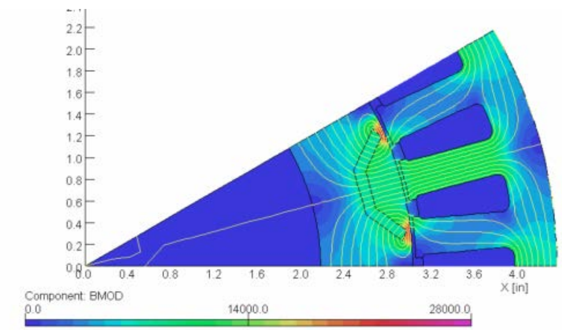
Permanent Magnet Development for Automotive Traction Motors

Includes: *Beyond Rare Earth Magnets (BREM)*

Iver E. Anderson
Ames Laboratory (USDOE)



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June 18, 2014

Project ID: APE015

Overview

Timeline

- Start - August 2001
- Finish - September 2014
- 96% Complete

Budget

- Total project funding
  - DOE share $15,350K (since FY01)
- FY13 Funding - $2000K
- FY14 Funding - $1700K (planned)

*2020 VT Targets

Barriers & Targets*

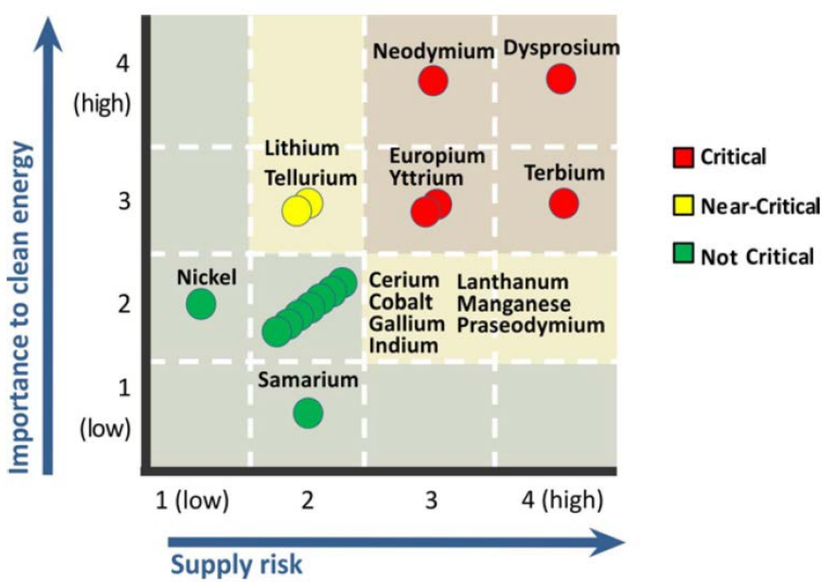
- High energy density permanent magnets (PM) needed for compact, high torque drive motors (specific power >1.4kW/kg and power density >4.0Kw/L).
- Reduced cost (<$8/kW): Efficient (>94%) motors require aligned magnets with net-shape and simple mass production.
- RE Minerals: Rising prices of Rare Earth (RE) elements, price instability, and looming shortage, especially Dy.
- Performance & Lifetime: High temperature tolerance (150-200˚C) and long life (15 yrs.) needed for magnets in PM motors.

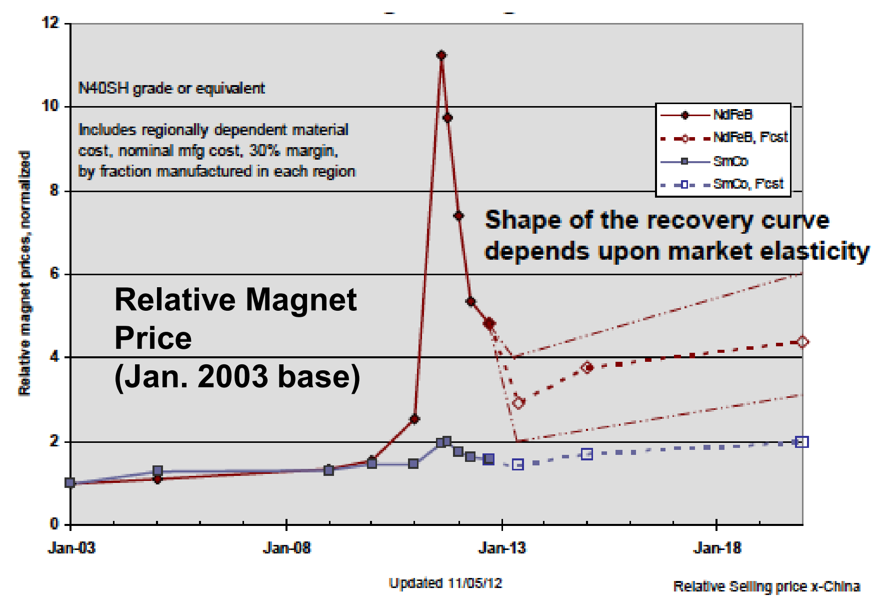
Partners

- Baldor, U. Wisconsin, GM, GE, UQM, Synthesis Partners (collaborators)
- ORNL, U. Maryland, U. Nebraska, Brown U., Arnold Magnetic Tech. (BREM 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.
- Increased RE costs and on-going reduction of import quotas (by China) for RE supplies (particularly Dy) motivates this research effort to improve (Fe-Co)-based permanent magnet alloys (modify or discover new) and processing methods to achieve high magnetic strength (especially coercivity) for high torque drive motors.
- **Objectives for the fully developed PM material:**
  - ✓ remain competitive at room temperature with current high energy (MGOe) magnets.
  - ✓ minimize or eliminate use of scarce RE, e.g., Dy, due to an impending world wide RE shortage.
  - ✓ achieve superior elevated temperature performance (150-200˚C) to minimize motor cooling needs.





Milestones for FY2013 and FY2014

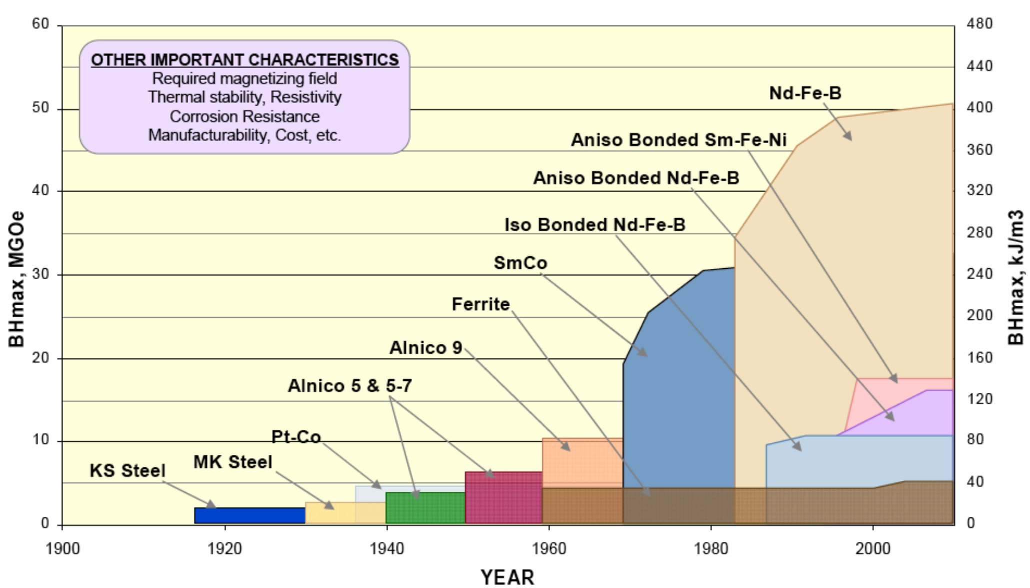
Month/Year	Milestone, Status, and/or Go/No-Go Decision
Sep-2013	**Milestone:** Demonstrate at least one mechanism for improving alnico magnetic properties by comparison of magnetic measurements with microstructure observations and supportive theory. **[completed]** **Milestone:** Complete modification of Genetic Algorithm code to permit initial peta-flop scale supercomputer calculations of selected Fe-Co-X alloys, accelerating exploration of stable structures with large unit cells that are promising magnet compounds. **[completed]** **Milestone:** Synthesize samples of Co-X, Fe-X, & Fe-Co-X (X=e.g., W, Ta, Mo, Zr) with cluster deposition and/or melt spinning. **[completed]**
June-2014	**Milestone:** Perform material and process cost analysis for optimized SSHD process for anisotropic RE (lowest Dy) magnets **[No go: SSHD shown effective with less process cost, but lack of funds/demand stopped work]** **Milestone:** Conduct tenth regular BREM workshop with team to exchange results and refine directions for non-RE magnet work. **[On schedule]** **Milestone:** Recommend directions from theoretical analysis of possible alnico interface/phase configurations and alnico alloying options for new processing trials to further enhance bulk alnico magnets. **[On schedule]**
Sep-2014	**Milestone:** Produce fully pre-alloyed alnico powder with modified (low Co) alnico 8 composition, and consolidate to full density with magnetic annealing and heat treatments designed on the basis of improved understanding of magnetic property optimization. **[On schedule]**

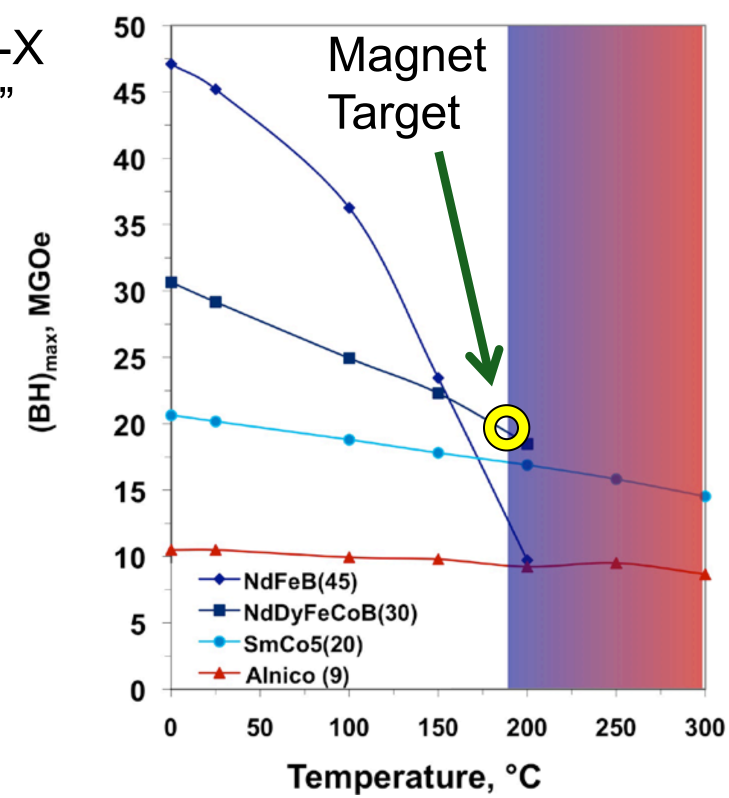
Approach/Strategy: Best Focus for Future

Near-term RE Magnets: Simplified process is promising for anisotropic mixed rare earth (Nd-based) magnets & **little or no Dy**, but further development for high temperature (HT) stability needs improved magnet alloy (insufficient funds to extend).

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

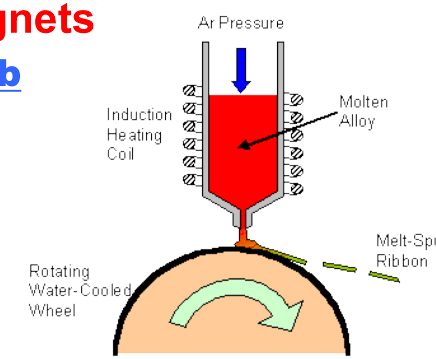
Long-term non-RE Magnets: Advances in Fe-Co-X magnet systems will be re-directed to “super-alnico” (tetragonal Fe-Co) with theoretical/synthesis efforts focused on design of other alnico additions.



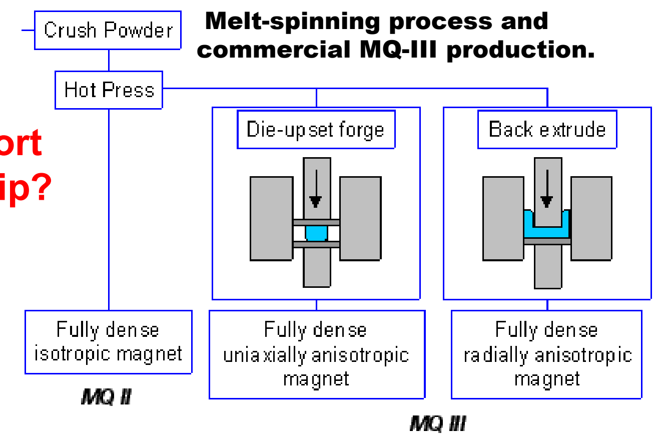


Development of Single-Stage Hot Deformation for Anisotropic Magnets from Zn-coated MRE-Fe-B Flake Powder

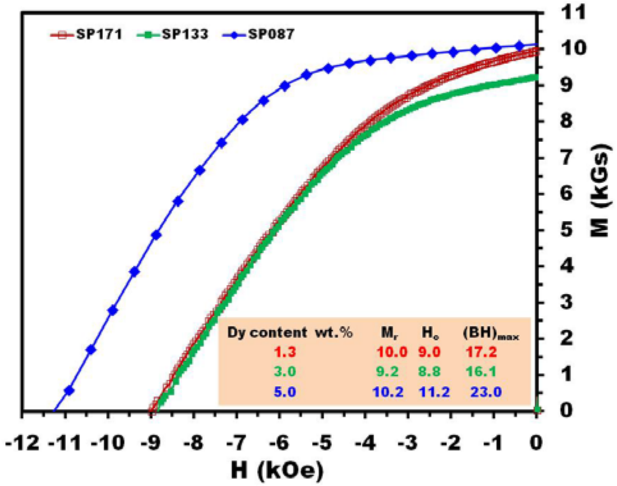
Ames Lab



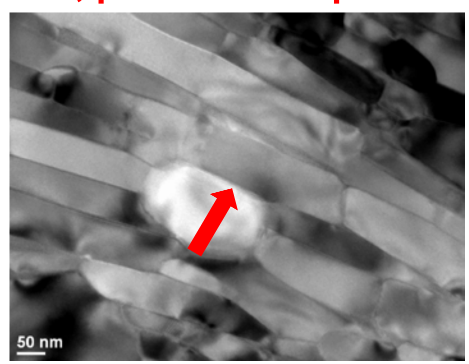
- Commercial anisotropic magnets (MQIII) used over-quenched flake particulate to avoid casting segregation/annealing, but needed ($$) two-stage hot deformation (hot press and die-upset forging) for fully dense Nd-Fe-B magnets.
- A new single stage hot deformation (SSHD) method is being developed to fabricate fully dense anisotropic magnets from Zn-coated glassy $MRE_2(Fe,Co)_{14}B$ flake: low Dy---need HT stability from improved alloy (with Ga?).
- Full density and stress-induced platelet grain growth (~30-60nm by ~150-300nm) results in 11.2 kOe and 23 MGOe for 5% Dy, and 9 kOe and 17.2 MGOe for 1.3 Dy.

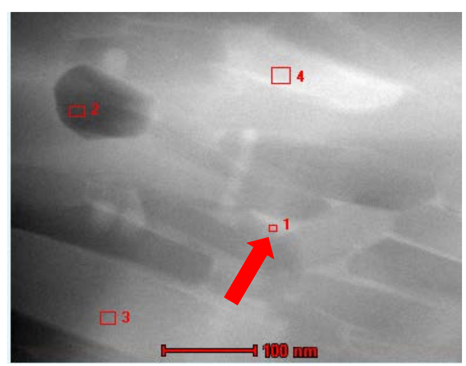


Melt-spinning process and commercial MQ-III production.



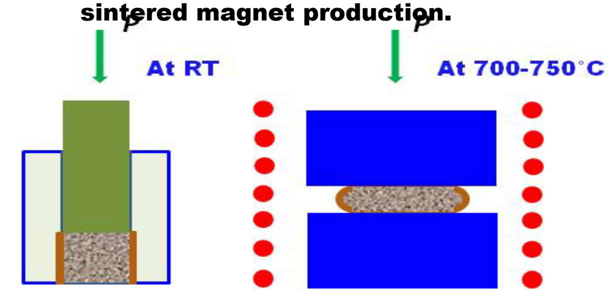
Funding decrease forced this effort to cease; potential MQ partnership?





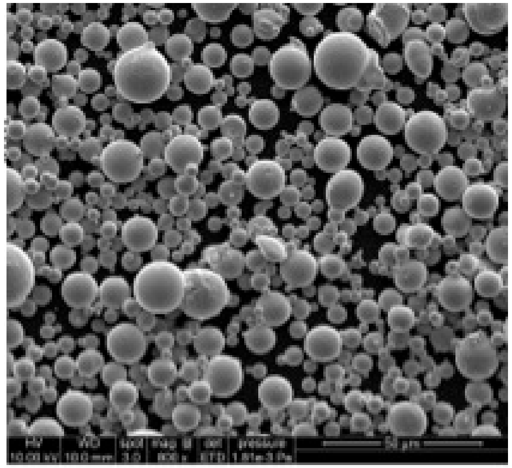
- STEM EDS detected Zn, only in the smooth grain boundaries (region 1)
- Zn diffused to grain boundaries from the interparticle regions.
- Zn helps retain a higher Hc after hot deformation (Dy from 5 to 1.3 wt%).
- Use of new alloy from MQ (with Ga) could demonstrate process value.

Melt spun flake and single stage hot deformation (SSHD) for anisotropic sintered magnet production.



W. Tang et al, "Anisotropic Hot-Deformed Magnets prepared by Zn-coated MRE-Fe-Co-B ribbon Powder", J. Appl. Phys. Vol. 115, 17A725(2014).

First Gas Atomized Pre-Alloyed Alnico 8H: Processing Innovation for Magnet Manufacturing



Dia.<20μm screened powder

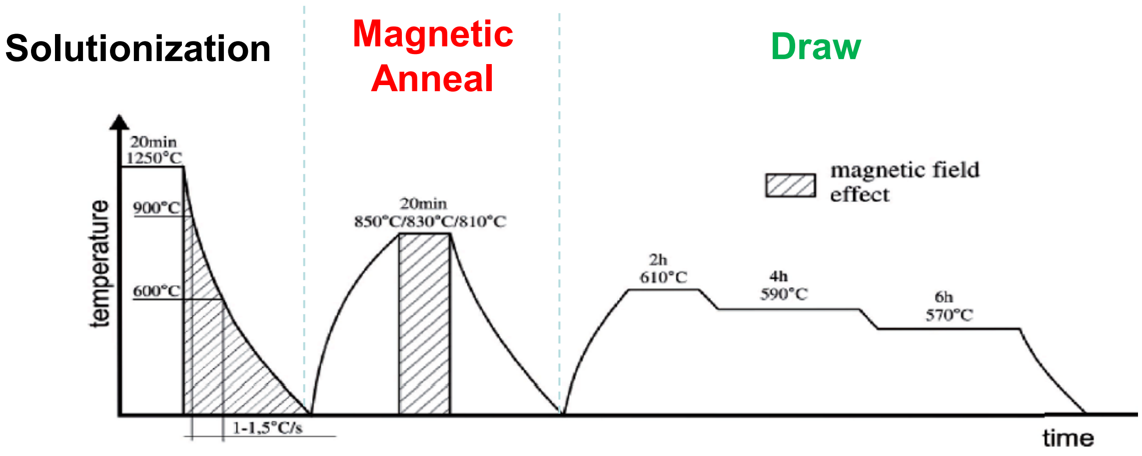
IEC Code	Class	Al wt%	Ni wt%	Co wt%	Cu wt%	Ti wt%	BHmax MGOe	Br Gauss	Hc Oersted	Hci Oersted
R1-1-13	Sintered Alnico 8H	7	14	38	3	8	4.50	6700	1800	2020
GA-194	HIP Alnico 8H	7.3	13	38	3	6.4		6200		1250

First attempt shows promise for new approach.

HIP 1250°C, 4 hrs,
60 MPa
< 20 μm powder
Furnace cooled

- Some grain boundary γ growth from slow cooling (0.5 vol.%)
- Degrades coercivity

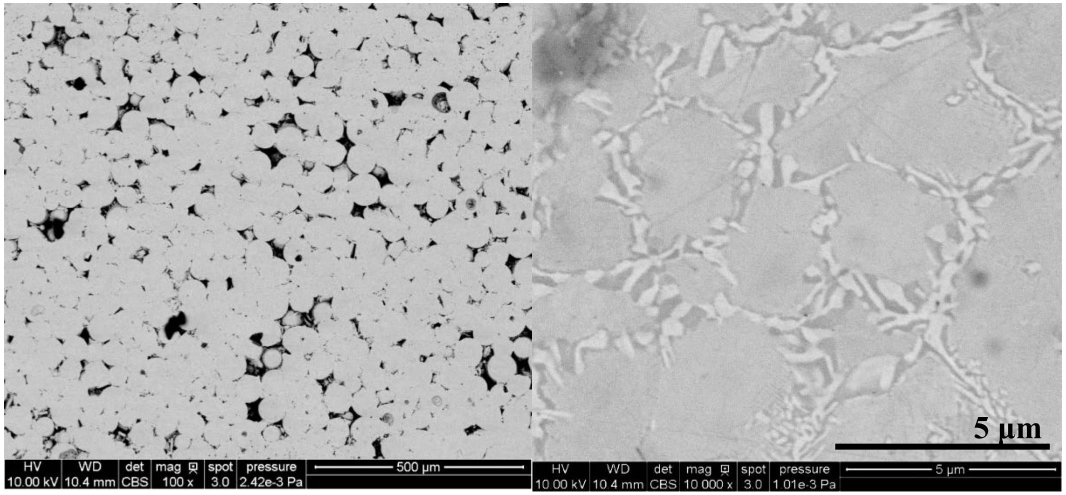
- Gamma phase forms on cooling commercial castings and HIP bars at conventional rates.
- Conventional practice to solutionize & cool with forced air to minimize gamma before magnetic annealing (MA)



- Gas atomized powders rapidly solidify to single phase alpha (B2)—**no gamma**
- **Can solution anneal be avoided for magnets made from gas atomized powders?**
- Can gas atomized powder be consolidated into magnets below MA temperature?
- Spark plasma sintering (SPS) tried to accelerate powder consolidation.

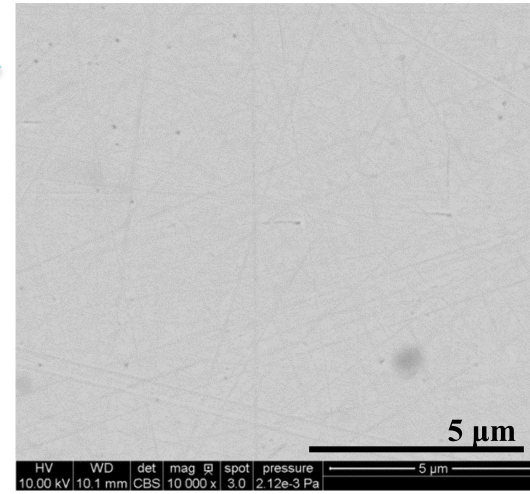
Processing Innovation for Net-Shape Alnico Magnets

Ames Lab



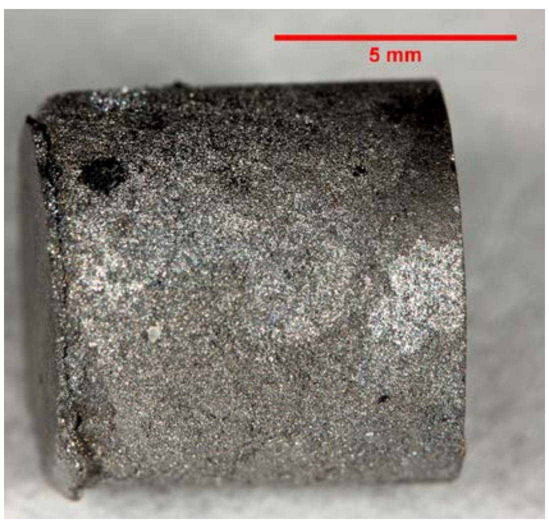
770C SPS, 5 min, 60 MPa: 91.5% dense, 18-20% gamma, poor bonding

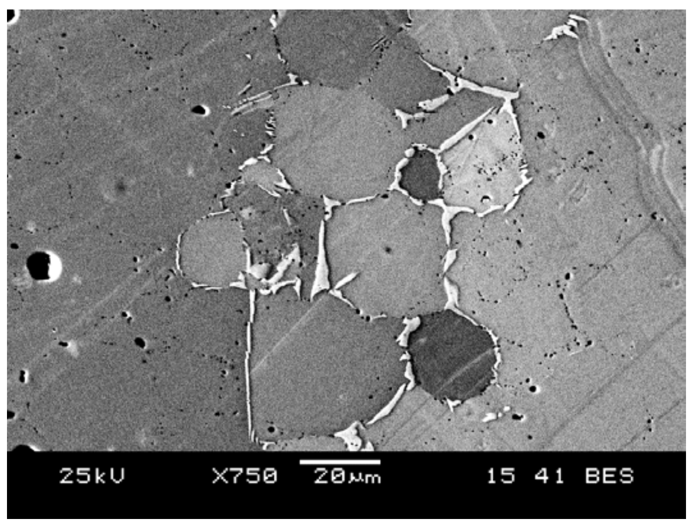
Flash anneal to remove gamma is effective



1250C 5 min anneal/water quenched

- High temperature solutionizing (1250C) needed to remove gamma phase: use for sintering
- Net-shape magnets & mass production are key objectives: try binder/compression molding





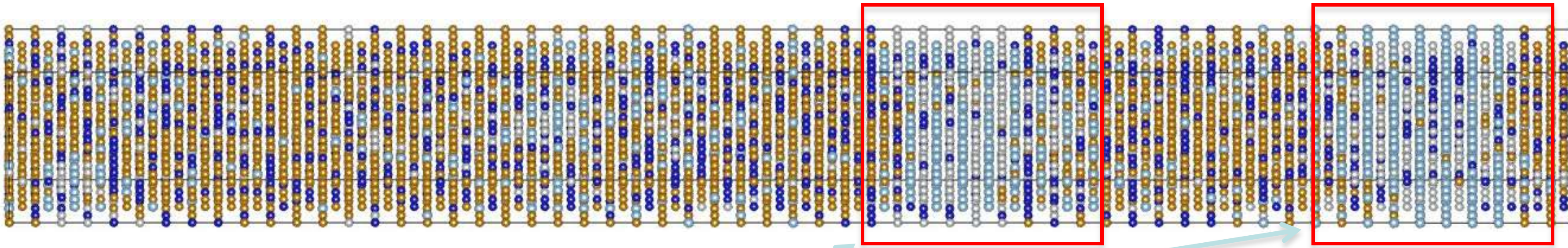
Compression molded at RT, debound at 300C/Vac. sintered 1250C, 8h, slow cooled

Average	Debinding Path	Bulk Density in g/cm^3	Total Porosity
Sample 1 (AGK-2-1)	2	7.1833	0.5725 %

- Dia.< 32µm alnico powders
- Poly-propylene carbonate binder (no carbon impurities)
- Uniform shrinkage, 99.5% t.d.
- 8mm dia. X 8mm tall, final
- Massive grain growth (to mm)
- Gamma on grain boundaries
- Need annealing/magnetics

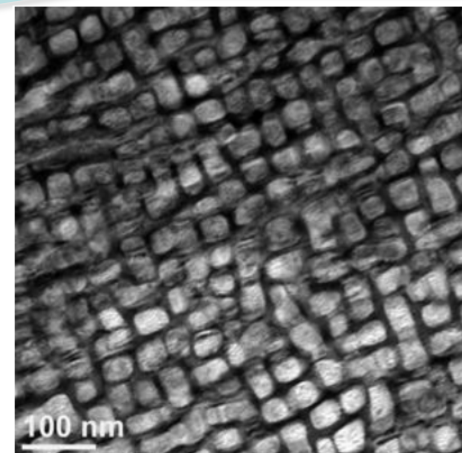
Atomic structure of interfaces by kinetic Monte Carlo (MC) calculations

Ames Lab

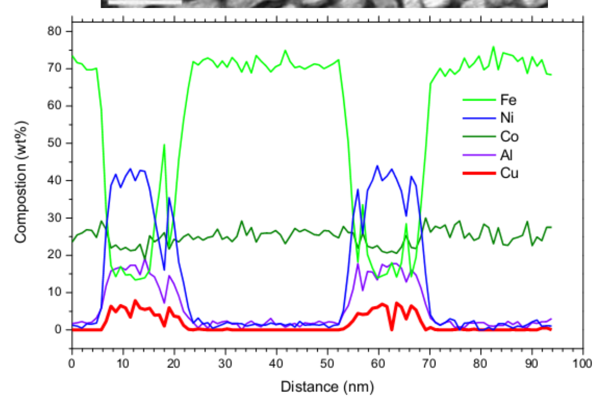


Correlate with 3DAP results: improvement from "draw" treatment

Aluminide phase



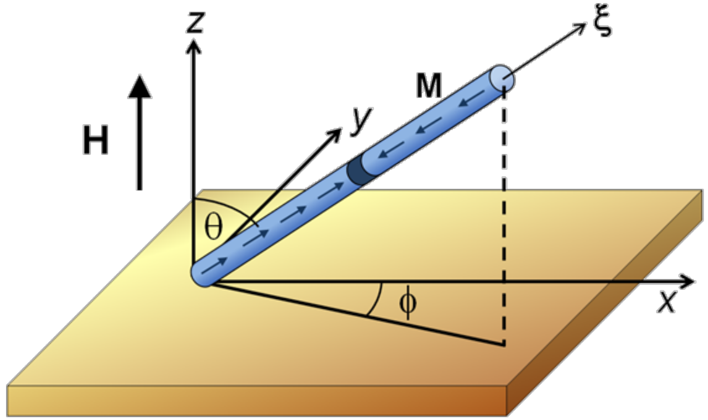
- Aluminide phases developed from spinodal with small size grains, 1 ~ 2 nm:
  - ✓ System falls into a local minimum => need new operators to MC code:
    - ○ cut-and-paste from genetic algorithm
    - ○ composition fluctuation wave from spinodal decomposition
  - ✓ Simulation supercell small compared with experiment (1/4)---will expand.



TEM and 3DAP of Alnico 5-7

Energy-Product Management in Alnico Magnet

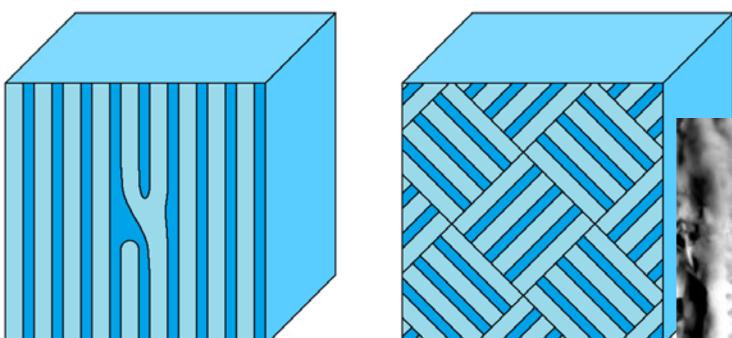
University of Nebraska



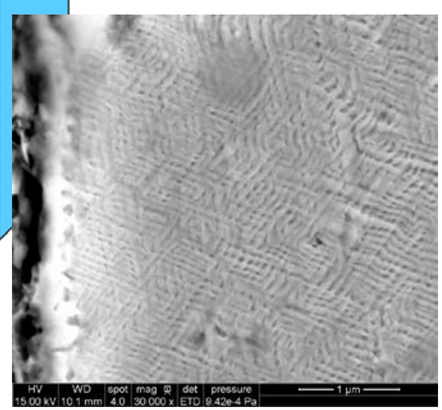
Geometry of Kondorski model

For "Small" Coercivity:

- $(BH)_{max} \sim M_r H_c$
- $M_r \sim \cos\theta$
- $H_c \sim 1/\cos\theta$



Situation in alnicos



Alnico 8H HIP/MA/drawn

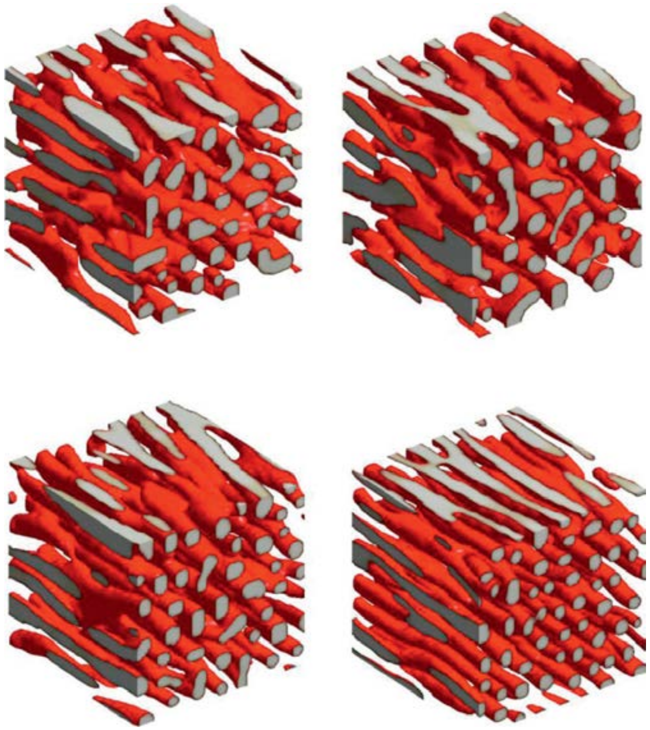
Conclusions

$(BH)_{max}$ is zero-sum game, but some freedom for motor design (shape of magnets).

Experiments on crystalline alignment effects needed (Now proposed for HTML).

Coercivity as function of Fe-Co nano-rod diameter

University of Nebraska



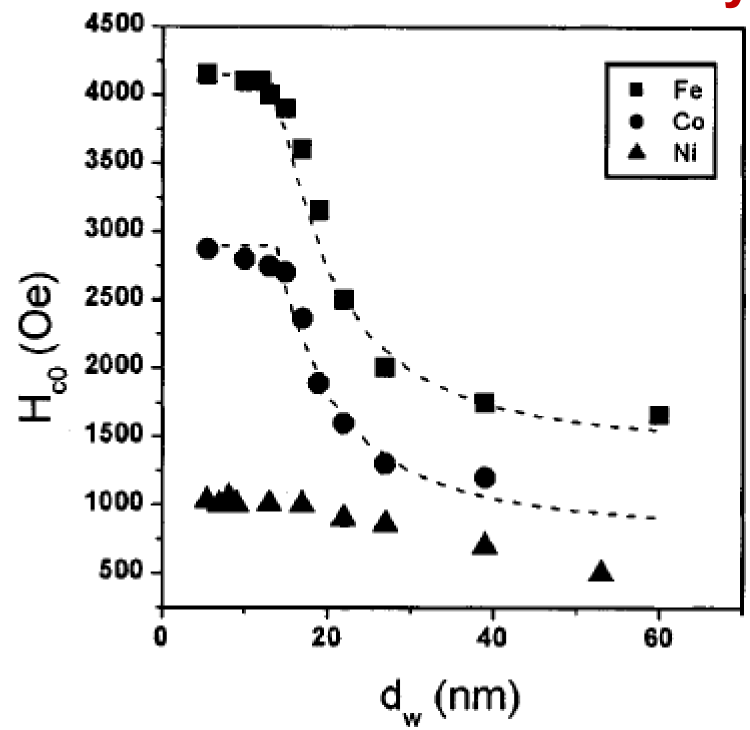


FIG. 7. Zero-temperature coercivity H_{c0} as a function of d_w for Fe and Co; the dashed lines are fits to Eqs. (9) and (10).

Meso-scale modeling predicts that more than 2X gain in coercivity for alnico is possible if spinodal spacing is maintained < 15nm, but must do controlled test

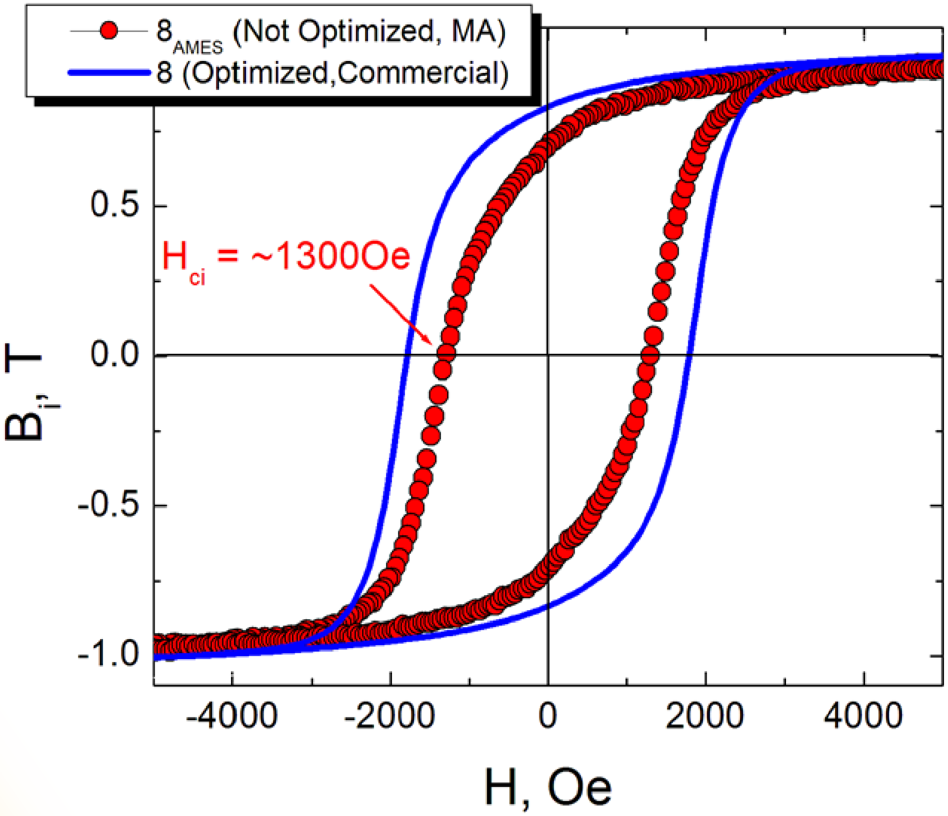
H. Zeng,[1,3,*] R. Skomski,[1,3] L. Menon,[2,3] Y. Liu,[3,4] S. Bandyopadhyay,[2,3] and D. J. Sellmyer[1,3]
PHYSICAL REVIEW B 65 134426

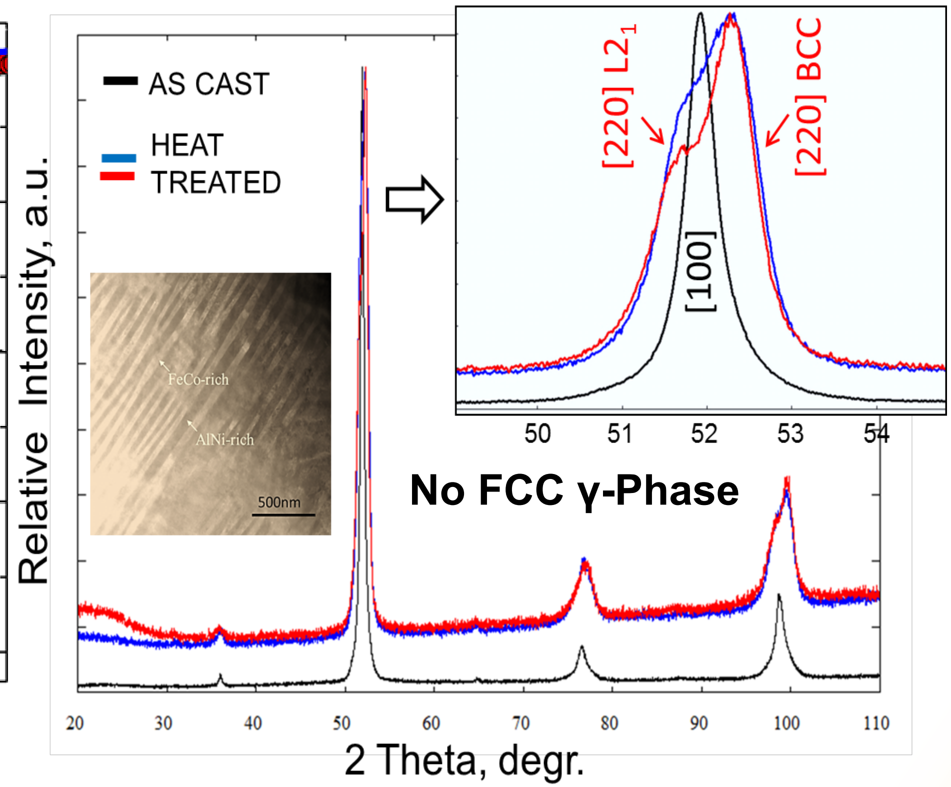
Alnico 8_{AMES}

GOAL: **Maintain or enhance** performance at a significantly reduced cost.

APPROACH: ***Novel alloying approach*** **to achieve a 40% reduction in Co content could result in a 30% cost reduction AND increased coercivity.**

INITIAL RESULTS: 1st alloy, crystallographically anisotropic, field annealed magnet.



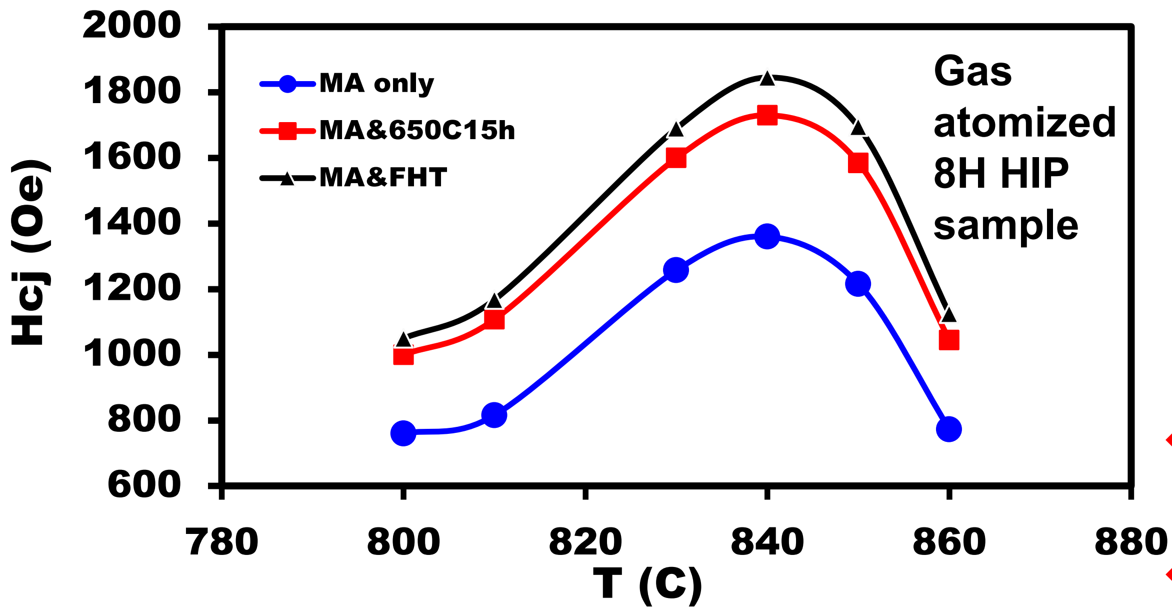


Type	BH_{Max} (MGOe)	B_r (T)	H_c (Oe)	H_{ci} (Oe)
Cast 8H	5.0	0.72	1900	2170
Sintered 8H	4.5	0.67	1800	2020
Gas Atomized HP 8H	**5.3**	**0.90**	**1530**	**1600**

Use of only 5min. at typical MA of 810C promoted reduced spinodal spacing (<30nm).

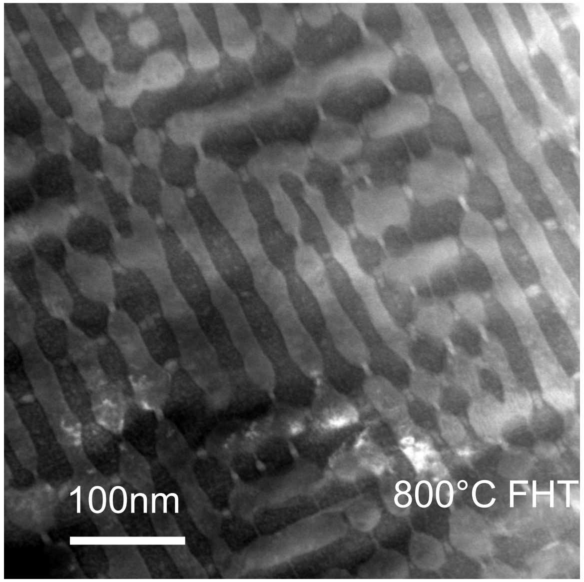
Recent Progress in Temperature Effects on Magnetic Annealing with Draw Cycle

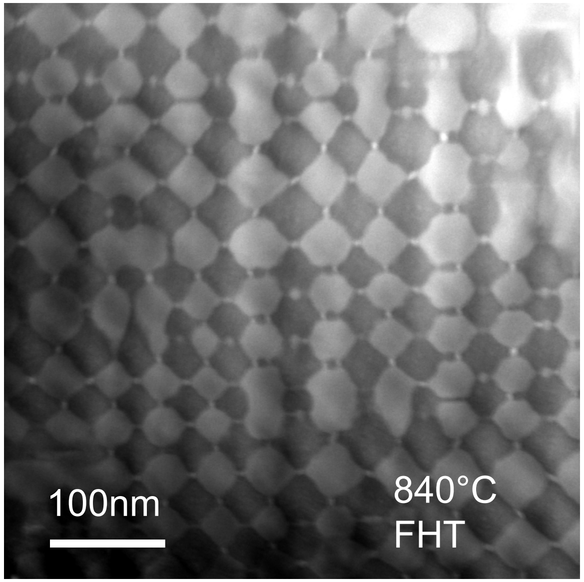
Ames Lab

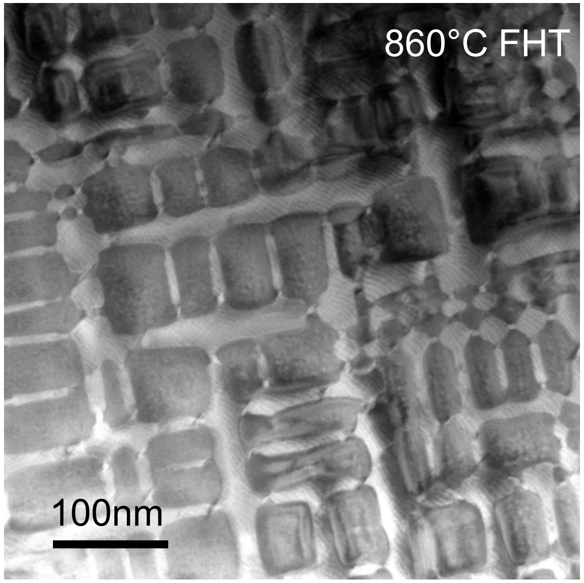


MA (10min.) + 2 Draws			
Ms (kGs)	Br (kGs)	Hc (Oe)	(BH)m
12.1	9.8	1050	3.7
11.9	9.5	1168	3.5
11.8	9.4	1690	5.2
12.1	9.6	1845	5.9
11.9	9	1695	5.4
12	8.8	1125	3.2

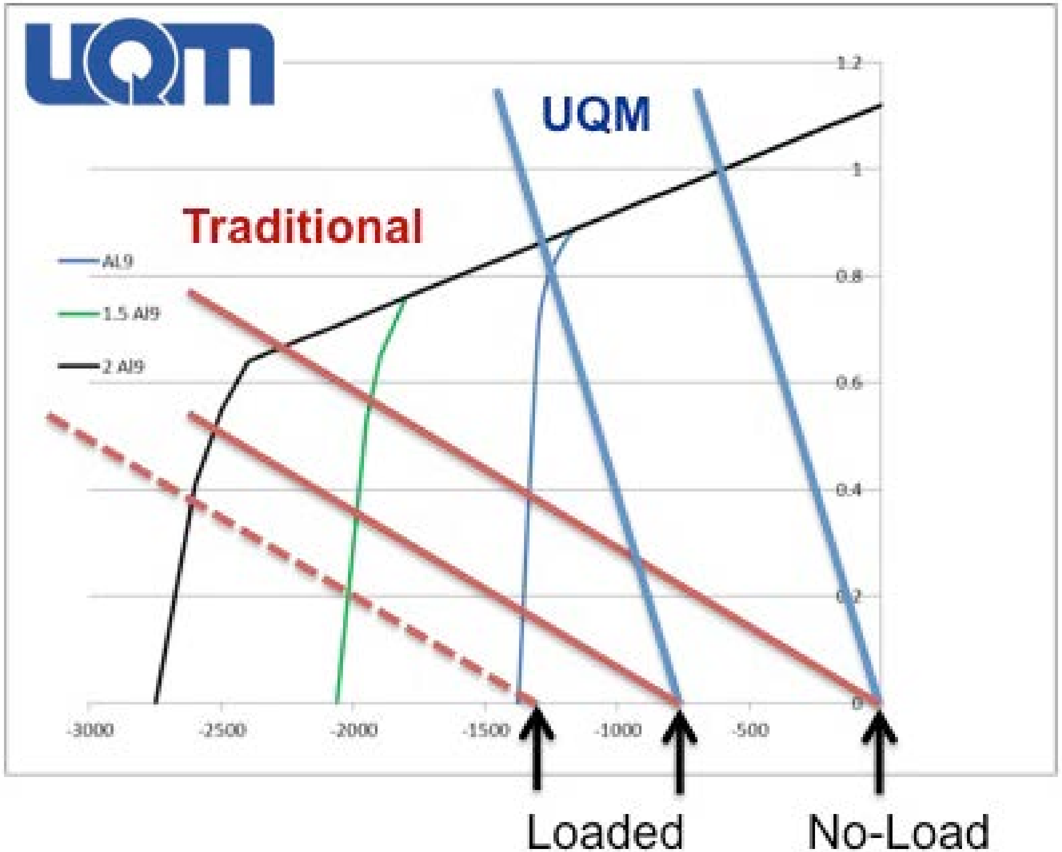
- Developed new method to accurately predict MA temperature for maximum coercivity and (BH)max.
- (BH)max 18% better than commercial

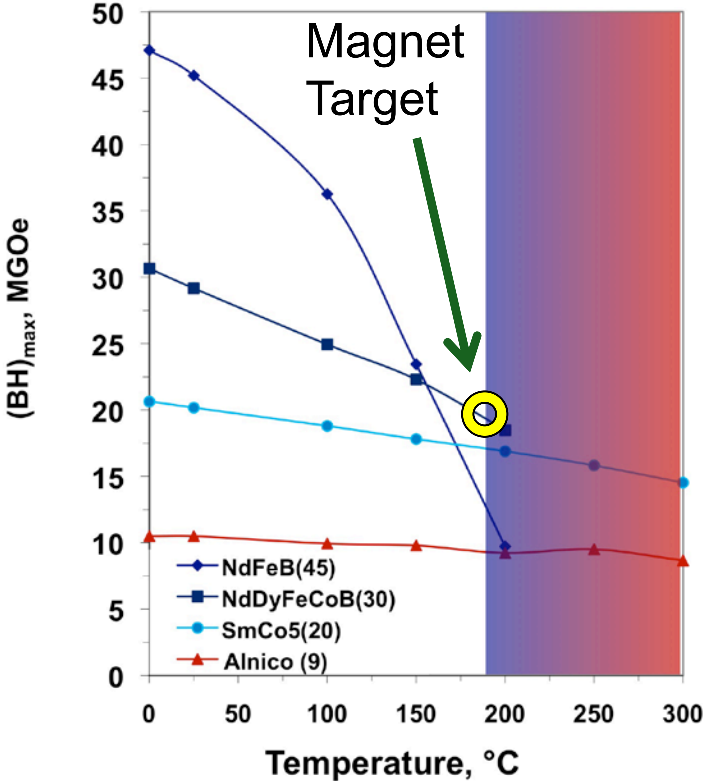






Outlook for Enhanced Alnico in Advanced PM Motor



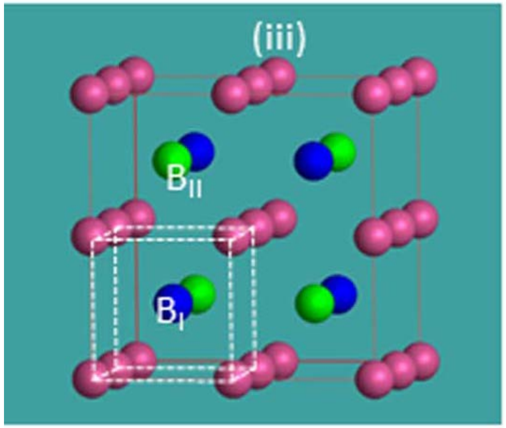


- "Operating load line of the UQM (motor) innovation is substantially higher than existing permanent magnet motors, allowing the use of lower coercivity magnets that support high flux densities." (e.g., alnico 9+)
- Improvement of alnico 8 (or 8/9) to useful range by powder processing for industrial partner's motor **is encouraging**.

Comparison: atom distribution in alnico 8 and 9 matrix phase ($L2_1$) AlNi experiment *

ORNL

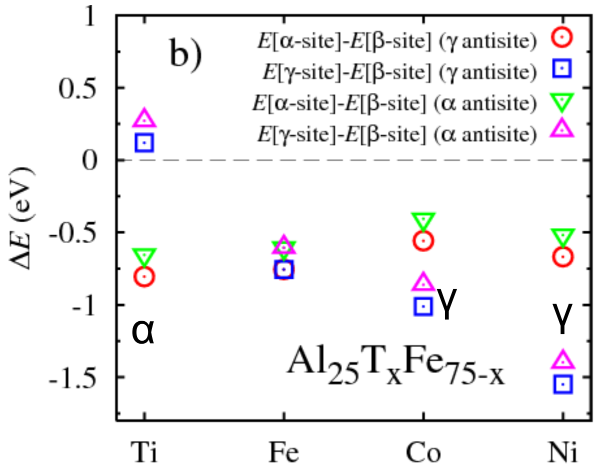
α_2 phase *

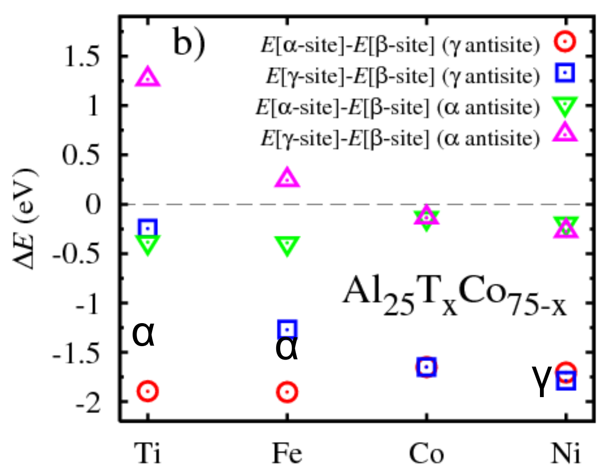


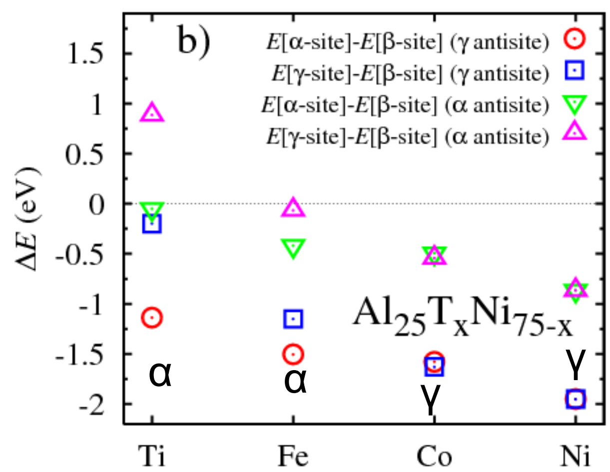
Experimental notations: *
A – γ, BI – β, BII – α
"measured" concentrations:
α(BII): $Fe_{0.20}Ti_{0.80}$
β(BI): Al
γ(A): $Ni_{0.48}Co_{0.52}$

Our work notations:
α (Fe) – (1/2,1/2,1/2)
β (Al) – (0, 0, 0)
γ(Fe) – ±(1/4, 1/4, 1/4)

DFT results







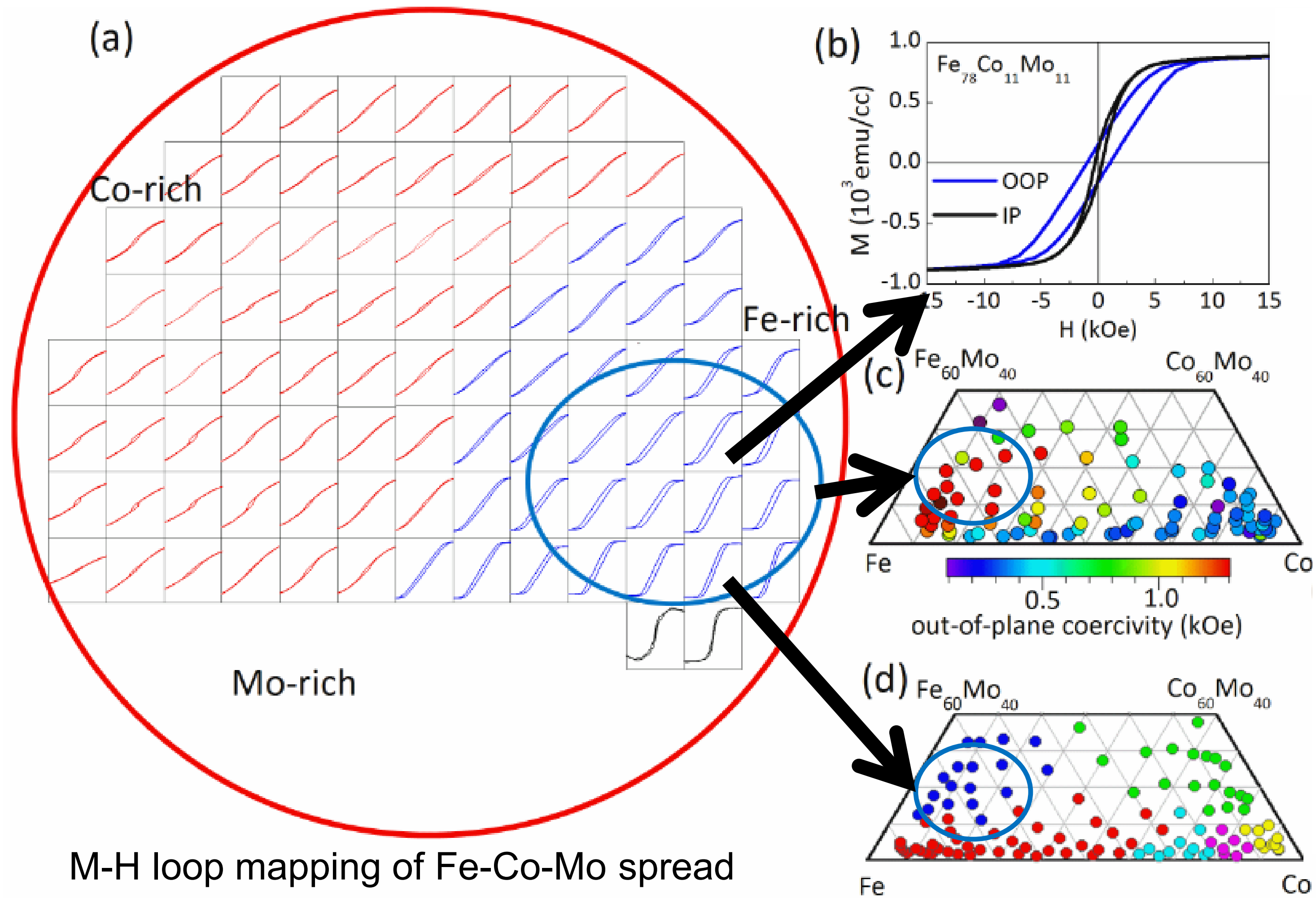
Theory gives site preferences: Ti, Fe – α; Co, Ni - γ

Expanded calculations valuable for high coercivity alloy design in alnico

*) P. Lu, L. Zhou, M. J. Kramer & D. J. Smith, SCIENTIFIC REPORTS | 4 : 3945 (2014).

Identification of tetragonally distorted Fe-Co-Mo from thin film composition spread

U. Maryland and Ames



M-H loop mapping of Fe-Co-Mo spread

From MOKE:
H_c ~ 1.2 Koe
(K ~ 30 μeV/atom)

H_c mapping

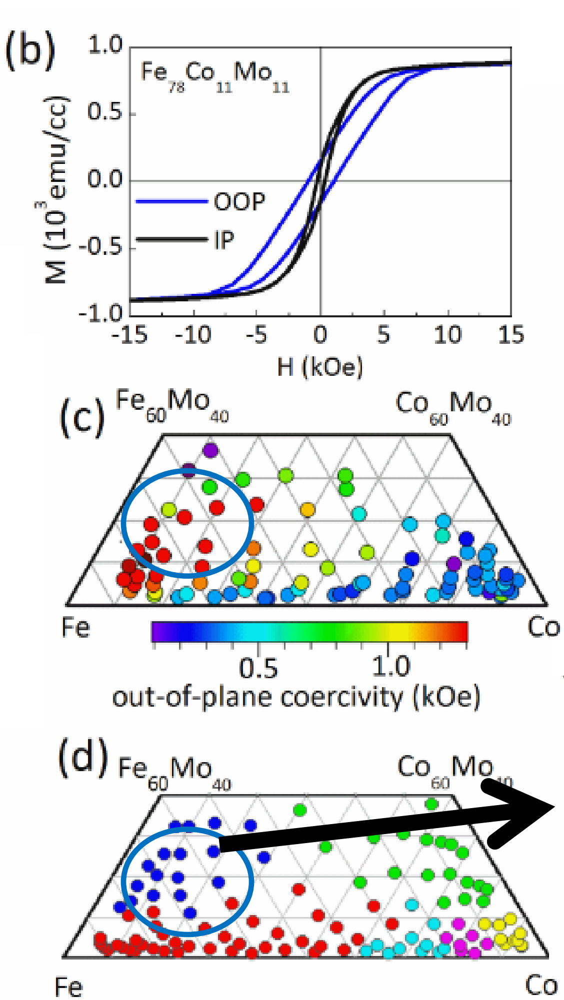
Grouping of structures based on synchrotron diffraction

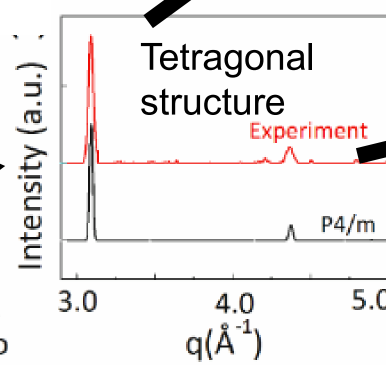
Magneto-crystalline anisotropy effect??

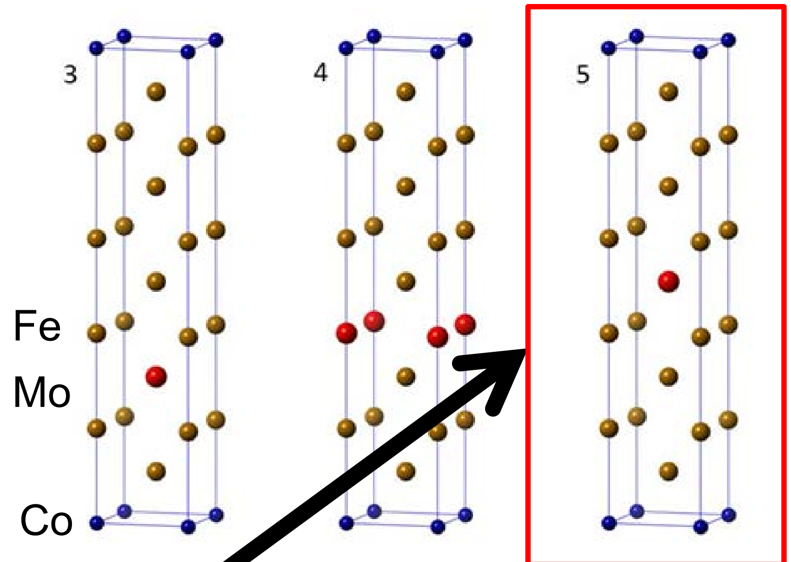
Composition region with enhanced coercive field identified (a,c); synchrotron diffraction identifies a tetragonal structure (d)

Theory predicts tetragonally distorted Fe-Co-Mo from thin film composition spread

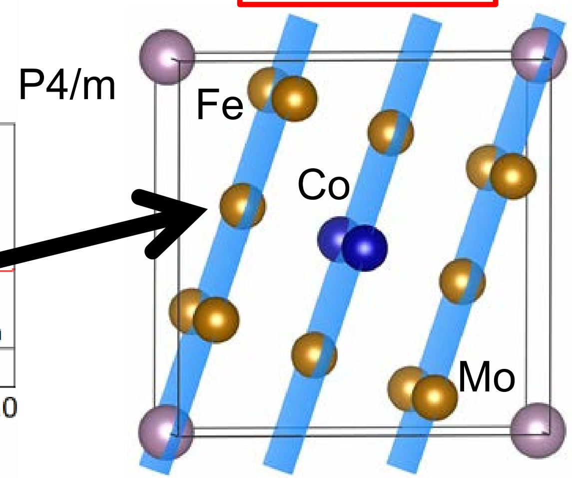
U. Maryland and Ames







Density functional theory gives K ~ 30 μeV/atom



Genetic algorithm leads to K ~ 20 μeV/atom

The identified composition Fe_8CoMo has a tetragonal structure; Genetic algorithm and DFT give K values agree with experiment.

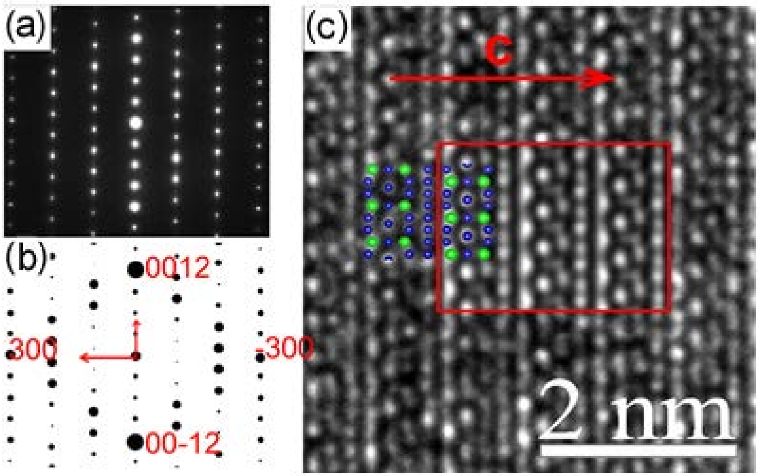
Possible boost in coercivity for alnico?

Hard Magnetism of Metastable Zr_2Co_{11}

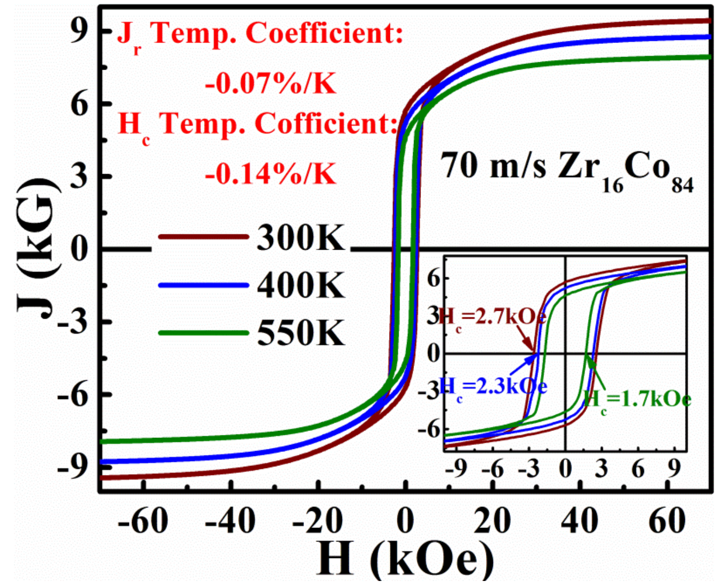
University of Nebraska

Identification of Hard-Phase Structure

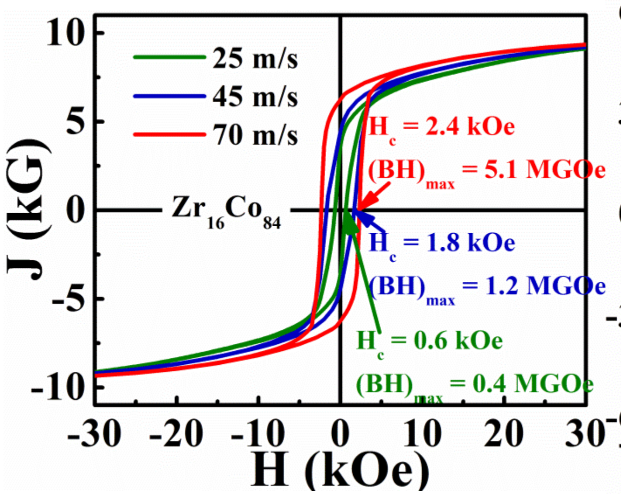
Rhombohedral (a = 4.5; c = 24.2 Å)

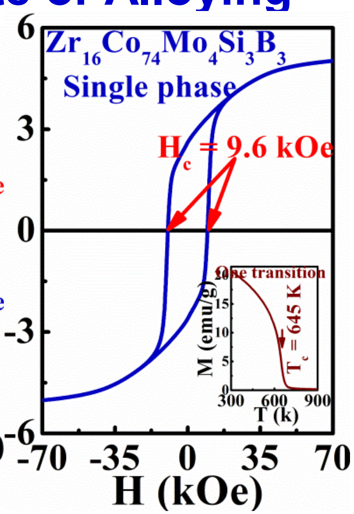


High-Temperature Magnetism



Tuning of Hard-Phase Content by Varying Quench Rate or Alloying





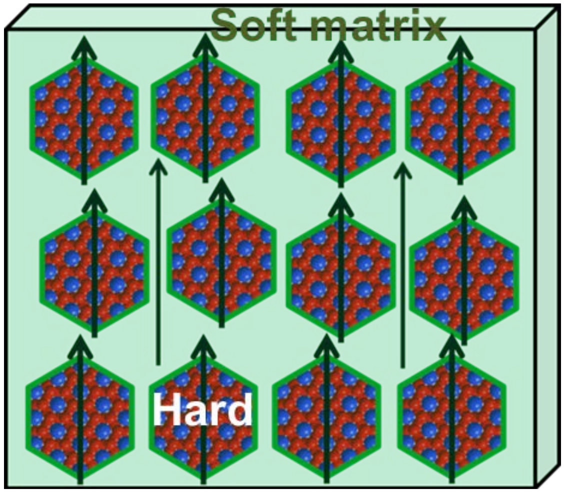
- The crystalline structure of the hard phase was identified.
- Single phase was obtained; $(BH)_{max}$ for isotropic "Zr_2Co_{11}" is 5.1 MGOe.
- Low temperature coefficients of J_r and H_c in 300~550K were found.
- $(BH)_{max}$ may be improved by hard-phase grain alignment—by SSHD??

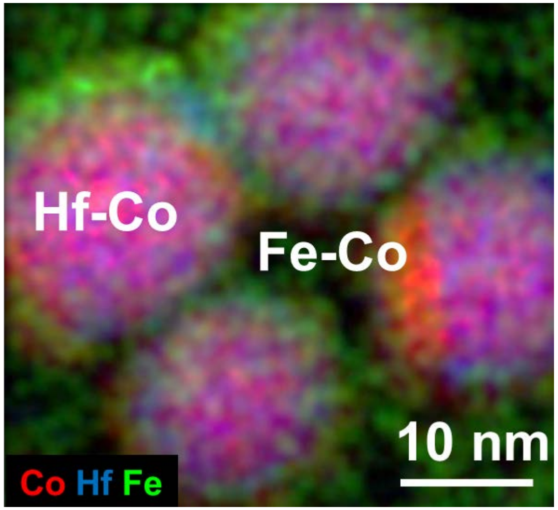
C.Z. Wang et al. Phys.Rev.Lett.112, 045502(2014). D.J. Sellmyer et al. J. Phys. D: Appl. Phys.46,135004(2013); J. Phys.: Cond. Matt.26, 064204(2014); J. Appl. Phys. 115, 17A739(2014); Mater. Sci. Eng. B 186, 64(2014); J. Alloy. Compd 587, 578(2014); IEEE Trans. Mag. 48, 3603(2012).

Aligned Two-Phase Nanocomposite Magnets

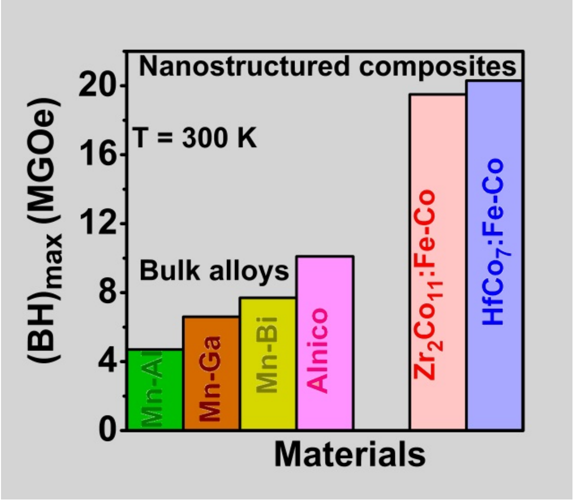
University of Nebraska

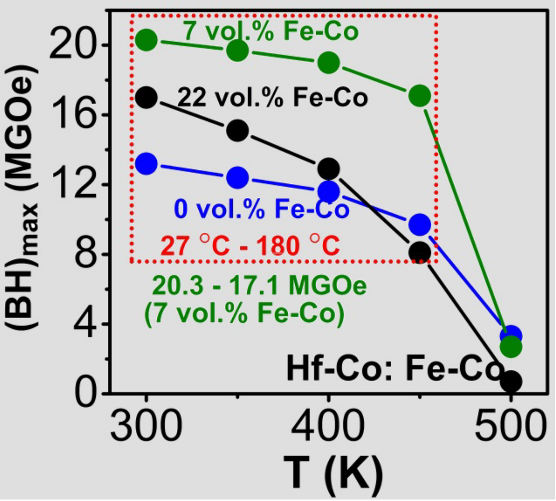
Fabricated by 2-stage cluster deposition process.





Hard phase: aligned Zr_2Co_{11} or $HfCo_7$ with soft phase: $Fe_{65}Co_{35}$





❖ **High energy product maintained up to 180 °C at preferred vol.% Fe-Co**

B. Balamurugan, B. Das, R. Skomski, W.Y. Zhang, D.J. Sellmyer

Appl. Phys. Lett. **101**, 122407 (2012); *Adv. Mater.* **25**, 6089(2013), *J. Phys. Condens. Mater.* **26**, 064204 (2014)

Response to Previous Year Reviewers' Comments

- In assessing the approach," the reviewer warned that for the second milestone in September 2013, a more solid validation study is necessary before using Genetic Algorithm (GA) to search for the best structure based on computer simulations….the reviewer cautioned that the magnetic properties of the new selected Fe-Co-X alloys calculated from the model may mislead the material design.

We agree that theory and experiment should be closely coupled to select the most fruitful direction to proceed. In fact, density functional theory (DFT) was used to estimate the intrinsic magnetic properties of each of the phases that were identified in the GA structural stability calculations, which were compared to the experimental diffraction patterns. The biggest experimental challenge to verification of the DFT magnetic property results was to obtain a single phase sample to measure in a magnetometer, especially with (at least) 2 metastable phases that were so close in stability. Some very recent work (report herein) that involved off-stoichiometry binary alloys with extremely high quench rates and significant alloy additions (Mo, Si, and B) were able to obtain a single phase rhombohedral structure and to verify the expectations of significant magnetic properties.

- In proposed future research, the reviewer expressed that the work is headed in the right direction, which bodes well for future research…..but that it would help to have more tangible goals (e.g., target magnet parameters) to allow industry to evaluate the impact of the new magnets…..on future motor designs…..even a rough idea of the possible improvements is helpful.

We certainly would like to provide useful predictions, but we must be guided by the magnetic measurements that result from the various microstructure, composition, and structure modifications that we are able to achieve. For example, we can achieve a relatively high magnetization in alnico, but we have not been able to expand the coercivity. Thus, among other things, we are thinking of purposefully lowering the fraction of Fe-Co phase, source of M(sat), to increase separation between these rods, which could decrease magnetic interactions and increase coercivity.

Collaboration and Coordination with Other Institutions during FY2014

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

BREM Team: B. Harmon, K.M. Ho, C.Z. Wang, V. Antropov, and R. Napolitano: AL

R. Skomski, D. Sellmyer and J. Shield: Univ. Nebraska-Lincoln

M. Stocks: ORNL

I. Takeuchi: Univ. Maryland

S. Constantinides: Arnold Magnetic Technologies, Inc.

Collaborators:

- Baldor (Mike Melfi): Electric motor manufacturing technology, BREM (VT) technology adviser.
- Univ. Wisconsin-Madison (Tom Jahns): Electric machine design, BREM (VT) technology adviser.
- Synthesis Partners (Chris Whaling): Automated search of permanent magnet literature, BREM project (VT) adviser.
- General Electric (Frank Johnson): Rare earth magnet technology and motor design, 2012 VT-PEEM Motor/Magnet partner (prime).
- Unique Mobility (Jon Lutz): Advanced motor design, 2012 VT-PEEM 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 Mathewson): Fe nitride PM, ARPA-E partner (prime)

UQM TECHNOLOGIES

imagination at work

ARNOLD MAGNETIC TECHNOLOGIES

Remaining Challenges and Barriers

Coercivity levels and maximum energy product values achieved thus far in experiments are insufficient to permit alnico magnet use in an advanced PM traction drive motor.

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 processing and alloying changes to make bulk magnets that achieve desired changes in microstructure and magnetic properties.

Establish capabilities of binder-assisted compression molding to fabricate alnico magnets in prototype sizes and shapes with significant magnetic properties.

To enable extensive experiments on compression molding and other bulk magnet fabrication methods, a new batch of gas atomized pre-alloyed powder must be produced with high purity and desired composition.

Outline of Future FY14 Plans

- Investigate influence of microstructure and composition features identified by theoretical analysis to further enhance bulk alnico magnets., e.g., test reduced spinodal phase spacing, influence of chemical gradient at Fe-Co/matrix interface, and influence of grain alignment/field direction.
- Establish methods to mitigate experimental issues with gamma phase formation at grain/cell boundaries.
- Perform sufficient experiments to select Cu concentration for low Co alloy to permit formation of well developed spinodal pattern.
- Perform intensive experiments on compression molding of alnico bulk magnet shapes, including solutionizing and magnetic annealing to estimate range of size scaling, i.e., for debinding/quenching/heating.
- Produce fully pre-alloyed alnico powder with modified alnico 8 composition, and consolidate to full density with magnetic annealing and heat treatments designed on the basis of improved understanding of magnetic property optimization.

Summary of FY13-14 Presentation

- Performed relative cost analysis of SSHD method for making fully dense anisotropic magnets from Zn-coated glassy RE-based (low Dy) magnet flake that revealed better cost and effectiveness for this method, but need for advanced RE alloy for best test.
- Studied processing of pre-alloyed powder-based alnico type 8H, along with chill cast rods and melt spun ribbon samples:
  - Discovered that as-atomized powders (and melt spun ribbons) were single phase (B2), but that cold pressing failed due to lack of plastic deformation (not shown).
  - Attempts to consolidate powders by spark plasma sintering well below 1250C was unsuccessful due to lack of sintering and to gamma phase formation
  - HIP consolidation provided fully consolidated samples for heat treating studies since solutionization and quenching apparently erased previous microstructure.
  - Quenching that suppressed gamma suggested the possibility of binder-assisted compression molding, which was demonstrated in preliminary testing.
  - Initial results of kinetic Monte Carlo show temperature dependence of spinodal reaction more accurately
  - HT of HIP samples indicated direct influence of draw cycle on coercivity.
  - Effective method to find maximum magnetic annealing temperature was discovered, producing improved magnetic properties over commercial levels.
  - Performed meso-scale theoretical analysis on desired reduction of spinodal scale.
- Results from studies of Co-X systems (X=5d and 4d, e.g., Zr, Mo) with combinatorial synthesis and melt spinning agreed with earlier theoretical predictions.

Technical Back-Up Slides

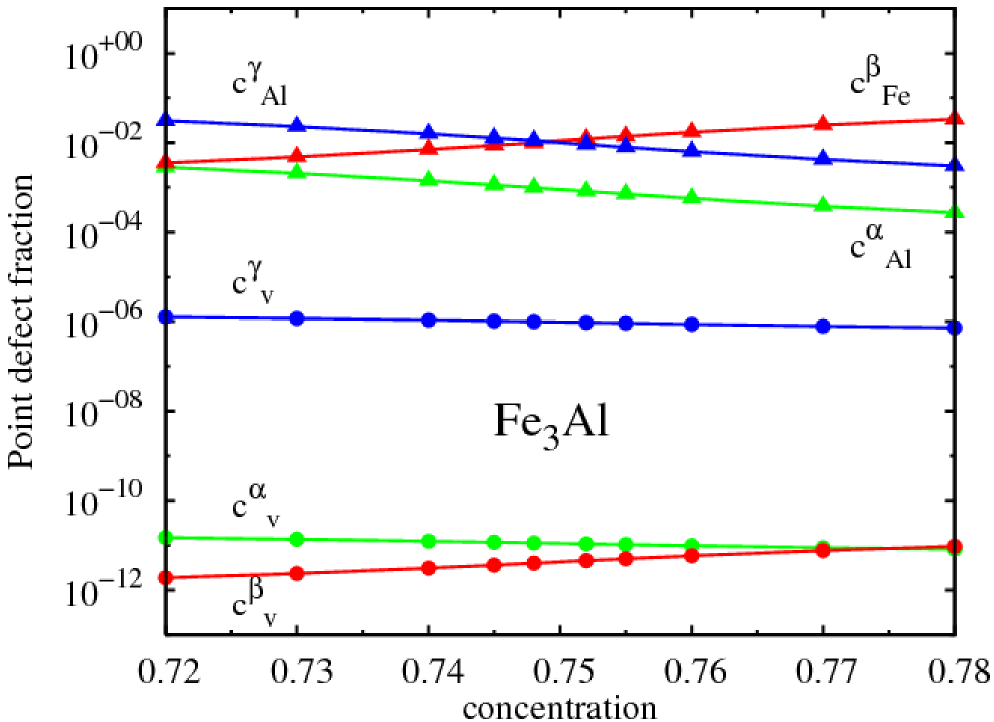
Ternary element site preference (DO_3) Fe_3Al, Co_3Al, Ni_3Al *

ORNL

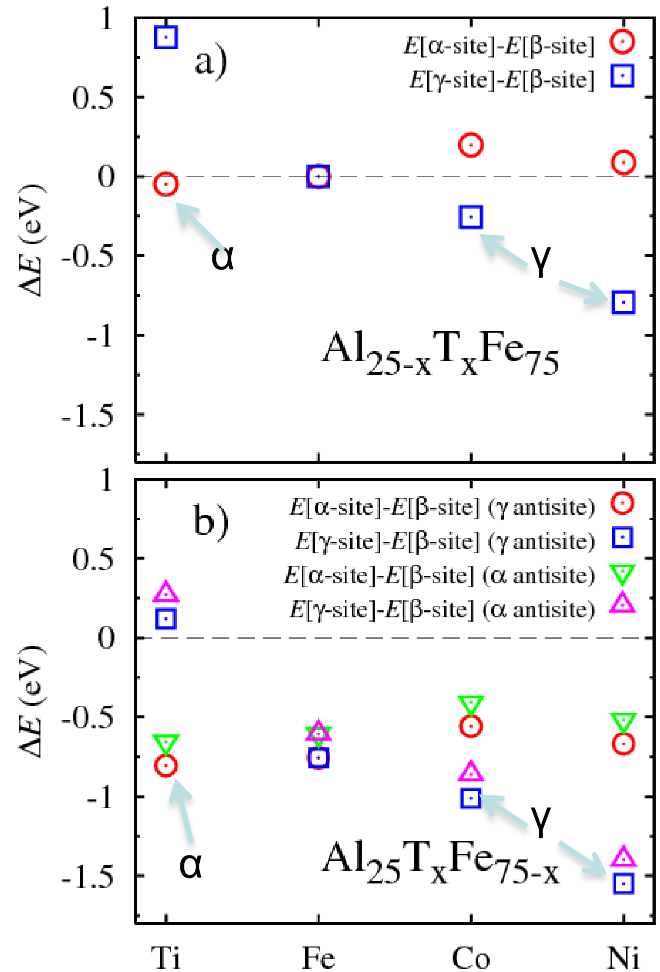
If α and γ sites are occupied by atoms with the same probability $L2_1$ becomes to DO_3

α (Fe) – (1/2,1/2,1/2)
β (Al) – (0, 0, 0)
γ(Fe) – ±(1/4, 1/4, 1/4)

Dependence of defect concentration on the composition at T=1000 K



Main defects in Fe_3Al correspond to anti-sites!



Ternary elements prefer Fe sub-lattice: Ti – α ; Co & Ni - γ

Valuable for alloy design decisions in alnico phases.

*) Samolyuk, Ujfalussy, Stocks, paper submitted for publication.

Wang-Landau Monte Carlo and Cluster Expansion

ORNL

Work in Progress for high performance Monte-Carlo calculation in cluster expansion:

•Use Wang-Landau Monte Carlo sampling to calculate the density of states g(E) (Micro-canonical Entropy) for all energies.

- Free Energy for fixed concentration (Canonical Ensemble) at all temperatures: **critical to apply to high temperature phases, e.g., alnico.**

•Replica Exchange Wang Landau [PRL 110, 210603 (2013)] allows for efficient parallelization of the g(E) calculation. This code has been implemented in the context of spin models and is being integrated with the cluster expansion **for multicomponent alloys, like alnico.**

- This algorithm allows super-linear speed up over conventional Wang-Landau sampling, reducing the number of energy evaluations needed

•Re-implement the energy calculation for Monte-Carlo, to be more efficient to evaluate **to radically reduce total calculation times.**

- Traditionally the cluster expansion contributions are expressed in coefficients for cluster functions. We pre-compute tensors indexed by the atomic species by expanding these cluster functions around a central site and recording its multiplicities and symmetries. Thus the energy difference entering the Monte Carlo acceptance can be efficiently evaluated by changing the site occupancy one at a time with the minimal number of sites contributing to this energy change. This allows for efficient evaluation on modern computer architectures