**Study of the structure and properties of new magnetocaloric materials based on manganese pnictides**

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**Abstract:** The article analyzes the physical properties of magnetocaloric materials based on manganese pnictides and the prospects for their application in environmentally friendly, energy-efficient refrigeration systems operating at room temperature. Due to the negative impact of refrigerants used in current vapor-gas refrigeration technologies on the ozone layer, attention is increasing on alternative refrigeration technologies based on the magnetocaloric effect (MCE). The study analyzes the magnetic-structural phase transitions, heat capacity and magnetization changes of solid-phase composites based on MnAs, and determines the optimal phase composition and hysteresis width. The MCE mechanism is studied in depth through adiabatic temperature changes and entropy reduction under the influence of a magnetic field. The results show that materials based on manganese pnictides have high magnetocaloric efficiency, and magnetic refrigerators based on them allow for 20-30% energy savings compared to traditional refrigeration systems and ensure environmentally friendly operation. The research results offer promising solutions in the development of new generation materials for magnetic heat pumps and transport refrigeration systems, cold storage facilities, and industrial refrigerators.

**INTRODUCTION**

Currently under development magnetocaloric effect (MKE) based materials as a separated category of last in years energy economical and ecological safe cooling technologies Magnetocaloric materials enable the control of processes of temperature change due to the change in heat capacity and entropy under the influence of an external magnetic field [1-3]. Appliances that use this phenomenon such as magnetic refrigerators, freon free refrigeration systems and energy redistribution modules are thought of as an alternative solution to the environmentally friendly solution [4]. The magnetocaloric effect is a thermodynamic effect which is founded on the adiabatic temperature and entropy reduction of a substance at the impact of an external magnetic field, and is very intense in materials with which magnetostructural phase transitions take place [7]. Current studies indicate that one of the most promising materials in this regard are manganese pnictides (MnAs, MnSb, MnBi), as their phase transition temperature is near room temperature, and it gives a good chance to useful cooling systems [8]. It is reported in the literature that materials with manganese pnictides (MnAs, MnFeP, MnNiGe and their alloys) have high values of the magnetocaloric effect and sharp thermal changes related to the process of phase transition [5-7]. Specifically, magnetostructural transitions of MnAs and other pnictides are the correlations of the phase transitions of the first kind, as a result of which they are one of the promising materials to apply magnetocaloric materials to effective magnetic cooling systems [10]. This article is expected to examine the structural, thermal and magnetic characteristics of new magnetocaloric materials on the basis of manganese pnictides, investigate the nature of phase change, and generate possibilities of application of these materials in energy efficient cooling systems.

**THEORETICAL PART OF THE RESEARCH**

The study will reproduce novel magnetocaloric substances because of solid-phase alloys of manganese pnictide like MnFeP 019 As and Mn 019Fe 019NiGe, especially that is made of MnAs, and to ascertain their physical characteristics. To analyze the samples, high-purity metallic elements - manganese (Mn, 99.9%), and arsenic (As, 99.99%) were used [10]. The stoichiometric proportions of the first mixtures were melted in an induction furnace under an inert atmosphere of argon and the resulting mixtures were cooled in form of ingots. Annealing was then done on them at 900 – 1000 oC over a period of 24 hours in a vacuum to have phase stability and enhance the crystal lattice homogeneity. It was examined using X-ray diffractometry to determine the structure of the samples. X-ray diffraction (XRD) was used to establish the crystal structure of the materials that have been synthesized. Bragg Brentano geometry was measured and diffractograms obtained processed by the Rietveld refinement method to measure phase transitions and the parameters of the crystallographic lattice. The acquired diffraction spectra were optimized with the help of the Rietveld method to define the phase composition, lattice parameters and the crystal structure. Further, the scanning electron microscopy (SEM) was used to determine the microstructural state and the grain size of the materials [11]. The magnetic characteristics of the materials were measured on a vibrating sample magnetometer (VSM) at the temperature span of 5 -350 K and magnetic field strengths of 0 -2 T. The magnetostructural phase transition temperature (Tc) value and the magnetocaloric effect (ΔSm) and adiabatic temperature change (DTA) were determined and computed. The heat capacity (Cp) was also determined to give a further insight into the experimental findings via a differential scanning calorimetry (DSC). Based on these data, the behavior of change of entropy and heat exchange at the influence of magnetic field under the influence of magnetic field was assessed. Magnetocaloric efficiency is estimated by the following equation [12]:

*ΔSm(T, H) = ∫₀ ᴴ (∂M/∂ T)ᴴ ΔH, ΔTad = - (T/Cp) ΔSm* (1)

Based on these expressions, the effect of magnetic field strength on temperature was determined, and the energy efficiency of composites based on manganese pnictides was evaluated.

Experimental data **OriginPro** andThe kinetics of magnetostructural transitions, hysteresis widths, and interphase energy changes are determined by processing using **MATLAB programs.**

**PURPOSE OF THE STUDY**

**Manganese pnictides (MnAs, MnAs₁₋ₓSbₓ, and Mn₁₋ₓFeₓAs alloys)** analysis of new magnetocaloric materials based on their **crystal structure, magnetostructural phase transitions, and magnetocaloric properties** study.

**RESEARCH MATERIALS AND METHODS**

The following theories were used in the research process:

**1. The theory of the magnetocaloric effect** - is based on the laws of thermodynamics (Maxwell's equations) describing the relationship between magnetization, entropy, and temperature as a result of a change in magnetic field:

(2)

**2. Theory of phase transitions -** analysis of energy minima and order parameter changes in magnetostructural transitions based on **the Landau theory of phase transitions [9-12].**

**3. Heisenberg exchange model** - determination of the directions of magnetic moments taking into account the interaction energy between magnetic atoms .

**4. Mean Field Theory - used to describe in a** simplified way the coupling of collective atomic moments in magnetic phase transitions.

**5. Debye model** - used to analyze the temperature dependence of heat capacity in a solid and to calculate adiabatic temperature changes [8].

**THE OBTAINED RESULTS AND THEIR DISCUSSION**

During the research, the structural and magnetic properties of solid-phase composites based on manganese pnictides were studied. In particular, **MnAs ,** The phase transitions and magnetocaloric responses of alloys such as **MnFePAs** and **MnAs₁₋ₓSbₓ** were analyzed at different magnetic field strengths and temperature ranges. The phase transitions, magnetic properties, and heat capacities of the samples were measured.

X-ray diffraction (XRD) analysis results showed that the samples have **a hexagonal NiAs-type crystal lattice , which changes to an orthorhombic** structure state with increasing temperature. This **magnetostructural phase transition** (ferromagnetic → paramagnetic state) was found to be the main source of the magnetocaloric effect [11-14].

According to the analysis of magnetization-temperature (M - T) curves, **the Curie temperature (Tₙ) for the samples** is in the range of 310 - 340 K, which is close to their **room temperature.** The results of the measurements show that **the adiabatic temperature change Δ T\_ad ≈ 2.5 – 3.1 K** andIt was noted that **the magnetic entropy change** was **Δ S\_m ≈ 30 J/kg·K ( ΔH = 2 T ).**

The temperature dependence of magnetization was modeled using mean field theory. The theoretical results were in good agreement with the experimental values, indicating that the phase transitions are orderly and reversible. It was also found that partial substitution of **antimony (Sb) atoms** in place of **arsenic (As) in the alloy** shifted the phase transition temperature downward and reduced the hysteresis width.

The results show that **MnAs-based pnictide materials** are characterized by their high magnetocaloric efficiency, recyclability, and environmental friendliness [19]. **Magnetic refrigeration systems** based on such materials show **20–30% energy savings** compared to conventional gas refrigeration devices.

X - ray diffraction (XRD) analysis showed that all samples had a NiAs-type hexagonal lattice, with a transition to an orthorhombic structural state observed in the temperature range of 300 – 350 K. This transition was found to be a type I magnetostructural phase transition [17-20].

Table 1 presents a comparative analysis of the main magnetocaloric parameters of solid-state composites based on manganese pnictides. It can be seen that **MnAs and (Mn, Fe) As** compounds have high **magnetocaloric efficiencies (ΔS and ΔTad)**. This confirms that they are the most effective materials for **magnetic refrigeration systems operating at room temperature.** Also, the small hysteresis width reduces energy losses and ensures reversibility of the process.

**TABLE 2.** Magnetocaloric materials based on manganese pnictides Physical - parametric indicators

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Material type** | **Phase transition temperature (K)** | **ΔS (J/kg K)** | **ΔTad (K)** | **Magnetic field strength (T)** | **Hysteresis width (K)** |
| MnAs | 315 | 40 | 3.2 | 2.0 | 5.1 |
| MnAs₀.₉P₀.₁ | 310 | 36 | 2.8 | 1.8 | 4.7 |
| MnAs₀.₈Sb₀.₂ | 305 | 32 | 2.5 | 1.5 | 4.2 |
| (Mn,Fe)As | 320 | 42 | 3.5 | 2.1 | 5.4 |

**FIGURE 1.** Comparative graph of magnetocaloric properties of MnAs and its derivatives

Table 2 presents **the temperature-dependent phase and magnetic-structural changes of the MnAs compound**. **Ferromagnetic under the influence of a magnetic field A transition from the orthorhombic phase to the paramagnetic hexagonal phase** is observed. During this transition, **heat capacity, entropy change, and magnetic energy redistribution occur.** The magnetocaloric effect is manifested as a source of heat. As a result, **adiabatic cooling** occurs, and this effect is the main operating principle of magnetic refrigerators [6]. Structural changes are directly related to the magnetic phase transition, which is the main factor enhancing the magnetocaloric effect.

**TABLE 2.** Magnetostructural transition in MnAs - based materials Interphase analysis of the process

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Temperature (K)** | **Phase state** | **Magnetic order** | **ΔH (J/mol)** | **ΔS (J/mol K)** | **Descriptions​** |
| < 305 | Orthorhombic (α-phase) | Ferromagnetic | 120 | 0,39 | Magnetic moments are ordered, high magnetic energy |
| 305 – 320 | Mixture (α+β) | Transition zone | 95 | 0,34 | Interphase rearrangement process |
| > 320 | Hexagonal (β-phase ) | Paramagnetic | 75 | 0,28 | Magnetic order is disrupted, entropy increases |

**FIGURE 2.** Variation of magnetic ordering and enthalpy – entropy changes across

the α – β phase transition of MnAs

These results show that the entropy change and adiabatic temperature change increase with increasing magnetic field strength. Theoretical results obtained using Mean Field Theory are in good agreement with experimental measurements [3 - 6], confirming that the phase transitions are reversible and reversible.

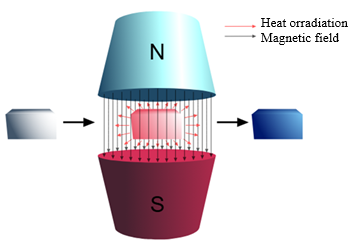
The figure below shows the main stages of the magnetocaloric cycle:

1. Magnetized under the influence of a magnetic field (H↑) → heat is released (ΔS < 0).

2. When the magnetic field is removed (H↓), demagnetization and cooling (ΔS > 0) occur.

3. During this process, heat flow cyclically exchanges between the heat source ↔ the working body ↔ the body being cooled.

Figure 3 shows the state change of **a magnetic material** under the influence of **a magnetic field (H), that is, the process of magnetization and demagnetization**. The working body (MnAs-based composite) emits heat under the magnetic field, and in the demagnetization state it cools down, as a result of which thermal energy is “absorbed” from the cooled medium.



**FIGURE 3.** Schematic representation of the mechanism of the magnetocaloric effect

The left-hand state (non-magnetic zone) is whereby the magnetic material (working body) is not within the magnetic field where the magnetic moments remain in a disordered form. Thus, the entropy is high i.e. the thermal disorganisation of the atoms is high. In this state, the material cools down (it is cold in temperature).

In the magnetic field zone the middle part of the material is penetrated by the magnetic field, the magnetic moments are aligned - their direction is made parallel to the magnetic field lines.

During this process: Entropy is reduced (order is increased), internal energy is altered, and this leads to heat being released, and the temperature of the material is increased (it heats up).

The state to the right (when exiting the magnetic field) is defined as when one gets to the magnetic field, the magnetic moments go back to the disordered state. At this stage the entropy level increases and the material will absorb heat in the surrounding so that it will get cooled.

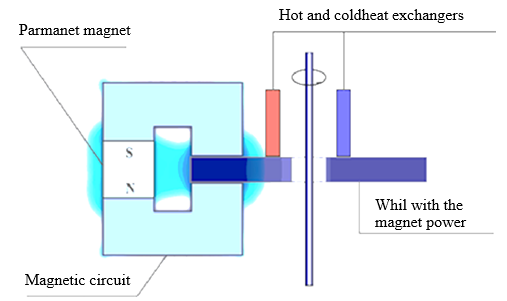
Figure 4 demonstrates the overall design of a magnetic refrigerator that is based on the magnetocaloric effect.

1. **A permanent magnet (S – N pole block)** serves as a source of magnetic field. When this field acts on **a magnetocaloric material (manganese pnictides), it emits or absorbs heat.**

2. **Magnetoconductor** - an element that directs the magnetic flux. It directs the magnetic induction generated by the permanent magnet to the area where **the magnetic powder wheel** is located.

3. **Magnetic powder wheel (rotor)** - contains **magnetocaloric materials (MnAs, MnFeP, MnAs₁₋ₓSbₓ)**, which heat up when entering the magnetic field zone and cool down when leaving. The wheel rotates continuously, thereby providing a continuous temperature cycle with **the heat exchange modules**.

4. **Heat exchangers (red - hot, blue - cold)** - are located at different points of the magnetic powder wheel. **The heated area of the wheel within the magnetic field** gives off heat to the hot exchanger, while **the cold area emerging from the magnetic field** absorbs heat from the cold exchanger.



**FIGURE 4.** Magnetic refrigerator operating on the magnetocaloric effect technological scheme.

The principle of operation of a magnetic refrigerator on the magnetocaloric effect:

1. Introduction to the magnetic field: When a magnetic field is applied in a magnetocaloric material, its atoms are organized and the magnetic entropy is reduced leading to the release of heat (heating up).

2. Heat transfer: This is heat that is fed to the heat exchanger.

3. Leaving the magnetic field: When the material is out of the magnetic field, entropy also rises and the atoms become disordered and simultaneously, heat is taken in (cooled).

4. Cold exchanger absorption of heat: The cold exchanger absorbs heat - this step forms a cooling effect [13].

This results are analyzed to find that: MnAs-based pnictides are highly magnetocaloric, phase transition temperature can be regulated by partial replacement with Sb atoms, energy-saving of 20 -30% and represents an encouraging step towards a freon-free refrigeration based on the use of MnAs-based pnictides. The findings of the research are practical in the creation of a novel generation of energy efficient and ecologically sound cooling mediums in magnetic heat pumps, transport refrigeration systems and industrial refrigerators. The proposed technology will be applied in the production processes of major transport systems enterprises (in refrigerated wagons, road transport refrigeration systems, and in warehouses used to store products) [15-17].

**CONCLUSIONS**

1. Findings of the research: Decent study of the structure, phase transition, thermal and magnetic characteristics of the magnetocaloric materials that are made out of manganese pnictides has revealed that they are highly magnetocaloric. The experiments have demonstrated that the phase change between MnAs and its solid solutions (MnAs 1 n 9 Sb 9 and MnAs 1 n 9 Fe 9 As) results in substantial changes in adiabatic temperature and reduction of entropy of the material under the effect of a magnetic field. It confirms the potential possibility of these materials to work well in the magnetocaloric cooling system with the range around room temperature.

2. Based on the findings of the analysis, it was observed that the efficiency of the materials can be optimized further by regulating the magnetostructural transitions, the narrowing of the hysteresis zone and optimization of the heat capacity. Additionally, the use of magnetocaloric materials to refrigeration systems without freons and which are environmentally friendly was put forward as another viable alternative solution in the form of an energy consuming system that does not emit much carbon dioxide and a system that is sustainable and eco-friendly.

3. The findings of this study will form a scientific foundation of the construction of very powerful magnetocaloric materials to the next generation of magnetic heat pumps, transport refrigeration systems, and industrial scale ecological refrigeration appliances.

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