ISSN 2181-8622

Manufacturing technology problems

Scientific and Technical Journal Namangan Institute of Engineering and Technology

INDEX COPERNICUS
INTERNATIONAL

Volume 10
Issue 3
2025

Google Scholar
Crossref doi
SLIB.UZ
Scientific library of Uzbekistan

IV TECHNICAL SCIENCES: MECHANICS AND MECHANICAL ENGINEERING

UDC 537.6+536.2+621.56

STUDY OF THE NATURE AND PROSPECTS OF PRACTICAL APPLICATION OF THE MAGNETOCALORIC EFFECT IN ENERGY-EFFICIENT COOLING SYSTEMS

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Abstract: This article presents the results of the analysis of the magnetocaloric effect (MCE), its physical nature, and the prospects for its application in magnetic cooling systems. Special attention is paid to the thermodynamic relationships describing the change in temperature of magnetic materials during adiabatic magnetization and demagnetization. Materials with giant MKE, including rare-earth magnetics and their alloys, were considered, with an assessment of their suitability for use in household and industrial magnetic refrigeration units. It has been shown that the efficiency of MKE depends on magnetic entropy, heat capacity, and the properties of the working medium. Modern approaches to calculating and experimentally determining MKE are presented, as well as models of magnetic refrigeration machines based on cyclic magnetization and demagnetization of working bodies.

Keywords: magnetocaloric effect, magnetic cooling, magnetic entropy, magnetic material, thermodynamics, magnetic refrigeration machines, rare earth alloys.

Introduction. Creating a compact, environmentally friendly, energy-efficient, and highly reliable refrigerator that operates within the room temperature range is extremely relevant at present. This is due to a number of serious complaints about the currently operating cooling systems. In particular, it is known that during the operation of currently used cooling systems, there may be leaks of working gases (refrigerants), causing serious environmental problems such as the destruction of the ozone layer and global warming [1, 3]. Among the diverse alternative technologies that could be used in refrigeration devices, magnetic cooling technology based on the magnetocaloric effect (MCE) is increasingly attracting the attention of researchers worldwide [1, 2]. Of all magnetothermal phenomena, the magnetocaloric effect (MCE) is of particular interest. On the one hand, the use of this effect allows physicists to study the interactions and changes in magnetic structures in magnets [1, 4], and on the other hand, on the practical side, recently, the possibilities of using MKE to create magnetic coolers and magneto-thermal pumps have emerged [2, 3].

Materials and methods. Both theoretical and comparative-analytical methods were used in the research process. The theoretical part of the work is based on the analysis of scientific literature devoted to magnetic phase transitions and the magnetocaloric effect (MCE) in various classes of magnetic materials, including rare-earth elements, intermetallic compounds, and Heusler-type alloys. Both classical approaches based on thermodynamic equations and modern methods for evaluating MKE, including direct and indirect measurements, were studied.

The following methods were used to assess the magnitude of the magnetocaloric effect:

Direct method based on measuring the change in sample temperature when the magnetic field is adiabatically turned on and off.

Indirect method, including the calculation of magnetic entropy change by magnetization isotherms using a thermodynamic relation: $\Delta S_M = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH.$

Thermodynamic analysis methods were also used, including the entropy model, which allows us to consider the interaction of magnetic moments subsystems and the crystal lattice. The calculations were based on the equations of the first law of thermodynamics, taking into account exchange energy, Zeeman interaction, and magnetic anisotropy.

Experimental data and comparisons were based on previously published results in scientific sources, as well as measurements performed using the MagEq MMS SV3 automated installation for magnetic entropy and heat capacity analysis.

II. Results and discussion. Magnetic-caloric effect is the change in the temperature of a magnetic material when the magnetic field is quickly (adiabatically) turned on and off.

The magnetic refrigerator is advantageously distinguished from traditional ones by its energy efficiency, reliability, environmental friendliness, and safety for humans. In these devices, it is not required to use, for example, chlorine-fluorine-carbon (CFC), which is considered to be responsible for the greenhouse effect, destruction of the ozone layer, and is also extremely harmful to humans when leaking near an open fire [4].

The magnitude of MKE and, consequently, the efficiency of the cooling process in a magnetic refrigerator are determined by the properties of the magnetic working bodies. Efficient alloys for the working bodies of magnetic refrigerators are being researched by numerous scientific collectives worldwide [1, 3]. In many works, numerous possible combinations of rare earth and magnetic metals and other materials have been analyzed from the perspective of finding optimal alloys for implementing magnetic cooling in various temperature ranges. Among the studied materials with high magnetic-caloric properties, the compound (iron alloy with rhodium) has the highest specific (per unit magnetic field) magnetic-caloric effect. The value of the specific MKE for this compound is several times greater than for other compounds. This alloy cannot be used in practice due to the high cost of rhodium, as well as significant hysteresis effects in it, however, it can serve as a unique standard with which the magnetocaloric properties of the studied materials should be compared [4, 5].

After Pecharsky and Gschneidner discovered a giant MKE in

$$Gd_5(Si_xGe_{1-x})_4 \qquad (1),$$

it turned out that other materials also show a giant MKE associated with a magnetic phase transition:

$$MnCoGeV_{0.02} \qquad (2),$$

$$Mn_{0.96}Cg_{0.04}CoGe \qquad (3),$$

$$Ni_{2.15}Mn_{0.85}Ga \qquad (4),$$

$$DyNiAI \qquad \text{(5) and others.}$$

The great advantage of the presented compounds, compared to the reference material Gd, is the possibility of obtaining a transition temperature in a wide range near room temperature.

However, none of the above-mentioned connections can be used for designing household refrigerators due to the need for high fields or high applied external pressure, which leads to a complication of the design and an increase in the size and cost of the final device [5, 6].

Finding the optimal connection for constructing magnetic refrigerators, as well as improving the experimental equipment for studying MKE, is a pressing task today [3, 7, 8].

This effect is determined by the direct method - by the change in ΔT of the substance's temperature (ΔT-effect), and by the indirect method - by the change in magnetization from the isotherms.

$$dW = Vd\left(\frac{1}{2}H^2\right) + HdM \qquad (6)$$

Let us consider a ferromagnet [6, 9], located in the field H of a solenoid. If the field is increased by dN,, then its magnetization increases by dM. The energy required for such an increase (at T=const) is the energy created by an external source of electrical energy: where V- is the volume of the body.

The first term of the right side corresponds to the work done to increase the field strength by a certain amount. This work is performed regardless of the presence of a magnet in the solenoid [5, 8]. The second term is the energy required to increase the magnetization of a substance:

$$dW = HdM \tag{7}$$

The sign of NdM is positive, similar to the work sign PdV (under the influence of H, the orderedness of magnetic moments increases, and under the influence of pressure, the volume of the body decreases; an increase in magnetization by dM and a decrease in volume by dV are thermodynamically equivalent processes).

Taking into account that $dSm = dQ/T$, according to the first law of thermodynamics,

$$dU = dQ + HdM = TdS_M + HdM \tag{8}$$

$$S = -H\left(\frac{dM}{dT}\right)$$

Substituting here $S_M = C_{PH}$ - we find

$$\Delta T = -\frac{T}{C_{P.H.}}\left(\frac{dM}{dT}\right)_H \Delta H \tag{9}$$

$\left(\frac{dM}{dT}\right)_H$- This is one of the most used thermodynamic relationships for the magnetocaloric effect. It is applicable both for paramagnetics and for magnetically ordered substances: ferro-, ferri-, and antiferromagnetics (in the region, the para-process decreases with increasing temperature), therefore, the DT effect at the rapid activation of the DN field has a positive sign (the magnetic temperature increases) [5, 8]. When the field is quickly switched off, i.e., when the sample is demagnetized, it has a negative sign (the temperature decreases).

From relation (8) follows another thermodynamic relation for the magnetocaloric effect. Since dU is a complete differential, then from (8) it follows:

$$\left(\frac{\partial U}{\partial S_M}\right)_H = \left(\frac{\partial U}{\partial M}\right)_{S_M} = H \tag{10}$$

Differentiating the first expression by M and the second expression by SM, we get

$$\Delta Q = T\left(\frac{\partial H}{\partial T}\right) \tag{11}$$

$dS_M = \frac{dQ}{T}$Substituting , we get

$$\Delta T = \frac{T}{C_{P.t.}}\left(\frac{\partial H}{\partial T}\right)_M \Delta M \tag{12}$$

These expressions show that this MKE is directly related to the change in magnetization DM, which creates a field momentum DN.

In the thermodynamic consideration of MKE, the entropy interpretation is useful, allowing for a better understanding of the physical essence of the phenomenon [9]. It arises from the condition of adiabaticity of the process and the idea that the magnetic material consists of two subsystems: a subsystem of magnetic moments with an entropy SM and a subsystem of lattice atoms with an entropy Sresolution. In this case, the condition for the adiabatic nature of the magnetization process is written as follows:

$$dS = d\left(S_M + S_{pesh}\right) = 0 \tag{13}$$

$$S = \left(S_M + S_{pesh}\right) = const \text{ i.e.} \tag{14}$$

Let's consider the adiabatic inclusion of the magnetic field. When the field is applied, the subsystem of moments is ordered, its entropy decreases, and an excess of energy is released, which is transferred to the subsystem of lattice atoms in the form of heat and raises its temperature by ΔT. A positive DT effect occurs.

When the magnetic field is adiabatically switched off, the substance becomes demagnetized, i.e., the process of breaking the magnetic order (demagnetization) in the subsystem of moments requires energy supplied by the subsystem of lattice atoms, which lowers its temperature. Thus, during the adiabatic magnetization and demagnetization of a substance, a reversible transfer of entropy, and consequently, heat, occurs from the magnetic subsystem to the lattice subsystem and back [7, 9].

The change in the magnetic part of entropy S_M can be determined by the formula:

$$\Delta S_M = S_M\,(H) + \ S_M(0) \tag{15}$$

Usually, MKE is determined by the change in sample temperature when the magnetic field is applied, or is calculated from the isotherms of magnetization's dependence on magnetic field magnitude based on the change in entropy DS [8, 9]. From thermodynamic considerations, formula (15) is derived, which connects the change in temperature ΔT with the derivative dM and the change in the field.

Change in entropy at constant magnetic field:

$$\Delta S_H = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH \qquad (16).$$

The relationship between the DT effect and the change in dS calculated from the magnetization isotherms can be obtained using formulas (15) and (16):

$$\Delta T = -\frac{T}{C_{p.H.}}\,\Delta\, S_M \tag{17}$$

The method of calculating MCE based on isotherms is indirect and often leads to erroneous conclusions. Moreover, to implement this method, it is necessary to conduct measurements of the temperature dependence of the material's heat capacity.

A more general thermodynamic analysis showed that MKE can be represented from four contributions related to exchange and magnetoelastic energies, magnetic anisotropy energy, and Zeeman energy [4, 9]:

$$\Delta T = -\frac{T}{C_{p.H.}}\left(\frac{\partial \Delta E_{\text{об.м}}}{\partial T} + \frac{\partial \Delta E_{\text{му}}}{\partial T} + \frac{\partial \Delta E_A}{\partial T} - H\frac{\partial \Delta M}{\partial T}\right) \tag{18}$$

$\Delta E_{\text{об.м}} \Delta E_{\text{му}} \Delta E_A H \frac{\partial \Delta M}{\partial T}$ where, - exchange interaction energy, - magnetoelastic energy, - magnetic energy - addend describing Zeeman energy (energy of magnetization interaction with magnetic field).

The effectiveness of magnetic cooling depends not only on the MKE value but also on the heat capacity value and the magnitude of the change in the magnetic part of entropy under the influence of the magnetic field [8, 9]. The magnitude of the lattice part of the entropy s_p, which increases significantly when heated, also plays a large role here, as a result of which magnetic coolers using paramagnetics as working bodies are ineffective at T > 20 K. At elevated temperatures, magnetically ordered substances are more effective as working bodies, in which large MKE arise in the region of magnetic phase transitions. In recent years, there has been an interest in creating new types of magnetic refrigeration machines (MCHM) based on the use of MKE. In this case, it is proposed to use rare-earth magnetics, Heissler alloys, manganese arsenide MnAs, compounds Gd5 (Si2Ge2), RCo2, La (Fe,Si) 13, etc., as working bodies, possessing a high MKE and a change in magnetic entropy in the temperature intervals convenient for operating such machines [9] (Fig.1.).

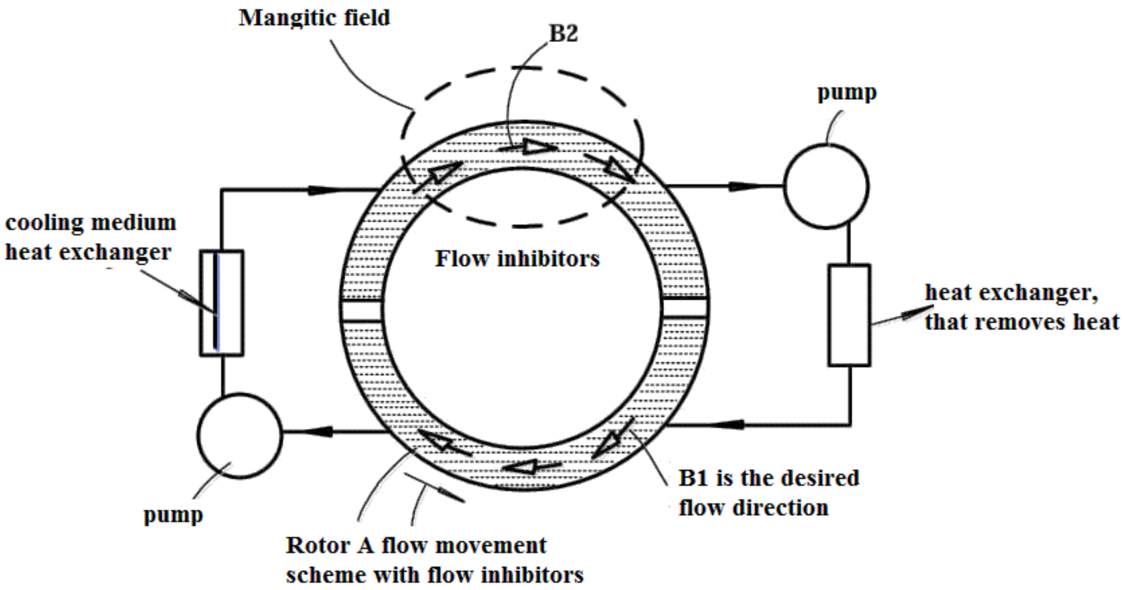

Figure.1. Magnetic refrigeration machine diagram

In one of the MXM designs, the solid working fluid - the magnetic fluid - cyclically moves between the receiver and the heat source (the cooled fluid) [7, 9]. In the zone of strong magnetic field, the working body is isothermally magnetized, and the heat released in the working body due to MKE is transferred to the heat receiver. In the zone where there is no magnetic field, the working body is demagnetized, resulting in a decrease in the working body's temperature and heat transfer to it from the heat source - the cooled body. After establishing equilibrium, the cycle is repeated. Thus, the MKE ensures the operation of the magneto-thermal pump, which "pumps" heat from the cooled body [8, 9]. Another promising model of a magnetic refrigeration machine is a device in which a liquid with a filler in the form of magnetic particles with a large MCE is pumped through a region where a strong magnetic field is created.

III. Conclusion. The analysis showed that the magnetocaloric effect (MCE) is a promising physical phenomenon for implementing environmentally safe and energy-efficient cooling systems, especially in the room temperature range. Based on the considered theoretical and experimental data, the following was established:

1. The magnetocaloric effect is closely related to the change in magnetic entropy and depends on the properties of the magnetic material, its heat capacity, and the nature of the magnetic phase transition.
2. Materials with giant MKE, such as Gd5 (SixGe1−x) 4, Mn-based alloys, and rare earth-based compounds, are of greatest interest for practical application.

3. Despite the high indicators of individual compounds, many of them are not yet suitable for mass use due to the high cost of components (for example, rhodium), hysteresis losses, or the need to create strong magnetic fields.
4. Both direct (measurement of ΔT) and indirect (calculation of entropy change by magnetization isotherms) methods are used to evaluate MKE, however, the most reliable results are achieved with a comprehensive approach.
5. A promising direction is the development of new magnetic materials and optimization of the designs of magnetic refrigeration machines, including using liquid working bodies with magnetic nanoparticles.

Thus, further research in the field of ICE should be aimed at creating affordable and efficient materials, as well as improving measurement methods and heat engineering designs that utilize this effect.

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