VSB TECHNICAL UNIVERSITY OF OSTRAVA | FACULTY OF ELECTRICAL ENGINEERING AND COMPUTER SCIENCE | DEPARTMENT OF PHYSICS

# INFLUENCE OF ANNEALING TEMPERATURE ON MAGNETISM AND DEFECTS IN $Mn_{2.4}Fe_{0.8}Al_{0.8}$ ALLOYS

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## ABSTRACT

Manganese-based alloys with the composition $Mn_2FeZ$, Z = Si, Al, have been widely investigated in recent years due to their potential applications in spintronics. $Mn_2FeSi$ alloy already prepared in the form of ingots, thin films, powders or ribbons exhibits a cubic full-Heusler ($L2_1$) or inverse-Heusler (XA) structure or combination of both. Contrary, the $Mn_2FeAl$ alloy has so far only been prepared in the form of an ingot and owns a primitive cubic ($\beta$-Mn type) structure. Present investigations are devoted to the $Mn_{2.4}Fe_{0.8}Al_{0.8}$ ingots prepared by induction melting technique and additionally annealed for 100 hours at 500 °C and 800 °C in the argon protective atmosphere. Their microstructural, defect, and magnetic properties are studied in dependence on annealing temperature and compared to the $Mn_2FeAl$ alloys. The scanning electron microscopy confirmed single phase alloys with compositions slightly different from the nominal one. The X-ray diffraction analysis revealed the origin of $\beta$-Mn structure with the lattice parameter of 0.6359(2) nm well comparable with the $Mn_2FeAl$ value of 0.6339(1) nm. Results of positron annihilation spectroscopy showed that vacancy concentration in prepared ingots is very low. Magnetic properties of $Mn_2FeAl$ alloys showed an antiferromagnetic-paramagnetic transition with the Néel temperature about 36-37 K without essential impact of annealing temperature. Similar magnetic characteristics are observed for $Mn_{2.4}Fe_{0.8}Al_{0.8}$ ingots in as-cast state and after annealing at 500 °C. Contrary, the sample annealed at 800 °C exhibited a weak ferromagnetic contribution at room temperature and its influence increased during cooling down to 5 K.

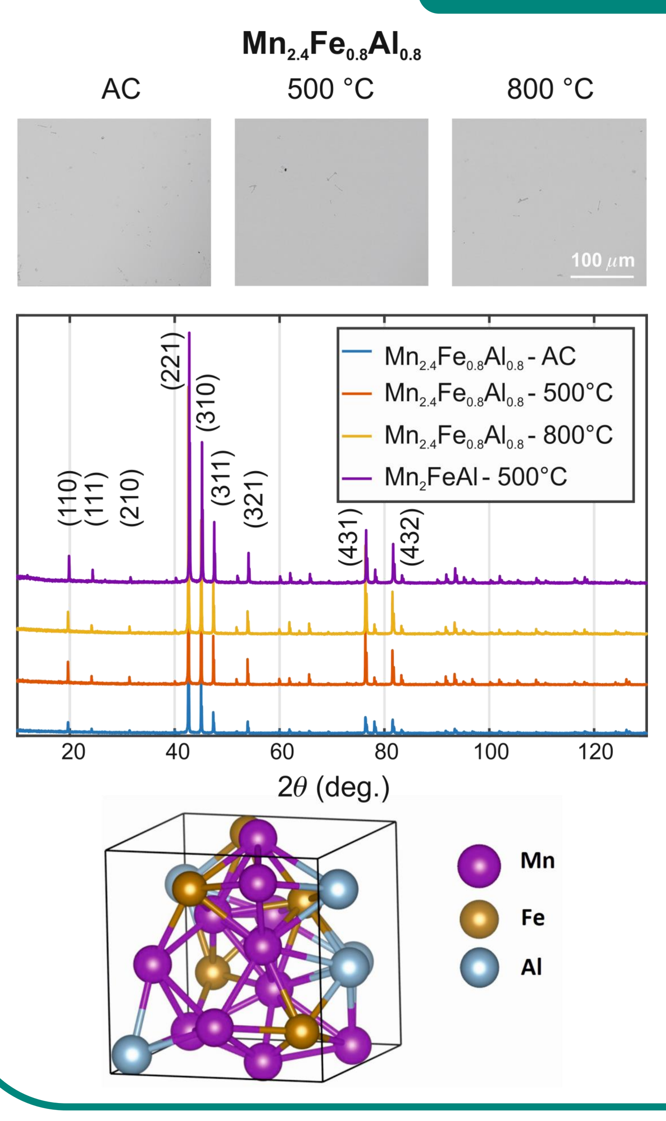
## SAMPLE PREPARATION

- **$Mn_2FeAl$ and $Mn_{2.4}Fe_{0.8}Al_{0.8}$ alloys** prepared from high purity elements (Mn – 99.9%, Fe – 99.95%, Al – 99.9%) using the conventional technology of **induction melting**
- due to the known volatilization of manganese during production and/or subsequent annealing of the high-Mn-alloys, **about 5% Mn more** added into the input compositions
- the induction Supercast Titan furnace (Linn High Therm, Germany) used for production of cylinder-type ingots, each ingot re-melted four times under Ar (6 N) of 90 kPa pressure to guarantee a good chemical homogeneity
- after induction melting the samples slowly cooled down to room temperature inside the furnace, denoted **as-cast or AC**
- the **annealing** performed **at 500 °C and 800 °C for 100 hours** in Ar protective atmosphere using a sintering furnace Xerion Advanced Heating (Ofentechnik, Germany), after annealing the samples slowly cooled down to room temperature inside the furnace

## EXPERIMENTAL TECHNIQUES

- **XRD (X-Ray Diffraction)** – diffractometer AXS D8 Advance (Bruker AXS GmbH, Germany) equipped with the position-sensitive LynxEye detector and Cu Kα radiation, 2θ range 20° - 95°, evaluation – Rietveld structure refinement method
- **SEM (Scanning Electron Microscopy)** – QUANTA 450 FEG (FEI, USA) equipped with an energy dispersive X-ray (EDX) APOLLO X analyzer
- **PAS (Positron Annihilation Spectroscopy)** – positron lifetime spectroscopy (PLT): identification of open volume defects like vacancies or dislocations; coincidence Doppler broadening (CDB): information about local chemical environment of defects; $^{22}$NaCl positron source with an activity of 1 MBq deposited on a 2 μm thick Mylar foil used
- **PPMS (Physical Property Measurement System)** – magnetometer Dynacool (Quantum Design, USA), magnetic properties of alloys at low and room temperatures 4.2 K - 300 K, applied magnetic field up to 4000 kA/m (5 T)
- **VSM (Vibrating Sample Magnetometer)** – magnetometer Microsense EZ 9, high-temperature experiments from room temperature up to 573 K at constant magnetic field 400 kA/m (0.5 T)

## MICROSTRUCTURE

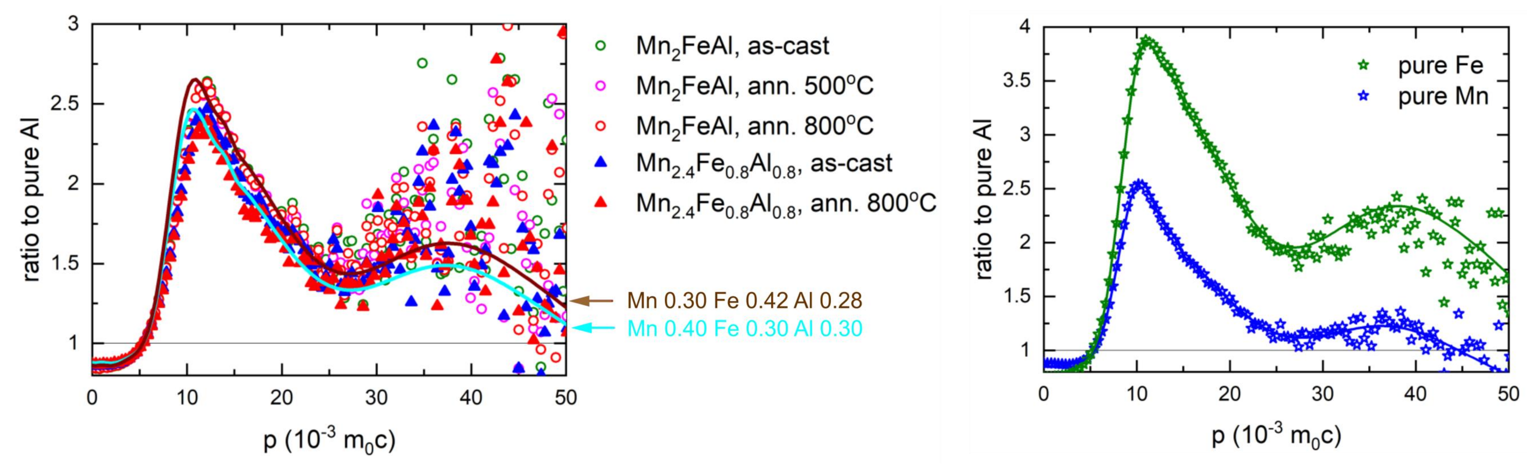



EDX analyses of samples from the area (1 × 1) mm$^2$

| nominal composition / annealing | Mn | | Fe | | Al | |
|---|---|---|---|---|---|---|
| | (wt.%) | (at.%) | (wt.%) | (at.%) | (wt.%) | (at.%) |
| $Mn_{2.4}Fe_{0.8}Al_{0.8}$ - AC | 63.8 | 56.5 | 23.3 | 20.3 | 12.9 | 23.2 |
| $Mn_{2.4}Fe_{0.8}Al_{0.8}$ - 500°C | 63.3 | 56.1 | 23.8 | 20.7 | 12.9 | 23.2 |
| $Mn_{2.4}Fe_{0.8}Al_{0.8}$ - 800°C | 63.9 | 56.7 | 23.8 | 20.7 | 12.3 | 22.6 |
| $Mn_2FeAl$ - AC | 54.5 | 47.4 | 29.8 | 25.3 | 15.7 | 27.3 |
| $Mn_2FeAl$ - 500°C | 54.8 | 47.6 | 30.2 | 25.8 | 15.0 | 26.6 |
| $Mn_2FeAl$ - 800°C | 54.8 | 47.6 | 30.2 | 25.8 | 15.0 | 26.6 |

- SEM back-scattered electron images with additional EDX analyses confirmed single-phase structure of all samples
- samples exhibit the primitive cubic structure of the $\beta$-Mn type (space group $P4_132$)
- obtained lattice parameter of $Mn_{2.4}Fe_{0.8}Al_{0.8}$ alloys, about 0.6359(2) nm, is slightly higher than that of $Mn_2FeAl$ samples having 0.6339(1) nm
- lattice constant values are not affected by alloy annealing

## PAS




CDB results - momentum distributions of annihilating electron-positron pairs presented as ratio curves relative to pure Al.

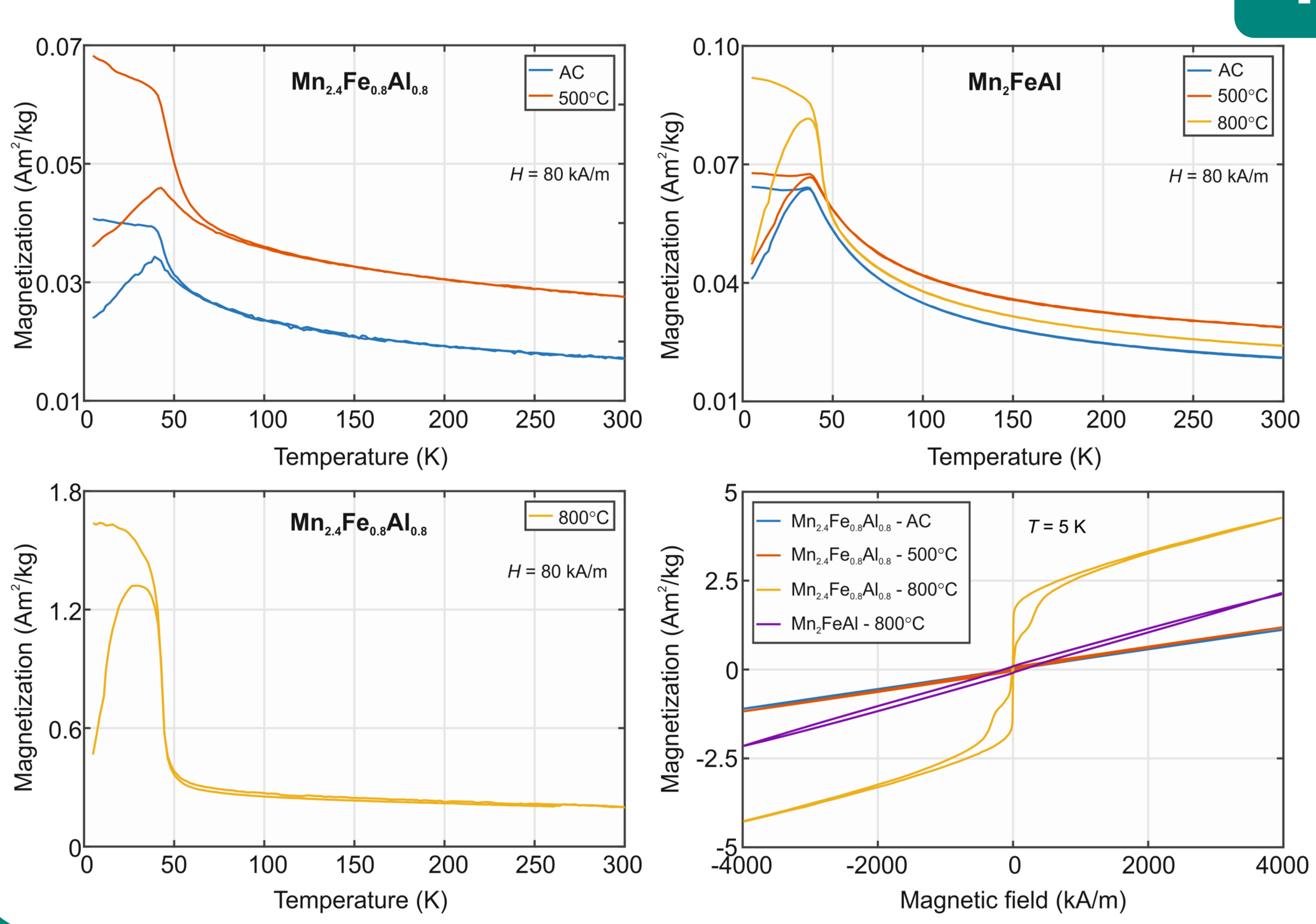
| $Mn_2FeAl$ | $\tau_1$ (ps) | $I_1$ (%) |
|---|---|---|
| as-cast | 124.5(5) | 100 |
| ann. 500 °C | 123.7(5) | 100 |
| ann. 800 °C | 125.1(5) | 100 |
| **$Mn_{2.4}Fe_{0.8}Al_{0.8}$** | **$\tau_1$ (ps)** | **$I_1$ (%)** |
| as-cast | 126.7(5) | 100 |
| ann. 500 °C | 125.8(5) | 100 |
| ann. 800 °C | 127.4(5) | 100 |

Results of PLT measurements (Table) are similar and contains one component with lifetime ≈ 125 ps corresponding well to the lifetime of free positrons in $Mn_2FeAl$ crystal with $\beta$-Mn structure and lattice parameter 0.637 nm.

Unlike other Heusler alloys, $Mn_2FeAl$-based alloys contain no defects and virtually all positrons annihilate as free in the delocalized state.

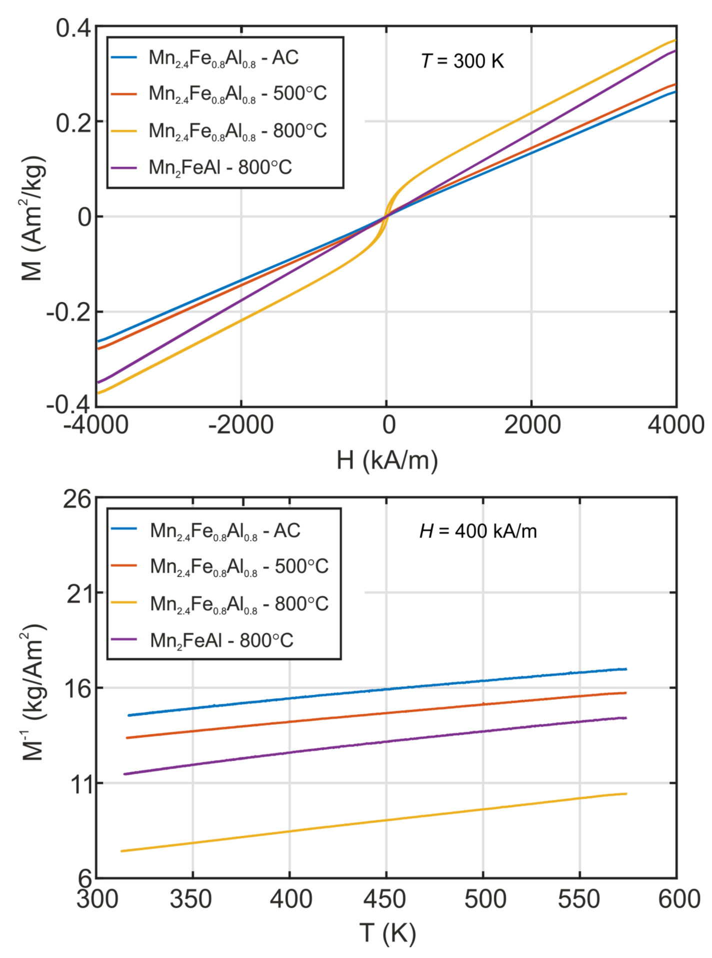
No significant change occurs either after annealing or when increasing the amount of Mn in the alloy composition.

## MAGNETIC PROPERTIES




Magnetic characteristics determined from the Curie-Weiss law and FC-ZFC curves; $C$ – Curie constant; $T_C$ – Curie temperature; $T_N$ – Néel temperature; $T_{irr}$ – temperature of irreversibility, $f$ – geometric frustration parameter, $\mu_{eff}$ – effective paramagnetic moment.

| | $Mn_{2.4}Fe_{0.8}Al_{0.8}$ | | | $Mn_2FeAl$ | | |
|---|---|---|---|---|---|---|
| | as-cast | 500°C | 800°C | as-cast | 500°C | 800°C |
| $T_N$ (K) | 39 | 43 | 29 | 36 | 37 | 36 |
| $T_C$ (K) | -1199 | -1120 | -319 | -657 | -718 | -670 |
| $C \cdot 10^{-4}$ (m³K/kg) | 2.6 | 2.7 | 2.1 | 2.1 | 2.3 | 2.2 |
| $T_{irr}$ (K) | 61 | 85 | 42 | 39 | 41 | 42 |
| $f = \|T_C\| / T_N$ (-) | 30.7 | 26.0 | 11 | 18.3 | 19.4 | 18.6 |
| $\mu_{eff}$ ($\mu_B$/f.u.) | 5.6 | 5.7 | 5.1 | 5.1 | 5.3 | 5.2 |

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