



Energy Efficiency & Renewable Energy

Development of Radically Enhanced alnico Magnets (DREaM) for Traction Drive Motors

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THE Ames Laboratory Creating Materials & Energy Solutions

Project ID # EDT015

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Overview

Barriers & Targets*

Timeline

- Start October 2014
- Finish September 2018 19% Complete

Budget

- Total project funding
 - DOE share \$1900K (new start in FY15)
- FY16 Funding \$2500K (proposed)
- FY17 Funding \$2500K (planned)

- High energy density permanent magnets (PM) needed for compact, high torque drive motors (specific power >1.4kW/kg and power density >4.0kW/L).
- Reduced cost (<\$8/kW): Efficient (>94%) motors require aligned magnets with netshape and simplified mass production.
- RE Minerals: Rising prices of rare earth (RE) elements, price instability, and looming shortage, especially Dy.
- Performance & Lifetime: High temperature tolerance (180-200°C) and long life (15 yrs.) needed for magnets in PM motors.

Partners

- Baldor, U. Wisconsin, GM, GE, UQM, Synthesis Partners (collaborators)
- ORNL, U. Maryland, U. Nebraska, Brown U., Arnold Magnetic Tech. (DREaM subcontractors)
- Project lead: Ames Lab

Project Relevance/Objectives

- To meet 2020 goals for enhanced specific power, power density, and reduced (stable) cost with mass production capability for advanced electric drive motors, improved alloys and processing of permanent magnets (PM) must be developed.
- Rising RE cost trend and unpredictable import quotas (by China) for RE supplies (particularly Dy) motivates this research effort to improve (Fe-Co)-based alnico permanent magnet alloys (with reduced Co) and processing methods to achieve high magnetic strength (especially coercivity) for high torque drive motors.

Objectives for the fully developed PM material:

- Provide competitive performance in advanced drive motors, compared to IPM motors with RE-PM.
- Eliminate use of RE, e.g., Nd, Dy, in high performance PM due to global strategic RE supply issues.
- ✓ Achieve superior elevated temperature performance (180-200°C) to minimize motor cooling needs.



DREaM Overall Approach/Strategy

Near-term non-RE Magnets: Best RE-free magnets (alnico) further enhanced (coercivity) by low-Co alloy design and bulk powder processing improvements using detailed and innovative analysis of micro/nano structure-magnetic property relationships, extensive theory results, and critical industry input.

Long-term non-RE Magnets: Advances in Fe-Co-X Magnet magnet systems will be shifted to "super-alnico" for Target added tetragonal distortion of Fe-Co magnetic phase for a further boost of coercivity from magnetocrystalline 50 anisotropy, coupling theory with 45 synthesis/characterization and bulk magnet processing 40 480 60 35 BH)_{max}, MGOe OTHER IMPORTANT CHARACTERISTICS 440 Required magnetizing field Nd-Fe-B Thermal stability, Resistivity 30 50 400 Corrosion Resistance Manufacturability, Cost, etc Aniso Bonded Sm-Fe-Ni 360 25 Aniso Bonded Nd-Fe-B 40 320 BHmax, MGOe Iso Bonded Nd-Fe-B kJ/m: 280 20 SmCo 240 BHmax, 15 Ferrite 200 Alnico 9 160 10 Alnico 5 & 5-7 NdFeB(45) 120 NdDvFeCoB(30) 5 10 Pt-Co 80 SmCo5(20) MK Steel KS Steel 40 Alnico (9) 0 50 100 150 200 250 300 1900 1920 1940 1960 1980 2000 YEAR Temperature, °C

FY15 DREaM Tasks

2014 Oct	Nov	Dec	2015 Jan	Feb	Mar	Apr	Мау	Jun	Jul	Aug	Sep
Develop enhanced alnico (non-RE) magnets Theory and Modeling: Calculate the atomistic energetics, site occupancies and nano-scale chemical gradients across the boundary of magnetic and matrix phases for alnico alloys as a function of nano-structural scale using efficient Monte Carlo codes in collaborative efforts with experiments. Develop phase field models for simulation of morphology evolution and magnetic behavior at											
micron-scale, including magnetic field application. Synthesis of Bulk Magnetic Samples: Pursue the enhancement of coercivity in the alnico systems by nano-structure refinement, by alloy modifications to reduce magnetic interactions between matrix and magnetic (Fe-Co) phase and by strategies to avoid γ phase inclusions									iverable: proved o magnet in		
Characterization: Data sharing Analysis of combinatorial libraries using synchrotron radiation combined large area scan with concurrent XRD and XRF Characterization of experimental samples to achieve enhanced structural, chemical, and magnetic measurements, utilizing XRD, HR-STEM, 3DAP, MOKE, SQUID, and hysteresigraph.											



Characterization Accomplishment: alnico 8 & 9 are most promising

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- Highest coercivity
- Complex mosaic tile pattern
 - Transverse to field
- Distinct Cu rods
- Additional Ni-rich phases
- L2₁ matrix phase (Ti effect)
- Fe-Co phase elongated with same spacing (may have 90° interrupting colonies

Longitudinal to field





Characterization Accomplishment:Magnetic Annealing Temperature2000Selection Identified by1800Pattern Perfection1600

- Highly anisotropic Fe-Co pillars (longitudinal)
 - Either extended length or with 90° interrupting colonies
- A well-faceted mosaic structure (transverse)
 - formed at optimum T_{MA} (840°C) for fixed time of 10 min.



HAADF STEM images of samples after 10 min. magnetic field heat-treatment only (cooled in furnace).







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HAADF STEM images of samples after full heat treatment (FHT 650°C for 5 hrs and 580°C for 15 hrs): No obvious morphology change comparing with the samples above, however, the size of FeCo rod is slightly increased (+~5-10nm).





Coupling of Theory and Experiment: Processing Effects on Nano-structure & Magnetic Performance

University of Nebraska, Oak Ridge National Lab, Ames Lab



To improve magnetic performance of alnico, especially coercivity, theory efforts must focus on:

- Spinodal decomposition process under applied field during magnetic annealing (MA) ~810-860°C
- Subsequent draw process, i.e., annealing at lower temperatures of 550-650°C.



M-C Simulation Accomplishment: Identified/predicted 4th phase in alnico 8



- M-C calculations on Al-Ni-Co-Fe-Ti model alloy revealed an unrecognized 4th alloy phase in alnico 8 (and 9) after low temperature annealing (drawing).
- The 4th phase is ~ Ni_3AI with DO_3 ordering.
- Al concentration is the same crossing Ni-Al-Ti and Ni-Al phases.

Accomplishment: Element Maps in High Resolution STEM Supported 4th Phase Identification



Re-examination of HR-STEM compositional data revealed a 4th phase very rich in Ni with a depletion of Fe, Co and Ti.

<u>University of Nebraska</u>



Observed Magnetic Anneal Time Effect

Apparent Fe-Co dia. 20-25nm after shortest (1.5 min.) MA

• Unexpected coercivity decrease Longer time MA leads to coarsening

• Developed bimodal Fe-Co size The well-defined mosaic structure

• Formed after ~ 5-10 min during MA Drawing cycles promote uniform increase

Mechanism not yet clear

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t (min.)



HAADF STEM images of samples with different MA time.







Site preference of ternary alloying addition (Ti, Fe, Co and Ni) in DO₃ Fe₃Al, Co₃Al and Ni₃Al – basic compound for alnico-8 magnetic materials Oak Ridge National Lab



Our goal is to find distribution of Ti, Fe, Ni Co, Al atoms between nonequivalent sites in $L2_1$ structure (AlNi-phase). For this purpose the energy change caused by substitution of atoms in AlNi-phase prototype compounds (Fe₃Al, Co₃Al and Ni₃Al) by ternary element was calculated.

If sites α and γ are occupied by the same group of atoms L2₁ (AlNi) structure is reduced to DO₃ structure: α -site (Fe): (1/2,1/2,1/2) β -site (Al): (0, 0, 0)

 γ -site (Fe): ±(1/4,1/4,1/4)

Grand canonical formalism was applied to find ground state configuration which corresponds to ternary atoms substitution

Ternary element substitution should be accompanied by antisite defect

Publication: Samolyuk, G.D.; Ujfalussy, B.; Stocks, G.M., J. Appl. Phys. 116, 173908 (2014).

Accomplishments from Ti Effect Study on alnico 8H

- Raising Ti to 7.5% increases Hc
- Ti ~>8% partly destroys the mosaic , structure.
 Optimum MA temperature
- Optimum MA temperature
 - sensitive to alloy composition.





HAADF STEM images of samples with different Ti concentration.





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Accomplishments from Processing and Alloy Design Explorations

- A well-faceted mosaic structure only forms in a narrow T close to the onset of the spinodal (~840°C), which results in optimum magnetic quality.
- External magnetic field aids the formation of well-faceted mosaic structure only at the optimal T.
- Drawing has no obvious effect on alloy's morphology, but does increase its coercivity, if the MA time is long enough.
- The base line 8H sample obtained a Hci of 1840 Oe and (BH)_{max} of 6 MGOe through optimizing magnetic annealing and drawing cycle.
- □ The magnetic properties were further improved by optimizing (B) the amount of Ti and Nb. A Hci of 2211 Oe and (BH)_{max} of 5.6 MGOe (BH)_m=6.0 were achieved so far.





Accomplished Size Scale-up of alnico 8H Magnet Forming



Lineal Shrinkage by Dimension



- "Clean" pre-alloyed powder and burnout of polypropylene carbonate binder leaves essentially no residuals.
- Full Density >99% after 4h sinter (1250°C)
- Isotropic linear shrinkage for near net shape magnet forming.
- Currently form magnets 27mm dia.x10mm and cut 3mm dia.x8mm samples for testing.

As-atomized powder size distribution

Percentile	Size
D50	31 µm
D70	42 µm
D84/D50	1.67

Sinter Time (min)	Carbon (ppm)	Sulfur (ppm)
Industry	~300 ppm	<2000
0 (base powder)	78	20
60	98	-
240	95.3	65.3
480	209	-
720	56*	6.5*

600

Sintering Time (minutes)

Determined Benefits of Prolonged Sintering: May Produce Aligned Grain Growth

- 8h sintered samples contain few large (>1mm) grains that may align: high Br & Bhmax.
- Comparing to aligned 9 magnets: 8h sintered magnet sample matches remanence and exceeds coercivity, with lower energy product and squareness
- Investigating how to promote highly aligned grain growth in sintered bulk magnets.

Br

G

8,523

8,789

8,626

6700

10600

1500

Sample

1 Hour Sinter

4 Hour Sinter

12 Hour Sinter

AMT Sintered

alnico 8H

AMT Cast

alnico 9

8 Hour Sinter 10,052



111

101

001

0.86

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ulk magr	nets.	1 m	1 mm			
Нс	Hk	Нсі	BHmax	Squareness		
Oe	Oe	Oe	MGOe	Hk/Hci		
1,521	459	1,632	4.87	0.28		
1,569	483	1,685	5.04	0.29		
1,608	601	1,688	6.5	0.36		
1,530	452	1,645	4.85	0.27		
1800	_	2020	4.5	_		

9.0

1500

Spinodal Decomposition of Amorphous Al-Ni-Co University of Nebraska



Amorphous Al-Ni-Co decomposes to the Fe-Co and (Ni,Al)-rich phases.
To make "super-alnico," can B be trapped in Fe-Co for tetragonal distortion?



Spinodal decomposition temp. lower, compared to standard MA temp. (>800°C).



Combinatorial investigation of Alnico magnets

- Composition spread optimization of alnico



Co-sputtering scheme

Ni

Mapped composition region Cluster analysis of synchrotron diffraction



In-plane H_c of 350 Oe is obtained in one region (RT deposition; 700°C anneal for 2h).

Useful tool to rapidly explore modified alnico alloys for unknown coercivity "hot spots."

Response to Previous Year Reviewers' Comments (on BREM)

 On future research, the reviewer mentioned that "the program was winding down and will hopefully continue through another program....and requested that if the work continues, to please continue working on AlNiCo and FeCo magnets, Co reduction, and to look at the mechanical properties...stating that improved AlNiCo mechanical properties (i.e., reduced brittleness) are desired."

Fortunately, the work is continuing through a more focused project on alnico magnets that includes Co reduction and is already involved in assessment of mechanical properties by bend tests. As a natural part of the preparation of well machined cylindrical specimens from larger samples, we know that our recent alnico samples survive the forces/impacts of center-less grinding. In new work that we are proposing for FY2016, we are adding NREL researchers to expand the measurement of magnet sample mechanical properties to include tensile ductility and impact.

• Under technical accomplishments, the reviewer reported that "the technical milestones for the magnetic performance of the new magnets to be developed were not very well defined.....and seemed to be towards the main goal of maintaining, or enhancing, the performance of AlNiCo 8 at a 30% reduction in cost associated with a 40% reduction in Co."

Beyond reduction of Co content to the maximum extent possible without significant reduction in saturation magnetization (Ms), the reduction in processing cost and addition of mass production capability by full development of the powder processing approach are technical goals for this project.

• "Good progress has been made towards enhancing the coercivity and energy product, although not yet at the level of the commercial magnets."

Latest alloying results do exceed current coercivity levels.

• "Demag curve tests at higher temperatures would have been useful in assessing the thermal stability of the AlNiCo samples with the highest coercivity".

Our proposal for new work in FY2016 does include expansion of ORNL work to include high temperature magnetic measurements.

• *"a* more clear correlation between the microstructure, processing conditions, and the achieved magnetic properties would have given a clearer picture on the feasibility of reaching the desired optimum microstructure."

Latest results allowed analysis of several issues related to the "optimum microstructure."

Ames Lab Partners/Collaborators for FY2015

Leadership Team: Iver E. Anderson, R. William McCallum, Matthew J. Kramer: AL

DREaM Team:

- K.M. Ho, C.Z. Wang, V. Antropov, and T. Wang: AL
- R. Skomski, D. Sellmyer and J. Shield: Univ. Nebraska-Lincoln
- M. Stocks: ORNL
- I. Takeuchi: Univ. Maryland
- S. Constantinides: Arnold Magnetic Technologies, Inc.



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ETIC TECHNOLOGIES





- Baldor (Mike Melfi): Electric motor manufacturing technology, DREaM technology adviser.
- Univ. Wisconsin-Madison (Tom Jahns): Electric machine design, DREaM technology adviser.
- Synthesis Partners (Chris Whaling): Automated search of permanent magnet literature, DREaM project adviser.
- General Electric (Frank Johnson): Non-RE magnet technology and motor design, started in 2012, VT Motor/Magnet partner (prime).
- Unique Mobility (Jon Lutz): Advanced non-RE PM motor design, started in 2012, VT Motor/Magnet partner (prime).
- Univ. Delaware (George Hadjipanayis): Development of high-energy permanent magnets, ARPA-E partner (prime).
- PNNL (Jun Cui): Friction-stir processing of permanent magnets, ARPA-E partner (prime)
- Case-Western Reserve Univ. (Dave Matthiesen): Transformation enabled nitride magnets absent rare earths, ARPA-E partner (prime)







Remaining Challenges and Barriers

- Coercivity levels achieved recently are sufficient, compared to current alnico magnets, but "safety margin" is needed for PM traction drive motor reliability.
- Must determine by linkage of theoretical analysis and experimental studies the most significant parameters or characteristics of alnico microstructure and nano-structure for enhancing magnetic properties, especially coercivity.
- Design and implement alnico alloying changes to make bulk magnets that achieve desired improvements in microstructure and magnetic properties with alloying elements of reduced cost.
- Establish capabilities of binder-assisted compression molding to fabricate alnico magnets in prototype sizes and shapes with anisotropic magnetic properties by developing solid state growth of highly aligned grains.
- To enable extensive experiments on compression molding and other bulk magnet fabrication methods, additional gas atomized pre-alloyed powder must be produced with high purity and desired composition.

Remaining FY15 DREaM Tasks

Develop Focused Theory & Simulation: Will continue calculations on Al-Ni-Fe-Co-Ti (simulating alnico 8 and 9) with composition variations to add value to nano-scale (equilibrium) Monte Carlo (M-C) simulations of spinodal decomposition and annealing. Output will be verified against new experimental results and used to provide magnetic properties and free energy driving forces to further development of phase field microstructure calculations.

Synthesize Test Samples: Additional bulk samples with nano-structure variations made to pursue promising modifications of alnico magnetic and thermal processing parameters from pre-alloyed powder (alnico 8H and "low-Co" alnico), exploring methods to control grain growth alignment. Bulk magnets of a sub-final size/shape (for UQM) will be made with learning from processing/characterization results with preferred nano-structure and microstructure. [SMART milestone] [Demonstration of bulk prototype alnico magnets with improved magnetic properties and mechanical properties for use in an advanced UQM motor system will be done under separate VT project.]

Perform Characterization: Continue detailed magnetic and microstructural characterization of extensive series of alnico 8 and low-Co samples to verify effects from processing parameter variations and to investigate correlations between magnetic properties and nano-structure. Include magnetic measurements with hysteresigraph (newly installed) and microstructural analysis with FE-SEM, HR-STEM with Lorentz imaging, and 3-D atom probe (ORNL visit).



Future (proposed) FY16 DREaM Detailed Tasks

Promote DREaM Team Interactions: Maintain regular WebEx discussions on specific project progress and conduct two face-to-face workshops per year with research team.

Develop Focused Theory & Simulation: Use theory methods to improve cluster expansion (CE) models to 6-element Al-Ni-Fe-Co-Ti-Cu systems (alnico 8 and 9) to permit realistic equilib. M-C simulations & enabling kinetic M-C to begin on spinodal & annealing. Verify with experimental results and use for calculating magnetic properties and driving forces to extend phase field microstructure calculations.

Perform Characterization: Continue characterization of extensive series of alnico 8 and low-Co samples to verify theory predictions/actual effects from alloy & processing variations and correlating magnetic properties and nano-structure. Utilize new NREL partnership to add magnet mechanical property data. Expand ORNL studies to include temperature dependent magnetic properties and FEM for motor design.



Synthesize Test Samples: Bulk samples with nano-structure variations made to continue pursuit of modified alnico alloys, using magnetic and thermal processing parameters for pre-alloyed powder (alnico 8, refined alnico "low-Co"). Refine methods to gain control over texturing effects in sintered bulk magnets to further improve magnetic properties. Analyze and report results of high magnetic field experiments on nano-structure and magnetic properties. [SMART milestone, complete September 30, 2016]

DREaM Project Summary

Project Duration: FY15 – FY18

Overall Objective (all years): Design and synthesize a high energy product alnico PM competitive with RE-PM (cost/MGOe/kg), but with sustainable supply and cost outlook in bulk near-final shapes by mass production methods.

FY15 Focus: Experimentally verify effects on nano-structure and microstructure of bulk samples from thermal-magnetic and annealing processing variations designed from theory findings and correlated with magnetic properties and the observed nano-structure changes.

Key Deliverable: Bulk, sub-sized alnico magnet with improved magnetic properties (vs. 8HE and/or 9) produced and delivered to UQM for specification of test magnets for advanced motor system.

Go/No Go Decision Point: Does bulk sub-sized alnico magnet have improved magnetic properties compared to alnico 8HE and/or alnico 9 or not?

FY 16-18 Focus: Develop refined low-Co alloy with reduced cost that undergoes spinodal transformation to produce desired nano-scale pattern and exhibits same higher levels of magnetic properties when subject to similar thermal-magnetic and annealing treatments.

Deliverables: Bulk alnico magnet samples made with reduced cost low-Co alnico alloy.





Technical Back-Up Slides

Increasing magnetocrystalline anisotropy in FeCo-rich particles in Alnico: a possible route to improve Alnico

- > It is desired to improve coercivity of Alnico without decreasing remanence.
- ➢ FeCo-rich phase in Alnico have a shape anisotropy about 0.15∼0.23 MJ/m³.
- Body centered tetragonal FeCo has a large magnetocrystaline anisotropy.
- FeCo-rich particle in Alnico is elongated along 001 direction.
- Interface anisotropy can be large (BREM results).

Recent related work from DREaM team:

Scientific Reports 4,6367 (2014)

- Tetragonal FeCo can be realized with V doping.
- Fe-Co-V films with MAE value of 0.3 MJ/m³ had been produced. Very good agreement between experiment and theory on MAE values had been found.



Recent related works from other groups: Interstitial Carbon/Boron doped FeCo

Carbon doped FeCo alloy

Theoretical prediction: Phys. Rev. B 89,144403 (2014)

- Carbon doped Fe-Co alloys form in a wide range of concentrations, in analogy with the formation of martensite in steels.
- > The alloys have a stable tetragonal distortion.
- > MAE up to 0.75 MJ/m³.

Experimental progress: J. Appl. Phys. 116, 213901 (2014)

2 at. % of Carbon leading to the formation of a spontaneously strained phase with 3% tetragonal distortion. (Fe_{0.4}Co_{0.6})_{0.98}C_{0.02} films have MAE above 0.4 MJ/m³.

Boron doped FeCo

Theoretical prediction: J. Phys. D: Appl. Phys. 47 (2014)

➢ MAE up to 0.8 MJ/m³

Conclusion

- > A large increase of MAE in bulk Fe-Co rich phase is possible.
- Interface anisotropy can be significant.

Magnetic Field Effect on Spinodal Results

Magnetic field

- helps the formation of wellfaceted mosaic structure only at 840°C.
 - without field Fe-Co remains connected---low coercivity.

Phase field modeling



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t*=1000s

HAADF STEM images of samples without MA at different temperature.







STEM Observations of Draw Effect

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- The well-defined mosaic structure
 - Longer MA time (~10min) helps preserve the faceting of Fe-Co rods on {110} planes after low temperature drawing.



HAADF STEM images of samples with different MA time but the same drawing process.







Motivation

- Study and reveal the relationship of composition, processing to microstructure and magnetic properties.
- ✓ Study the role of Co, Ti and Nb on magnetic properties

Experimental details

Sample preparation

- ✓ HIP samples were fabricated by GA powder with the base line composition 32.4Fe-38.1Co-12.9Ni-7.3Al-6.4Ti-3.0Cu (in wt.%).
- Casting samples with different Co, Ti and Nb weight percentages which were substituted by Fe based on the base line composition.

Heat treatment

- ✓ Magnetic annealing at 800-860°C for 1.5-60 min.
- Drawing cycle: 650°C5hrs+580°C15hrs

Magnetic properties measurement

 ✓Ø3x8 mm samples were measured by a Laboratorio Elettrofisico Engineering Walker LDJ Scientific AMH-5 Hysteresisgraph.

Microstructure characterization

✓ FEI Tecnai F20 (200kV, FEG) with high-angle annular-dark-field detector.





