

## Recent developments of manganese-aluminium as rare-earth-free magnets

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**Abstract.** This article reviews findings and progresses in the past decade on manganese-aluminium (MnAl) based magnets as the interest has been revived to fulfill their potential as commercial magnets. The challenges in developments of these rare-earth-free magnets are to acquire a high remanence and coercivity from the ferromagnetic  $\tau$ -phase in MnAl alloys. To this end, the phase transformation to this  $\tau$ -MnAl with  $L1_0$  body centered tetragonal structure has been promoted by a variety of methods and a few percents of carbon (C) is often added to prevent the phase decomposition. Magnetization and coercivity are not only influenced by the phase composition but also the microstructure. The fabrication processes and factors affecting the phase and microstructure are therefore covered. Finally, the productions of bulk MnAl magnets are addressed.

**Keywords:** rare-earth-free magnet; manganese-aluminium; ferromagnetic phase; coercivity; magnetization

### 1. Introduction

Permanent magnets have been utilized in hard disk drives, electrical generators, motors and other electrical appliances. The advents of novel wind turbines, flywheels, magnetic refrigerators and electrical vehicles ensure the prolonged demands of magnetic components. The quest for new rare-earth-free magnets is driven by the concern over the supply of rare-earth elements. To fill the gap between high-performance NdFeB magnets and low-cost ferrites, Mn based alloys have been developed (Poudyal and Liu 2013, Li *et al.* 2016, Hirosawa *et al.* 2017). Whereas Mn is antiferromagnetic, alloying Mn with Bi, Ga or Al leads to ferromagnetism at room temperature (Patel *et al.* 2018, Yang *et al.* 2018).

The implementation of ferromagnetic MnAl was initially proposed by Kono (1958) and the research had significantly progressed in 1970s (Ohtani *et al.* 1977). However, MnAl magnets have never been widely commercialized because the maximum energy products are still lower than those of rare-earth magnets. Whereas the coercivity and saturation magnetization of MnAl are moderate,

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the magnetocrystalline anisotropy (1.50-1.77 MJ/m<sup>3</sup>) and the Curie temperature (650 K) of MnAl are relatively high (Cui *et al.* 2018). Mechanical properties, weight and cost are considered as its advantages over other materials.

In this article, recent developments on MnAl magnets are reviewed. The issues from the structure to productions of magnets are addressed. The ferromagnetic phase in MnAl is a metastable and the synthesis of pure phase is not straightforward. Attempts have been made to stabilize and promote the ferromagnetic phase by various means. Magnetic properties have been enhanced by controlling structural defects and grain sizes. Finally, the prospect of producing bulk MnAl magnets is presented.

## 2. Formation and phase transformation of ferromagnetic MnAl

Intermetallic MnAl alloys exhibit ferromagnetism when the (0, 0, 0) and (½, ½, ½) sites in the L1<sub>0</sub> body centered tetragonal crystal are respectively occupied by Mn and Al atoms. In this structure referred to as “τ-phase”, the expansion of Mn atomic distances to over 2.9 Å by the presence of layers of Al atoms give rise to ferromagnetic ordering at the expense of antiferromagnetic coupling (Cui *et al.* 2018). The real MnAl crystals differ from this ideal equiatomic L1<sub>0</sub> structure. Ferromagnetic τ-phase occurs when the Mn composition is 51-59 at.%. Some Mn atoms occupy Al sites and couple antiferromagnetically with Mn atoms in the Mn layers. The saturation magnetization is reduced because of competing ferromagnetic and antiferromagnetic interactions (Anand *et al.* 2014). Park *et al.* (2010) reported the saturation magnetization of 98.3 emu/g from the experiment and compared to 144 emu/g from the first principle calculation. The latest report suggested that the antiferromagnetism due to the super-exchange interaction between Mn atoms via p-electrons of Al atoms should be suppressed (Sato *et al.* 2020). The magnetization of MnAl can be raised by substituting excess Mn atoms in the Al layers and increasing the tetragonal distortion.

C has widely been used in substitutional alloying and stabilizing the τ-MnAl. A small amount of graphite is commonly added but the introduction in forms of manganese carbide (Mn<sub>23</sub>C<sub>6</sub>) led to homogeneous powders with improved magnetic properties (Tyrman *et al.* 2017). Zhao *et al.* (2018) suggested that the solubility limit of C in τ-MnAl was around 3% and high loadings gave rise to the C-rich phase which is detrimental to magnetic properties. Several experimental works confirmed that the τ-phase in MnAl is stabilized by C (Liu *et al.* 2015, Fang *et al.* 2016, Zhao *et al.* 2018, Qian *et al.* 2019, Si *et al.* 2019a, Saetang *et al.* 2020). Yang *et al.* used the neutron diffraction to prove that C substitute Mn in (½, ½, ½) sites (Yang *et al.* 1984). Alternatively, Han *et al.* (1993) suggested that C atoms tended to enter the octahedral interstitial sites (½, ½, 0) and increased the c/a-ratio as a result. It follows that the antiferromagnetic ordering is decreased and the structure stability is increased. The magnetization is increased as the phase decomposition is suppressed (Zhao *et al.* 2018). Moreover, the mechanical properties are likely improved and the possible Mn<sub>3</sub>AlC phase can pin domain walls thereby enhancing the coercivity (Palanisamy *et al.* 2017). The upper limits of the maximum energy products in MnAl and MnAlC are respectively estimated as 13.2 and 16.8 MGOe (Cui *et al.* 2018). However, the Curie temperature is lowered by the C doping (Wei *et al.* 2014).

The pure ferromagnetic τ-phase cannot be obtained from the melting process directly but it is commonly derived from the high-temperature ε-phase which is stable above 863°C (Cui *et al.* 2018). According to the Mn-Al phase diagram (Shukla and Pelton 2009), the τ-phase is formed when the hexagonal close packed (hcp) ε-phase is either cooled at a control rate or quenched and then annealed. There are different pathways in this ε→τ phase transformation and Genc *et al.* (2019) pointed out the influence of the composition. The ε→τ phase transformation was analyzed by taking into account

of the local activation energy (Lu *et al.* 2016a). Importantly, the resulting  $\tau$ -MnAl is metastable and tends to decompose into nonmagnetic  $\gamma_2$  ( $\text{Mn}_5\text{Al}_8$ ) and  $\beta$  (Mn) phases. Although the annealing is commonly required to promote the  $\tau$ -phase, prolonged heat treatments at high temperatures inadvertently lead to the decomposition into nonmagnetic phases (Cui *et al.* 2018, Palanisamy *et al.* 2019). Qian *et al.* showed that the annealing at 680°C had adverse effects on the  $\tau$ -MnAl (Qian *et al.* 2019). Kobayashi *et al.* (2019) applied magnetic field during the annealing to promote the  $\varepsilon \rightarrow \tau$  phase transformation and suggested that the gain of Zeeman energy of  $\tau$ -phase also inhibited the  $\tau \rightarrow \beta$  transformation.

The  $\varepsilon \rightarrow \tau$  phase transformation in MnAl and MnAlC alloys were composed of displacive mode and the massive mode (Si *et al.* 2017). In the low temperature regimes of 237-377°C for MnAl and 267-493°C for MnAlC, the transformation is driven by the displacive mode. With increasing temperature, the massive mode with the diffusive nucleation and growth processes is increasingly dominant (Si *et al.* 2017, 2019a, Palanisamy *et al.* 2019). Si *et al.* (2019b) observed the massive phase transformation of the melt-spun MnAl and MnAlC ribbons in the range of 387-466°C and 427-485°C, respectively. The onset temperature of phase transformation in these melt-spun MnAlC ribbons was a lower than those of water-quenched MnAlC.

### 3. Synthesis and annealing conditions

The previous section underlines that the main challenge in the MnAl developments is to enhance hard magnetic properties from the stabilized ferromagnetic  $\tau$ -phase. To this end, a variety of techniques including drop synthesis, gas atomising, arc melting, induction melting, melt spinning, strip casting and mechanical alloying have been implemented and combined to synthesize MnAl and MnAlC in forms of ribbons and powders. The composition, heating profile as well as other synthesis conditions leading to this specific phase have been investigated and magnetic properties from some works are exemplified in this section. Since the conditions for the highest magnetization and coercivity are often different, the lists for both parameters are separately presented in Tables 1 and 2.

The magnetization is highly influenced by compositions of Mn, Al as well as C. From Table 1, the compositions of 54-55% Mn and 2% C were mostly employed. The deviations from this range likely result in  $\gamma_2$  and  $\beta$  phases. The annealing process for MnAl and MnAlC was optimized based on the measured temperature range of  $\varepsilon \rightarrow \tau$  phase transformations. Annealing temperatures of 350-650°C are commonly used (Janotova *et al.* 2018) but Jiménez-Villacorta *et al.* (2014) demonstrated that the temperature as low as 295 °C was sufficient for the ferromagnetic phase transformation in  $\text{Mn}_{55}\text{Al}_{45}$  ribbons.

As previously suggested, the cooling of MnAl melts is also influential. By controlling the cooling rate in the melt-spinning process, high purity  $\tau$ -MnAl and hence large magnetization could be obtained without subsequent annealing (Xiang *et al.* 2018a). Moreover, substantial  $\tau$ -MnAl were directly synthesized from the drop synthesis, strip casting and induction melting (Fang *et al.* 2016, Shao *et al.* 2017, Charoensuk *et al.* 2020). Interestingly, experiments suggested that a large fraction of  $\tau$ -MnAl could be directly formed without transformation from the  $\varepsilon$ -phase. Kinemuchi *et al.* (2016) employed high pressure exceeding 5 GPa to derive the  $\tau$ -phase from the synthesis at 900°C. Sato and Irie (2019) obtained substantial  $\tau$ -phase in electrodeposited MnAl film but the annealing was still employed to enhance the coercivity.

Table 1 Maximum mass magnetization reported in selected works using different processes

Reference	MnAl(C)	Process	Max. mass magnetization (emu/g)
Jiménez-Villacorta <i>et al.</i> 2014	Mn <sub>55</sub> Al <sub>45</sub>	melt spinning → annealing at 345°C	18
Lu <i>et al.</i> 2016b	Mn <sub>57</sub> Al <sub>43</sub>	melt spinning → mechanical alloying	62
Crisan <i>et al.</i> 2018	Mn <sub>55</sub> Al <sub>45</sub>	arc melting → melt spinning	60
Xiang <i>et al.</i> 2018a	MnAl	induction melting → melt spinning at 5 m/s	105
Shao <i>et al.</i> 2017	Mn <sub>54</sub> Al <sub>46</sub>	induction melting → strip casting → grinding	114
Janotova <i>et al.</i> 2017	Mn <sub>55</sub> Al <sub>45</sub>	induction melting → planar flow casting → annealing at 440°C for 1 h	85
Fang <i>et al.</i> 2016	Mn <sub>54</sub> Al <sub>46</sub> C <sub>2</sub>	drop synthesis → cryo milling for 4 h → annealing at 600°C for 30 min	94
Chaturvedi <i>et al.</i> 2014a	Mn <sub>54</sub> Al <sub>46</sub>	gas atomizing → milling for 20 h → annealing at 600°C for 30 min	55
Sato and Irie 2019	MnAl	electrodeposition → annealing at 450°C for 16 h	111

Table 2 Maximum coercivity reported in selected works using different processes

Reference	MnAl(C)	Process	Coercivity (kOe)
Jiménez-Villacorta <i>et al.</i> 2014	Mn <sub>55</sub> Al <sub>45</sub>	melt spinning → annealing at 345°C	2.5
Lu <i>et al.</i> 2016b	Mn <sub>57</sub> Al <sub>43</sub>	melt spinning → mechanical alloying	5.3
Crisan <i>et al.</i> 2018	Mn <sub>55</sub> Al <sub>45</sub>	arc melting → melt spinning	2.0
Wei <i>et al.</i> 2014	Mn <sub>54</sub> Al <sub>46</sub>	arc melting → melt spinning at 15 m/s → annealing at 450°C for 45 min	5.0
Xiang <i>et al.</i> 2018a	MnAl	induction melting → melt spinning at 10 m/s	1.8
Shao <i>et al.</i> 2017	Mn <sub>54</sub> Al <sub>46</sub>	induction melting → strip casting → grinding	2.8
Janotova <i>et al.</i> 2017	Mn <sub>55</sub> Al <sub>45</sub>	induction melting → planar flow casting → annealing at 400°C for 1 h	1.6
Shafeie <i>et al.</i> 2019	Mn <sub>55</sub> Al <sub>45</sub> C <sub>2</sub>	induction heating through drop synthesis → kept at 1400°C for 5-10 min	0.7

Table 2 Continued

Reference	MnAl(C)	Process	Coercivity (kOe)
Fang <i>et al.</i> 2016	Mn <sub>54</sub> Al <sub>46</sub>	drop synthesis → cryo milling for 4 h → annealing at 600°C for 30 min	5.3
Fang <i>et al.</i> 2018	Mn <sub>55</sub> Al <sub>45</sub> C <sub>2</sub>	drop synthesis → cryo milling for 2 h	3.8
Chaturvedi <i>et al.</i> 2014a	Mn <sub>54</sub> Al <sub>46</sub>	gas atomizing → milling for 20 h → annealing at 600°C for 30 min	5.0
Law <i>et al.</i> 2017	Mn <sub>54</sub> Al <sub>46</sub>	gas atomizing → milling for 3 min → annealing at 355°C for 10 min	4.9
Jian <i>et al.</i> 2015	Mn <sub>53.3</sub> Al <sub>45</sub> C <sub>1.7</sub>	mechanical alloying for 12 h	5.2
Nguyen <i>et al.</i> 2018	Mn <sub>54.2</sub> Al <sub>43.8</sub> C <sub>2</sub>	mechanical alloying → annealing at 1050°C for 1 h and 500-535°C for 45 min	1.8
Sato and Irie 2019	MnAl	electrodeposition → annealing at 400°C for 16h	12.3

Interestingly, the mechanical milling directly leads to  $\tau$ -MnAl by producing microstrain in the  $\epsilon$ -phase (Cui *et al.* 2018). Corresponding to the high magnetization, Nguyen *et al.* (2018) successfully synthesized as high as 99%  $\tau$ -MnAl by the ball milling. Grain boundaries affect the thermal stability of MnAl because they likely become nucleation sites of ferromagnetic  $\tau$ -phase in the massive transformation (Bittner *et al.* 2017b, Janotova *et al.* 2017). The grain growth is detrimental to the coercivity. According to the experiment by Xiang *et al.* (2018a), the coercivity tended to increase with increasing amount of grain boundaries. The grain refinement of magnetic materials is normally achieved by the mechanical milling (Gabay and Hadjipanayis 2015).

There were several reports on coercivity enhancement by milling MnAl and MnAlC powders synthesized by other methods (Chaturvedi *et al.* 2014a, Fang *et al.* 2016, Lu *et al.* 2016b, Qian *et al.* 2018a). The mechanical milling increases the coercivity by virtue of the grain size reduction but the saturation magnetization is often reduced because of phase decomposition and defect formation. Jian *et al.* (2015) reported the increase in coercivity of MnAlC with the milling time and suggested that the reduced magnetization by structural defects during milling could be recovered by the annealing. Chaturvedi *et al.* (2014a) employed the milling times up to 20 h on Mn<sub>54</sub>Al<sub>46</sub> produced via gas atomization, melt spinning, and rapid solidification rate processing.

To avoid the rise in temperature during the milling, short milling times were also tested. The annealing after milling for 3 min also greatly enhanced the coercivity in gas atomized Mn<sub>54</sub>Al<sub>46</sub> powders (Law *et al.* 2017). Rial *et al.* (2017) used the flash milling for only 30 s to promote the phase transformation and microstrain in gas-atomized Mn<sub>54</sub>Al<sub>46</sub>. The  $\tau$ -MnAl was enhanced by the microstrain induced during the cryogenic milling (Marshall *et al.* 2016). Fang *et al.* (2016) also used the cryogenic milling on the drop synthesized MnAl and demonstrated in another work that the flash heating after the ball milling could increase the saturation magnetization (Fang *et al.* 2018). Su *et al.* (2015) suggested that the microstrain and defects in MnAl after the surfactant-assisted ball milling pinned the domain walls, thereby increasing the coercivity. In another work, they demonstrated the inhibition of phase decomposition by the addition of C and Cu in the surfactant-assisted ball milling

(Su *et al.* 2019). MnAlC flakes with varying sizes and thickness were also derived from this method (Liu *et al.* 2015).

#### 4. Microstructural changes and element doping

Variations in magnetic properties in Tables 1 and 2 are not only due to the phase contents but also the microstructures of MnAl alloys. Although the high purity  $\tau$ -phase led to high magnetization, the values of coercivity in some experiments were moderate. The coercivity of  $\tau$ -MnAl may be reduced by the reversed magnetic domains, antiphase boundaries and twin defects (Bance *et al.* 2017). Arapan *et al.* (2019) investigated the effect of antiphase boundaries on  $\tau$ -MnAl by using the first principle calculation. Palanisamy *et al.* (2019) also revealed the effect of micro-twin on phase transformation. However, the structural defects capable of pinning domain walls enhance the coercivity. Bittner *et al.* (2017a) used electron backscatter diffraction to confirm the domain wall pinning by dislocation. Precipitated phases with comparable size to the domain wall width can be introduced by either solid state diffusion or doping for domain wall pinning. The doping is therefore a route to improve hard magnetic properties of MnAl.

In addition to C, the doping of other elements in MnAl is also a topic of interest. In the case of Ni and Ti, magnetic properties are not significantly modified. The effects of B have been contradictory. Xiang *et al.* (2018b) reported that the addition of B simultaneously enhanced coercivity and magnetization. The latter occurred since B tended to substitute excess Mn with B in the position of Al atoms. The B doping also decreased the onset temperature for  $\epsilon \rightarrow \tau$  phase transformation. By contrast, other works suggested that B did not stabilize the  $\tau$ -MnAl (Liu *et al.* 2012, Fang *et al.* 2017, Kontos *et al.* 2019). The  $c/a$ -ratio was reduced after the B doping (Kontos *et al.* 2019).

For some elements, the trade-off between coercivity and saturation magnetization was observed. Qian *et al.* (2018a) reported that the doping by Fe raised the Curie temperature and coercivity but reduced the saturation magnetization. According to the first principle calculation (Manchanda *et al.* 2014), the magnetic anisotropy is increased but the magnetization is diluted with increasing Fe. The Si doping gave rise to a similar trend and the reduction in magnetization was related to the decomposition into nonmagnetic  $\gamma_2$ - and  $\beta$ - phases. The addition of Si increased the onset temperature of the massive  $\epsilon \rightarrow \tau$  phase transformation (Qian *et al.* 2018a).

Coercivity and saturation magnetization of MnAlC were simultaneously increased by the doping by Zr (Geng *et al.* 2015) and Zn (Wang *et al.* 2011). The addition of either Co or V also enhanced the formation and improved thermal stability of ferromagnetic  $\tau$  phase in nanocrystalline MnAl alloys (Xiang *et al.* 2019, 2020). It was suggested that the anisotropy constant could be enhanced by the 3d-4f electron interactions from Dy and Pr doping but experiments revealed that the magnetic properties of MnAlC alloys were not significantly improved (Liu *et al.* 2012). Tb was introduced as nanoprecipitates to increase the coercivity of MnAl by virtue of the domain wall pinning (Zhao *et al.* 2019a). Finally, the substitution of Al by Ga was also reported and it was suggested that the ternary alloy exhibit better magnetic properties than those of MnAl and MnGa (Mix *et al.* 2017). The experiment and theoretical treatment agree that Ga occupies ( $\frac{1}{2}$ ,  $\frac{1}{2}$ ,  $\frac{1}{2}$ ) whereas Co and Cu atoms tend to enter (0, 0, 0) sites (Zhao *et al.* 2019b).

## 5. Consolidation of MnAl-based magnets

To implement rare-earth-free magnets, MnAl and MnAlC powders have to be consolidated into bulk forms. The hot compaction has predominantly been employed for permanent magnets but high temperatures during this traditional method likely decompose the  $\tau$ -MnAl. Madugundo *et al.* (2016) compared MnAl magnets prepared by hot compaction, microwave-sintering and hot-deformation. The optimum saturation magnetization, remanence and coercivity of respectively 82 emu/g, 50 emu/g and 2.2 kOe in hot-deformed magnets are attributed to the texture developed during the deformation. The hot extrusion was implemented for MnAlC magnets by Thielsch *et al.* (2017). Using the equal channel angular extrusion at a relatively low temperature of 375°C, Chaturvedi *et al.* (2014b) produced bulk nanocrystalline magnets from gas atomized MnAl powders. The coercivity was the highest for an extruded sample annealed at 400°C for 30 min. The spark plasma sintering was also successfully used to produce bulk MnAl (Saravanan *et al.* 2015a) and MnAlC (Pasko *et al.* 2014). The coercivity and saturation magnetization were further increased with the temperature in the rapid thermal annealing from 500-700°C (Saravanan *et al.* 2015b).

High pressure offers an alternative to produce bulk magnets. Magnetic properties of the MnAl based magnets could be enhanced because the high-pressure compaction reduces the grain size and increases the grain boundaries. Tyrman *et al.* (2018) observed the dependence of the coercivity of MnAlC magnets on the compaction pressure up to 400 MPa. After the high-pressure compaction of MnAl and MnAlC powders, the coercivities of the bulk magnets were increased from 0.05 T and 0.08 T to 0.39 T and 0.22 T, respectively (Qian *et al.* 2019). Gas-atomized MnAlC powders were consolidated into magnets with enhanced coercivity and remanence by the high-pressure compaction (Si *et al.* 2019a). Si *et al.* (2019b, 2020a) also employed the high-pressure compaction on melt-spun MnAlC and the high-pressure torsion (6 GPa) on B-N-doped MnAl. Furthermore, MnAl based magnets can be benefited from subsequent heat treatments. The heat treatment at 1050 °C for 6 h promoted the  $\tau$ -MnAl. A higher annealing temperature (1100°C) and a longer time (10 h) gave rise to enhance the  $\tau$ -phase and homogeneous composition (Qian *et al.* 2019). For the MnAl disk prepared by the severe plastic deformation (Si *et al.* 2020b), the magnetization of was increased after the annealing at the expense of the coercivity from 5.90 to 3.96 kOe.

MnAl based magnets can also be prepared in forms of composites. The incorporation of soft magnetic Fe<sub>65</sub>Co<sub>35</sub> gave rise to the domain walls pinning in MnAlC and magnetic properties were improved by the exchange coupling with the hard magnetic Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> (Qian *et al.* 2018b). Saravanan *et al.* (2017) used the spark plasma sintering to reinforce MnAl composites with carbon nanotubes. Palmero *et al.* suggested that the magnets of varying shapes could be produced using the 3D printing on polymer bonded MnAlC (Palmero *et al.* 2018). Electron beam melting was recently investigated as binder-free additive manufacturing technique for MnAl magnets (Radulov *et al.* 2019).

## 6. Conclusions

MnAl possesses required attributes for the gap magnets between ferrites and rare-earth magnets and the understanding of influential factors is crucial for the MnAl implementation. Arc melting, induction melting, melt-spinning, gas-atomising, and drop synthesis process are used to initiate the ferromagnetic  $\tau$ -phase. The annealing after the alloying is required in most cases in order to enhance the  $\tau$ -MnAl. Alternatively, it is shown that the formation of the  $\epsilon$ -phase is not a necessary condition

for the formation and the  $\tau$ -phase is directly obtained. The  $\tau$ -MnAl is more stable when C is added and the latest report also indicates the C contribution in stabilizing the precursor  $\varepsilon$ -phase and promoting  $\tau$ -phase formation (Mitsui *et al.* 2020). Microstructures and structural defects also influence magnetic properties. The mechanical milling and the doping of elements including B, Fe, Si, Co, Zn, Zr, V are therefore employed but the trade-off between magnetization and coercivity is often obtained. The fabrications of bulk MnAl and MnAlC magnets using high temperature and pressure have been demonstrated. The prospect of increasing use of simulation designs, recently exemplified by Kovacs *et al.* (2020), certainly facilitates the development of MnAl as rare-earth-free magnets.

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