linear configurations, each presenting distinct trade-offs in controllability, mechanical complexity, and overall coefficient of performance (COP).

Electromagnet-based systems use current-driven coils to generate strong and precisely controllable magnetic fields, often exceeding 2 T. These configurations are primarily employed to investigate the upper limits of the magnetocaloric effect and material performance rather than energy efficiency, as continuous power input and cooling are required for coil operation. Consequently, electromagnets are mainly used in

laboratory-scale studies for material characterization, model validation, and benchmarking under idealized high-field conditions.

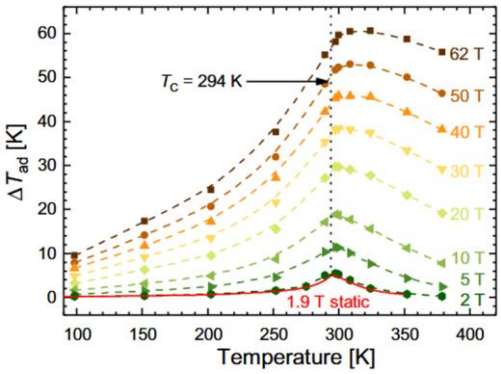

Figure 1. Adiabatic temperature change of gadolinium [18]

Figure 1 shows the adiabatic temperature change under a high magnetic field. When an electromagnet is used, it produces a much larger magnetocaloric effect compared to that generated under a low magnetic field. For example, gadolinium exhibits an adiabatic temperature change of about 60 K at 62 T, which is significantly higher than that obtained using conventional permanent magnets. In addition, the magnetic field can be precisely controlled by adjusting the electric current, making phase control easier. This is the primary reason for employing electromagnets. However, generating such strong magnetic fields requires substantial electrical power. Therefore, electromagnets are typically operated below 5 T, and even in that range, their coefficient of performance (COP) remains low. Consequently, electromagnets are mostly limited to laboratory-scale research applications.

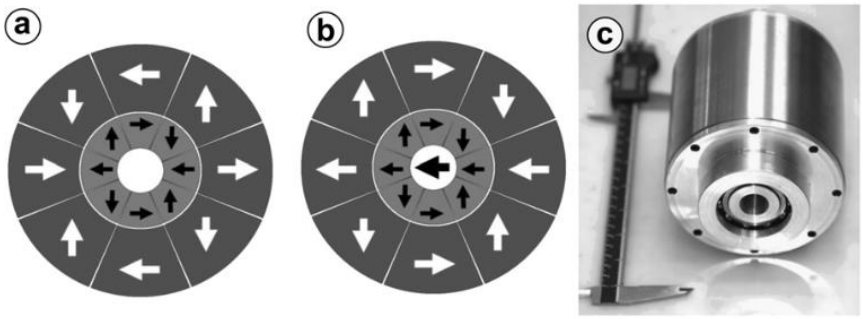

Figure 2. Rotating permanent magnet systems [19]

Rotating permanent magnet systems generate a time-varying magnetic field by mechanically rotating permanent magnets. A representative configuration is shown in Fig 2., where the outer magnet remains stationary while the inner magnet rotates, producing a periodic change in the magnetic field at the center. This rotational mechanism allows the magnetic field to vary without requiring large physical space, offering a compact design advantage. Moreover, since the magnetic field is generated by permanent magnets rather than current-driven coils, the power consumption is significantly lower than that of electromagnet-based systems. However, this approach is limited by the magnetic strength of the permanent magnets, typically achieving a maximum field of about 1.7 T. The major drawback of this configuration is that the minimum magnetic field cannot be reduced to zero, leaving a residual field of approximately 0.05 T. Although several studies have attempted to address this issue, further research is still required to completely eliminate the residual field [20].

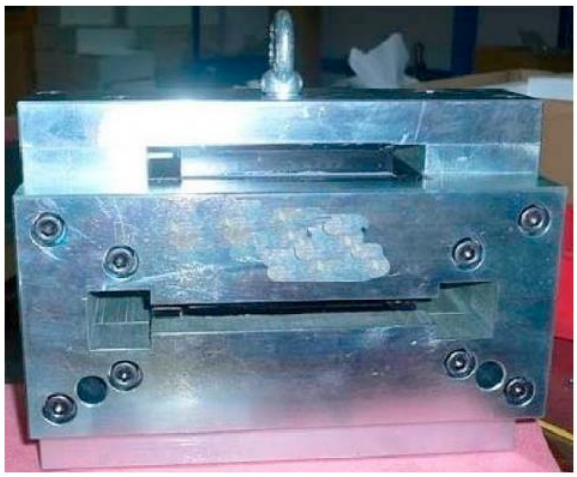

Figure 3. Reciprocating linear magnet [21]

The reciprocating linear magnet system features a relatively simple structure in which the magnetocaloric regenerator moves back and forth through a fixed magnetic field region, undergoing repeated magnetization and demagnetization cycles, as shown in Fig. 3. Unlike rotating systems that rely on flux cancellation, demagnetization is achieved by physically removing the material from the field region, allowing the minimum magnetic field to approach zero and providing superior field modulation. However, the required linear reciprocating motion generally demands more mechanical space than rotating configurations, limiting system compactness in practical applications.

3. Future Outlook and Conclusion

Multilayer active magnetic regenerators (AMRs) based on Curie temperature gradients are an effective approach to broaden the temperature span of magnetic refrigeration systems. However, magnetocaloric materials remain expensive and scarce, and achieving high cooling capacity requires a large quantity of them. Reducing material costs and developing more abundant compositions while maintaining strong magnetocaloric performance are therefore essential steps toward commercialization.

From a system-level perspective, recent studies indicate that simply pursuing higher magnetic field strengths is not necessarily an effective strategy for improving the overall coefficient of performance (COP). While the magnetocaloric effect, quantified by the magnetic entropy change ΔS_m, typically scales with the applied magnetic field as $\Delta S_m \propto B^n$ (with $n \approx 0.6–0.8$ for many practical materials), the mechanical interaction forces and torques in permanent-magnet-based systems scale approximately with B^2. Consequently, higher magnetic fields can lead to disproportionate mechanical losses and parasitic power consumption, ultimately reducing system-level COP. These considerations indicate that future research should emphasize efficient magnetic circuit design under moderate magnetic fields by accounting for the relative permeability of magnetocaloric materials, rather than solely maximizing field strength.

In addition, reversible magnetocaloric cycles and strategies aimed at enhancing the intrinsic magnetocaloric effect have been extensively investigated over the past decades. Consequently, further gains in performance are increasingly limited by irreversible processes at the system level, such as heat transfer resistance, fluid–solid thermal coupling, flow maldistribution, and other parasitic losses within active magnetic regenerators. Addressing these irreversible mechanisms, rather than further amplifying the reversible magnetocaloric effect alone, is expected to play a decisive role in achieving scalable, high-COP magnetic refrigeration systems. Overall, overcoming the combined challenges of material cost, magnetic field efficiency, and irreversible system losses will be critical for the successful commercialization of magnetic refrigeration technology

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