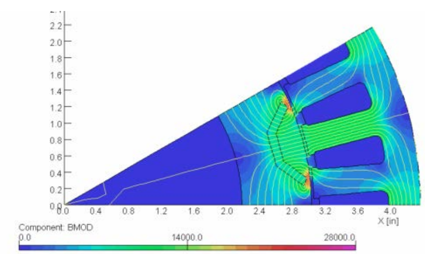
Permanent Magnet Development for Automotive Traction Motors

Includes: ***Beyond Rare Earth Magnets (BREM)***

Iver E. Anderson

Ames Laboratory (USDOE)



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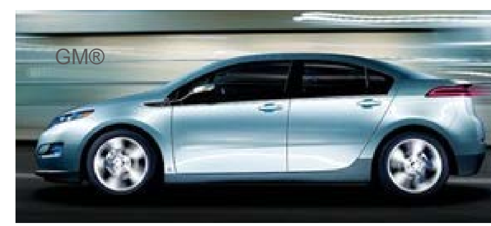


May 15, 2013

Project ID: APE015

This presentation does not contain any proprietary, confidential, or otherwise restricted information

Overview

Timeline

- Start - August 2001
- Finish - September 2015
- 90% Complete

Budget

- Total project funding
  - DOE share $12,950K (since FY01)
- FY12 Funding - $2400K
- FY13 Funding - $2400K (planned)

*2020 VT Targets

Barriers & Targets*

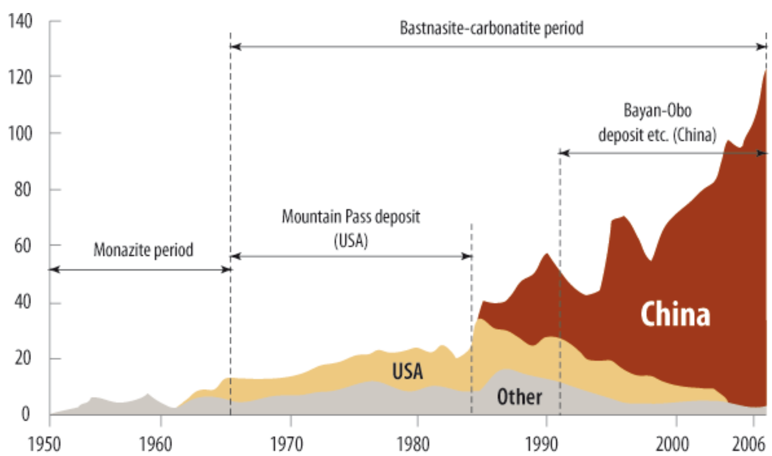
- High energy density permanent magnets (PM) needed for compact, high torque drive motors (specific power >1.4kW/kg and power density >4.0Kw/L).
- Reduced cost (<$8/kW): Efficient (>94%) motors require aligned magnets with net-shape and simple mass production.
- RE Minerals: Rising prices of Rare Earth (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

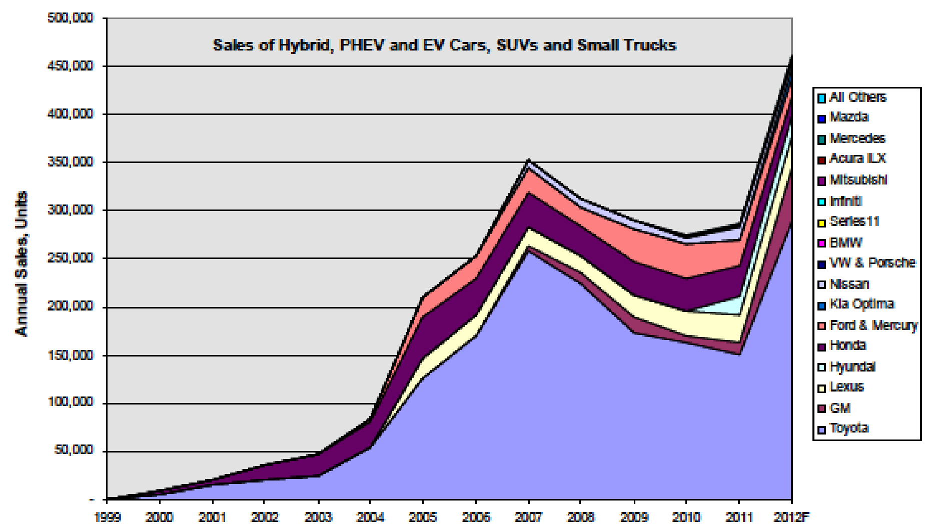
- Baldor, U. Wisconsin, GM, GE, UQM, Synthesis Partners (collaborators)
- ORNL, U. Maryland, U. Nebraska, Brown U., Arnold Magnetic Tech. (BREM subcontractors)
- Project lead: Ames Lab

Project 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.
- The fully developed PM material must:
  - ✓ remain competitive at room temperature with current high energy (MGOe) magnets to conserve valuable materials and contain cost.
  - ✓ minimize or eliminate use of scarce RE, e.g., Dy, due to an impending world wide RE shortage, or be developed as RE-free magnet alloys
  - ✓ achieve superior elevated temperature performance (150-200˚C) to minimize or eliminate motor cooling needs.



The World's Production of Rare Earth Metal Oxides (in 1,000 t). Source: USGS



Data for USA; www.hybridcars.com

Project Relevance

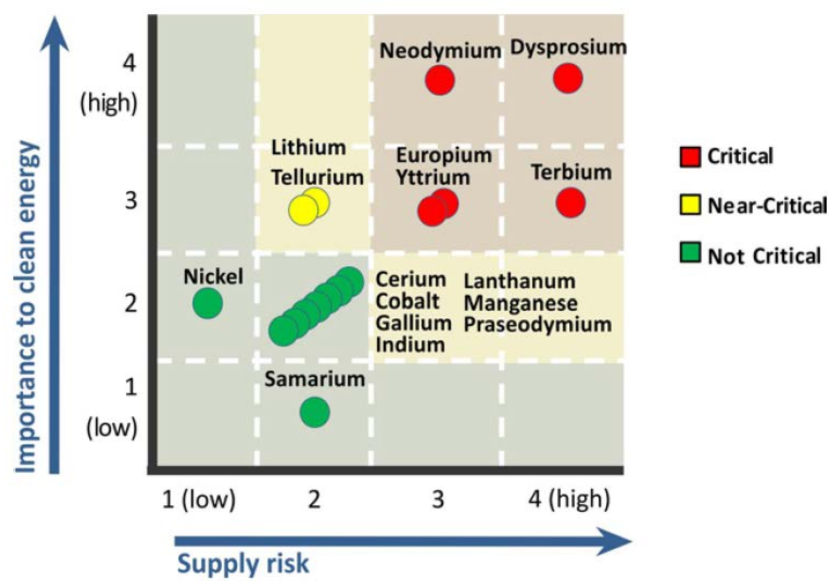
Parallel Pathways

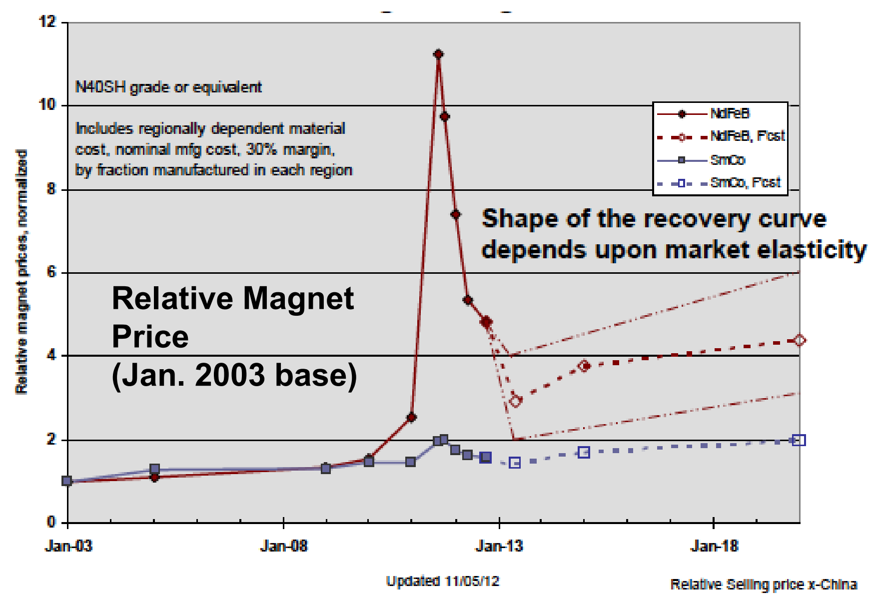
- Further develop anisotropic sintered RE magnets to achieve highest energy product (4-6X isotropic bonded) with high temperature stability & little or no Dy.
- Develop non-RE permanent magnets with sufficient coercivity and energy product for advanced internal permanent magnet (IPM) traction motors.

Uniqueness and Impacts

- Increased RE costs and on-going reduction of import quotas 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.

- US/EU/Japan filed WTO case on March 13, 2012, on Chinese restraint of RE trade.





Milestones for FY2012 and FY2013

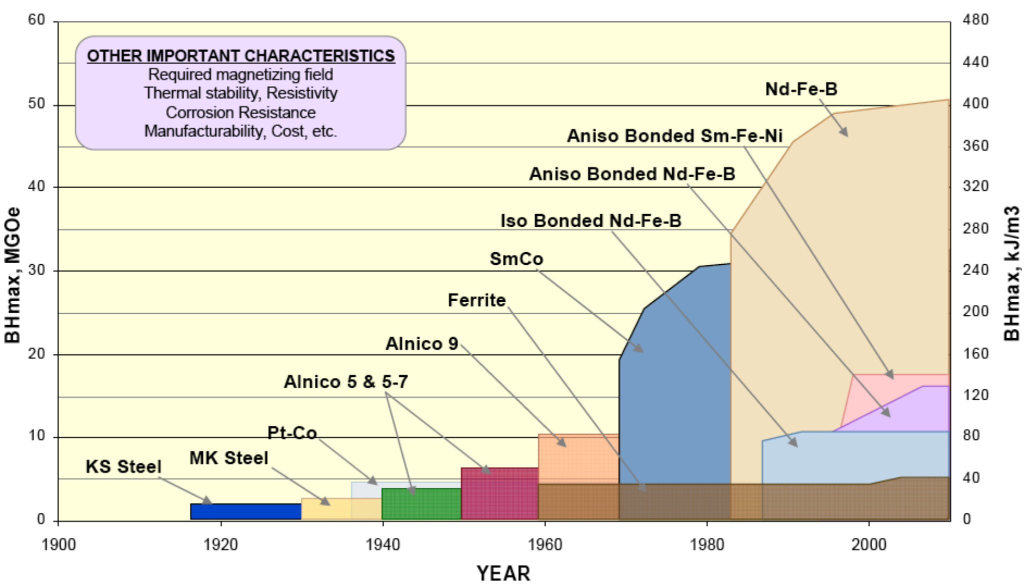
Month/Year	Milestone, Status, and/or Go/No-Go Decision
Sep-2012	**Milestone:** Complete analysis of commercial alnico: alloy influences on spinodal partitioning and ultimate reduction of spinodal scale. **[Complete]** **Milestone:** Theoretical tools expanded for the investigation of potential new Fe-Co-X phases on clusters and supercomputers. **[Complete]** **Milestone:** Extend experimental investigations of Co-X, Fe-X, & Fe-Co-X systems (X=e.g., W, Ta, Mo, Hf, Zr) with combinatorial synthesis, cluster deposition, and chemical synthesis **[Complete]**
May-2013	**Milestone:** Conduct eighth regular BREM workshop with team to exchange results and refine directions for non-RE magnet work. **[On schedule]** **Milestone:** Expand coercivity enhancement study in alnico 8 consolidated from pre-alloyed gas atomized fine powder, exploiting observed micro/nano-pinning of magnetic domains in mis-aligned grains. **[On schedule]**
Sep-2013	**Milestone:** Demonstrate at least one mechanism for improving alnico magnetic properties by comparison of magnetic measurements with microstructure observations and supportive theory. **[On schedule]** **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. **[On schedule]** **Milestone:** Synthesize bulk samples of Co-X, Fe-X, & Fe-Co-X (X=e.g., W, Ta, Mo, Zr) with cluster deposition and/or melt spinning. **[On schedule]**

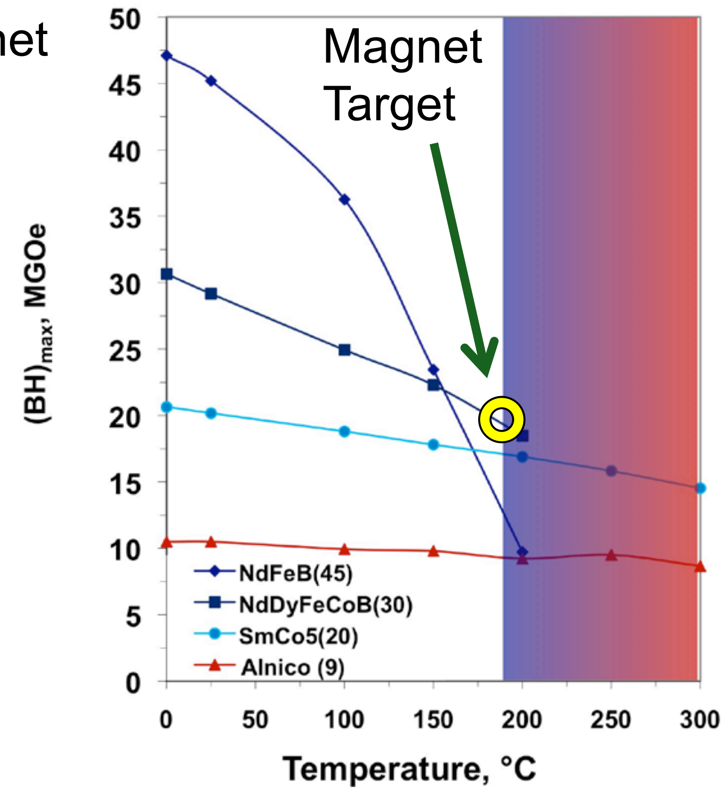
Approach/Strategy: Parallel Paths & Timing

Near-term RE Magnets: Anisotropic mixed rare earth (Nd-based) magnets will be developed with high temperature (HT) stability & **little or no Dy**, using, e.g., Y substitutions and simplified processing to near-net shape magnets.

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

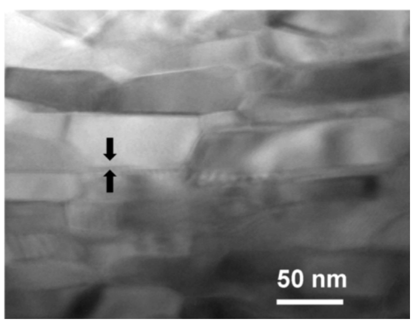
Long-term non-RE Magnets: New Fe-Co-X magnet systems will be sought by theoretical efforts, experimental surveys of magnet compounds, and prototype magnet fabrication and characterization.

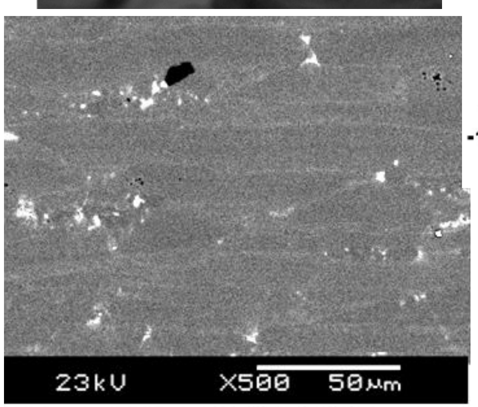


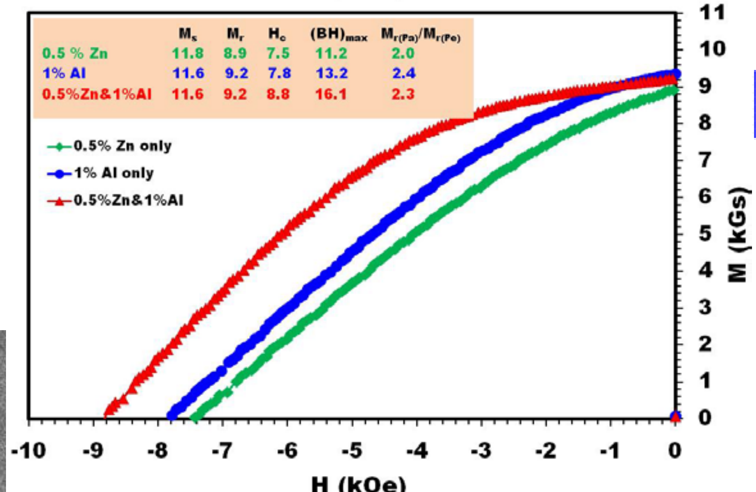


Development of Single-Stage Hot Deformation for Anisotropic Magnets from Zn (+Al) coated MRE-Fe-B Flake Powder (McCallum, Tang @ Ames)

- 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) to produce fully dense magnets from Nd-Fe-B alloys.
- 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 with HT stability and low Dy.
- Full density and stress-induced platelet grain growth (~30-60nm by ~150-300nm) results in 10.8 kOe and 21.3 MGOe for 1% Zn. With only 3wt.% Dy & 1%Al added, had 8.8 kOe and 16.1 MGOe.

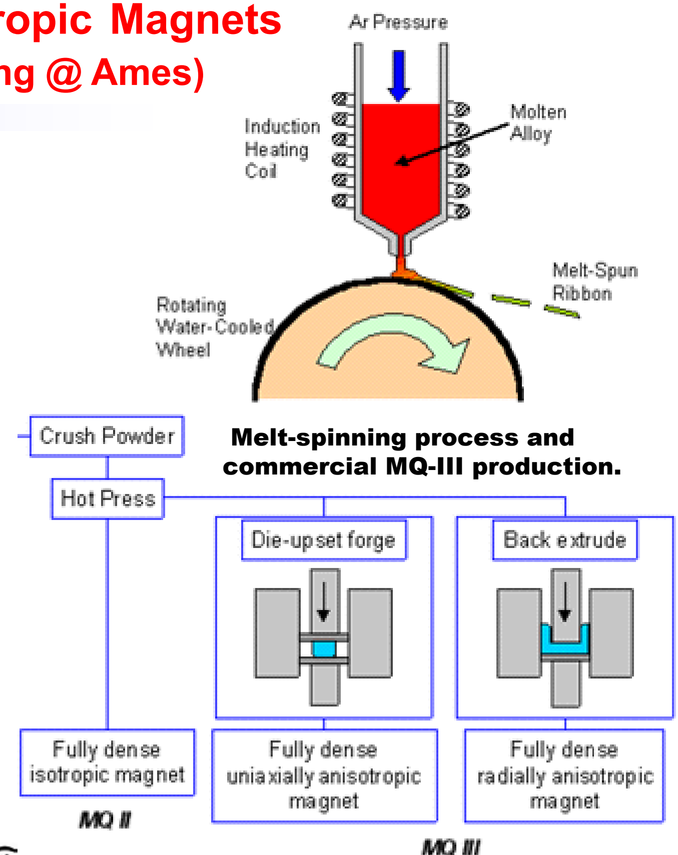






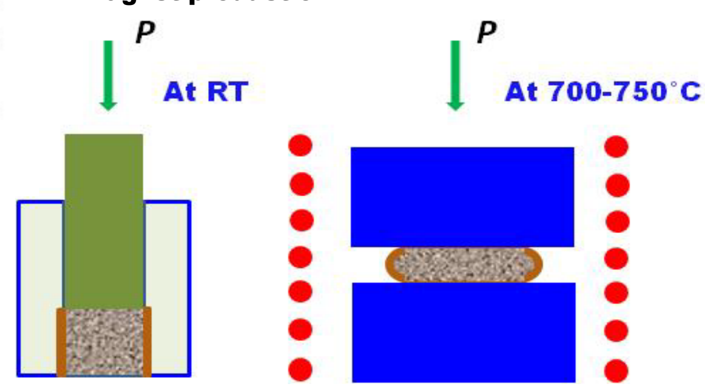
◆TEM to probe interface reactions with Zn and Al for further reduced Dy (from 5 wt%, down to ~3 wt% Dy).

W. Tang et al, "Anisotropic Hot-Deformed MRE-Fe-B Magnets With Zn Powder addition", IEEE Trans. Magn. Vol. 48, 2012(in press).



Melt-spinning process and commercial MQ-III production.

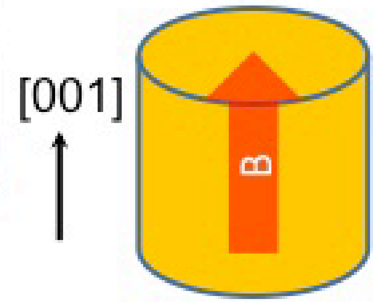
Melt spun flake and single stage hot deformation (SSHD) for anisotropic sintered magnet production.

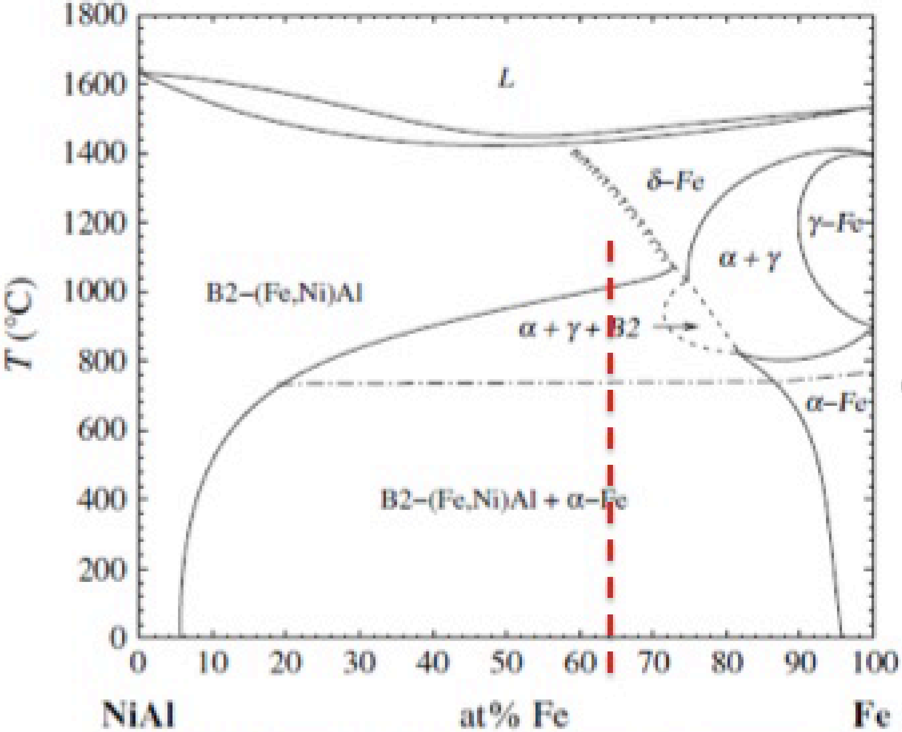


Alnico Design/Processing

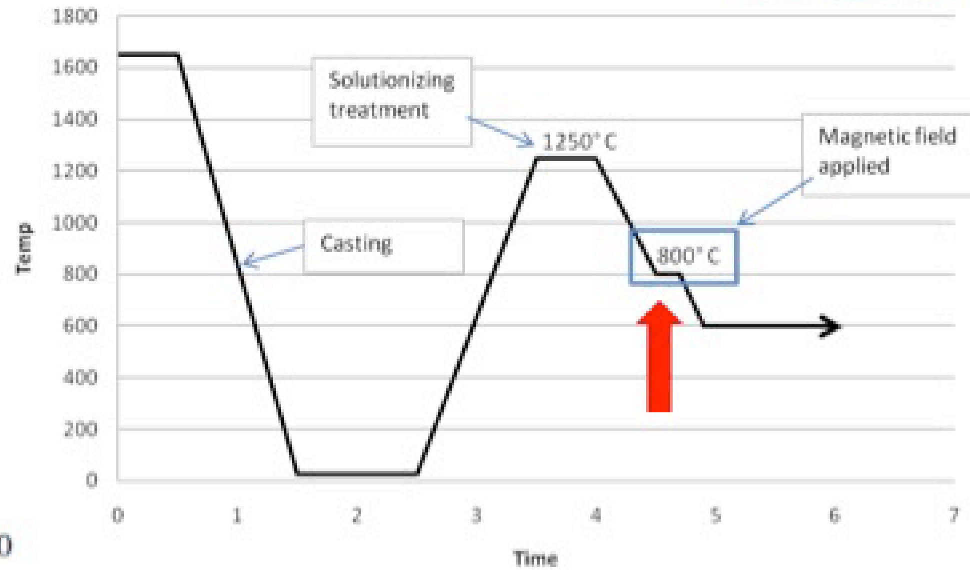
- Spinodal alloy based on AlNi-Fe(Co)
- Co additions promote higher T_{Curie}

	Al	Ni	Co	Cu	Fe
At %	15.6	12.5	21.4	2.5	48





Eleno, Luiz, Karin Frisk, and Andre Schneider. "Assessment of the Fe-Ni-Al system." *Intermetallics* 14 (2006): 1276-1290

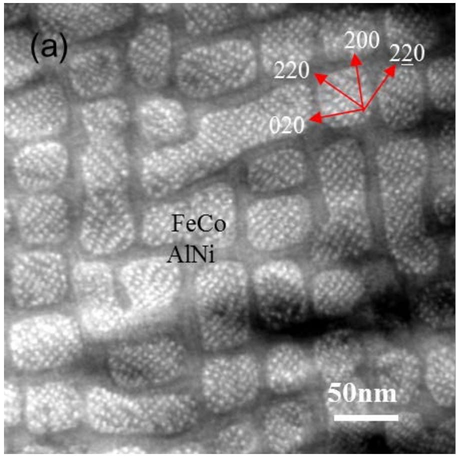


Typical thermal history of an alnico magnet

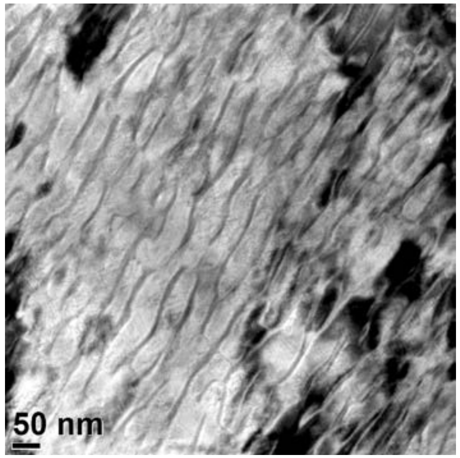
◆ Best alnico magnets (type 9) from directionally solidified fine-grained castings, but need higher remanence and additional coercivity (and mass production).

Composition Across Interface in alnico 5-7

(Kramer @ Ames with Miller @ ORNL)

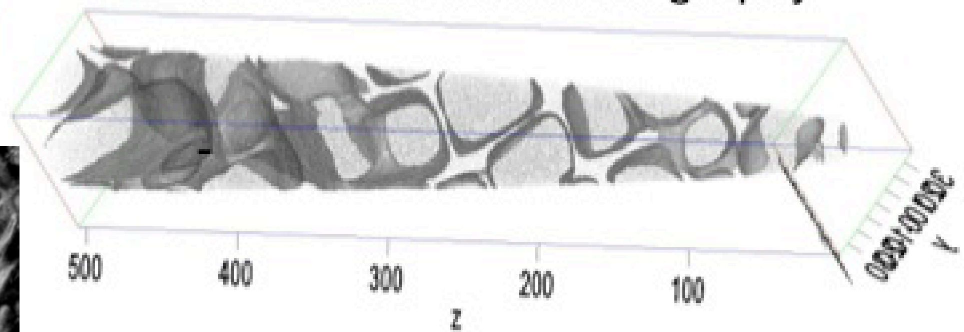


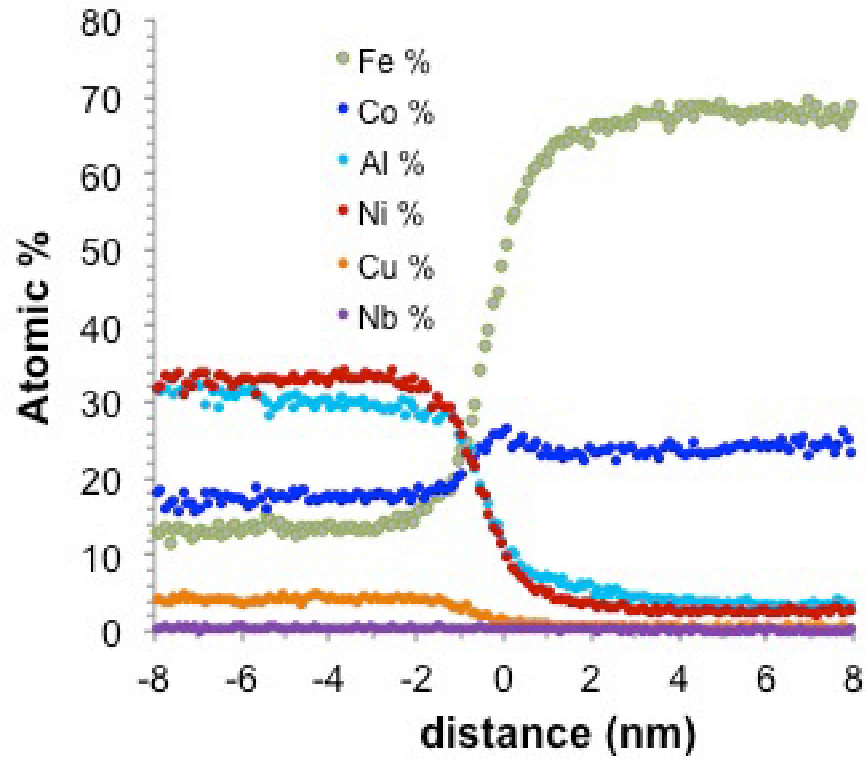
Transverse, HAADF STEM, alnico 5-7



Longitudinal, HAADF STEM, alnico 5-7

3-D Atom Probe Tomography



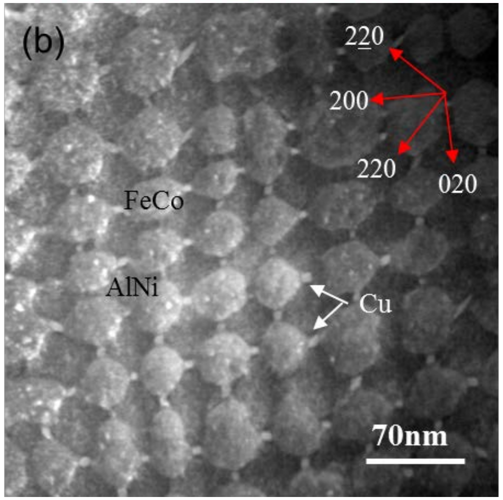


- Prismatic blocks ("bricks") of well faceted –(001), bcc-(Fe-Co) ~ 40-60 nm wide, moderate aspect ratio (< 5).
- Thin matrix ("mortar") phase, ~ 5 nm across, B2 (Ni,Al,Co,Fe), with minor Cu in solution.
- Fully coherent interfaces.
- Volume fraction of bcc:B2 ~ 61:39.

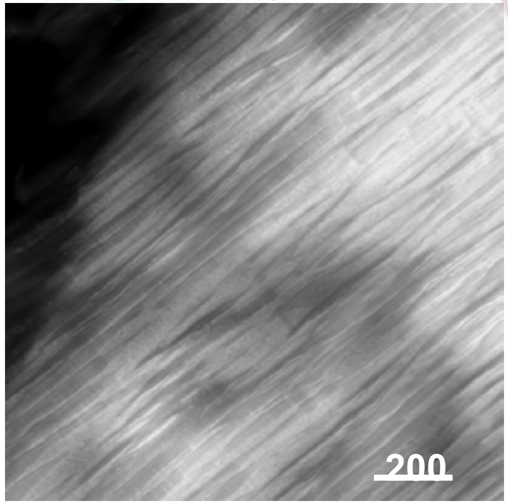
◆ Classical alnico with spinodal structure.

Composition Across Interface in alnico 8 (& 9)

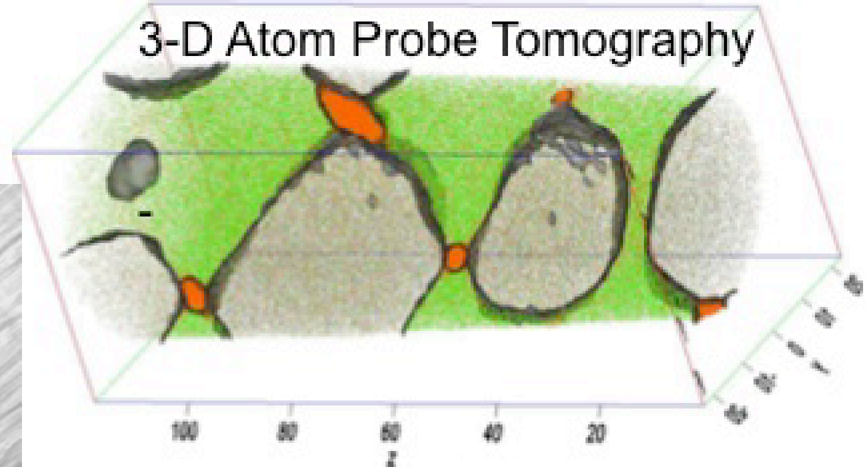
(Kramer @ Ames with Miller @ ORNL)

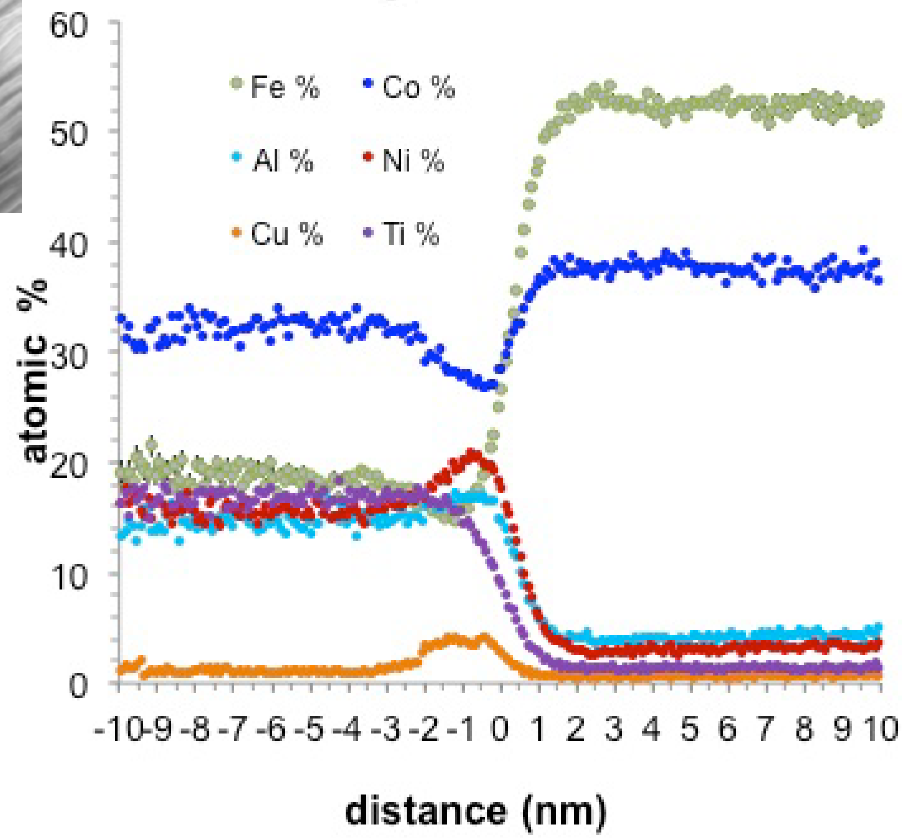


Transverse, HAADF STEM, alnico 8



Longitudinal, HAADF STEM, alnico 9

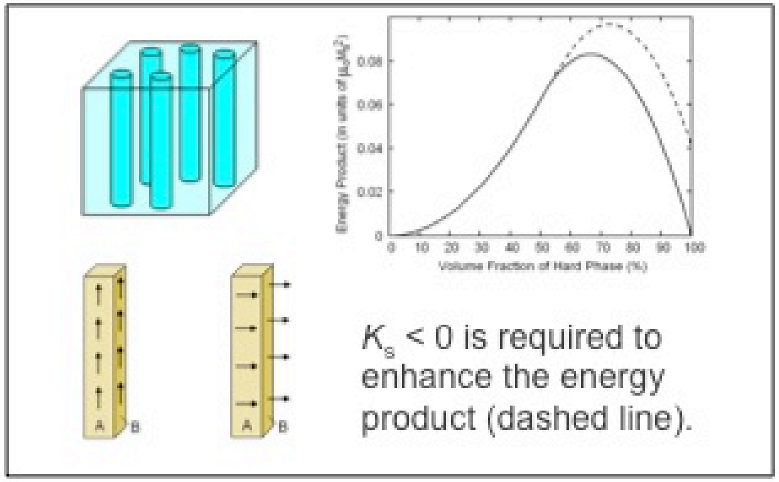




- Blocky ("mosaic tile") bcc-(Fe-Co) rather than prismatic, high (>10) aspect ratio, ends more tapered.
- Broad (near-equal, continuous) matrix phase $L2_1$, $(Ni,Co,Fe)_2AlTi$
- Cu precipitates at corners of Fe-Co tiles (& Fe-Co ppts.)
- Coherent interfaces (not Cu)
- Volume fraction of bcc:$L2_1$ ~ 50:50.

◆ First observation of "mosaic tile" alnico.

Micromagnetic Modeling (Skomski @ UNL)

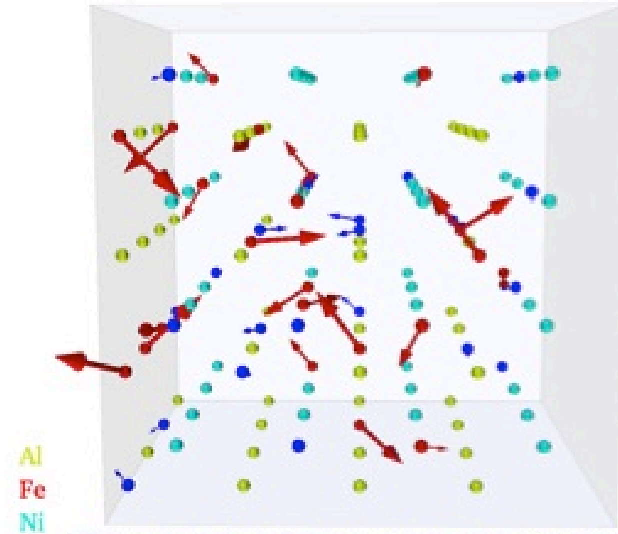


Interface anisotropy contribution to the energy product of alnico.

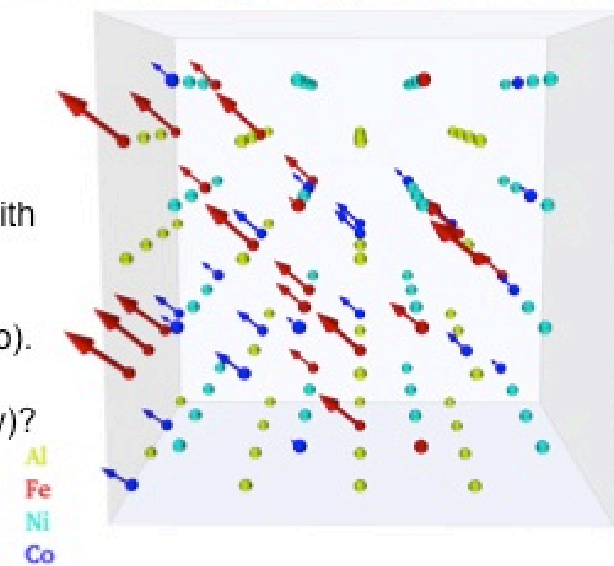
- **Indicates clear role of alnico interface anisotropy and need for high Fe-Co%.**

Moment Configurations

Initial $L2_1$ (matix) Moments



Final: Net Magnetic Moment



LSMS: Relaxation to Ground State (Stocks @ ORNL)

•Alnico 5-7: Aluminide phase (B2) is likely to be magnetically active with local moments that are ferro-magnetically coupled.

•Alnico 8 (&9): Aluminide phase ($L2_1$) more (?) magnetic (more Fe, Co).

•Effect of this on magneto-crystalline and shape anisotropy (coercivity)?

•Need to mitigate intermixing of Co and Fe into aluminide phase?

◆ Excess Co and Fe in AlNi phase may degrade coercivity severely.

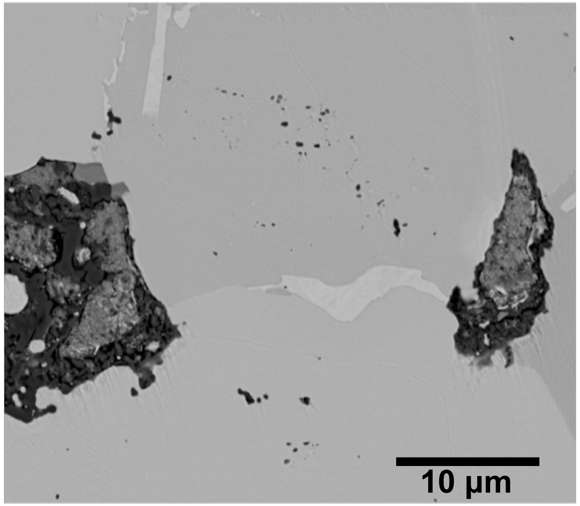
Known Properties and New Results Indicate Powder Option

PERMANENT MAGNET MATERIALS -- Ref: MMPA STANDARD 0100-00

IEC Code	Class	Al wt%	Ni wt%	Co wt%	Cu wt%	Ti wt%	BHmax MGOe	Br Gauss	Hc Oersted	Hci Oersted
R1-0-2	Cast Alnico 3	12	25	0	3	0	1.35	7000	480	500
R1-1-1	Cast Alnico 5	8	14	24	3	0	5.50	12800	640	640
R1-1-2	Cast Alnico 5DG	8	14	24	3	0	6.50	13300	670	670
R1-1-3	Cast Alnico 5-7	8	14	24	3	0	7.50	13500	740	740
R1-1-4	Cast Alnico 6	8	16	24	3	1	3.90	10500	780	800
R1-1-5	Cast Alnico 8	7	15	35	4	5	5.30	8200	1650	1860
R1-1-7	Cast Alnico 8H	8	14	38	3	8	5.00	7200	1900	2170
R1-1-6	Cast Alnico 9	7	15	35	4	5	9.00	10600	1500	1500
R1-0-4	Sintered Alnico 2	10	19	13	3	0	1.50	7100	550	570
R1-1-10	Sintered Alnico 5	8	14	24	3	0	3.90	10900	620	630
R1-1-11	Sintered Alnico 6	8	15	24	3	1	2.90	9400	790	820
R1-1-12	Sintered Alnico 8	7	15	35	4	5	4.00	7400	1500	1690
R1-1-13	Sintered Alnico 8H	7	14	38	3	8	4.50	6700	1800	2020

Isotropic (unoriented) grades: Alnico 2, 3, 4

Anisotropic (oriented) grades: Alnico 5, 6, 8, 9

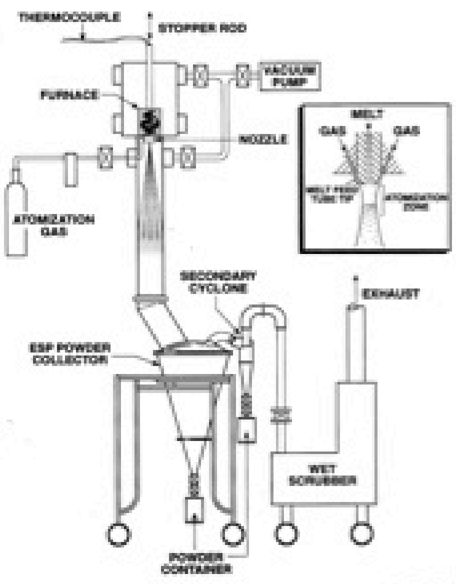


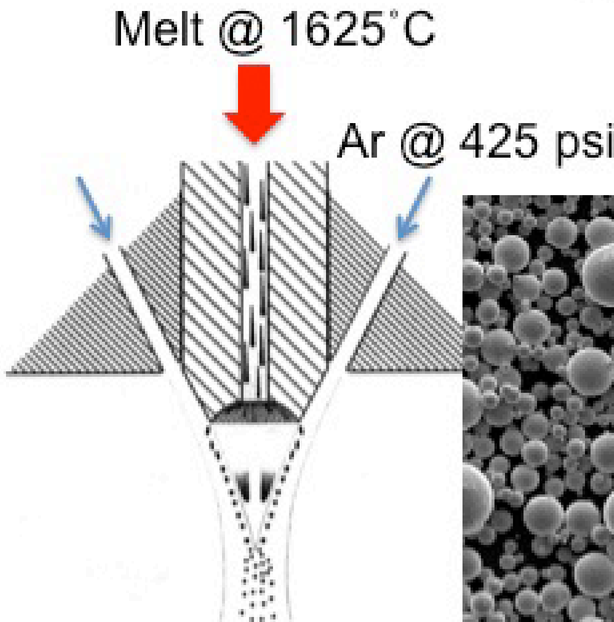
FE-SEM of sintered alnico 8 with large inclusions of impurity (Ti, Al, O, C) phases.

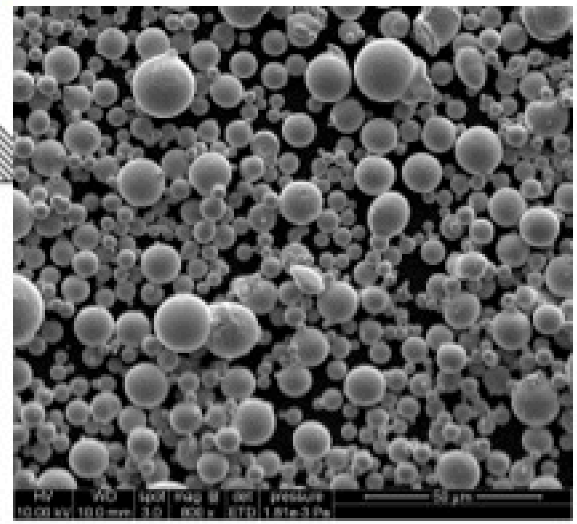
- Gas atomization of **pre-alloyed** alnico 8H was attempted to generate isotropic fine grains (micro-pinning from mis-orientation) with non-continuous spinodal pattern (nano-pinning) and greatly reduced vol.% impurities (suppressed domain nucleation).

- Potential for increased coercivity and saturation with high volume manufacturing capability.

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







Dia.<20μm screened powder

Melt stream before spray onset.

Powder Yield: Avg. particle dia.= 30 μm

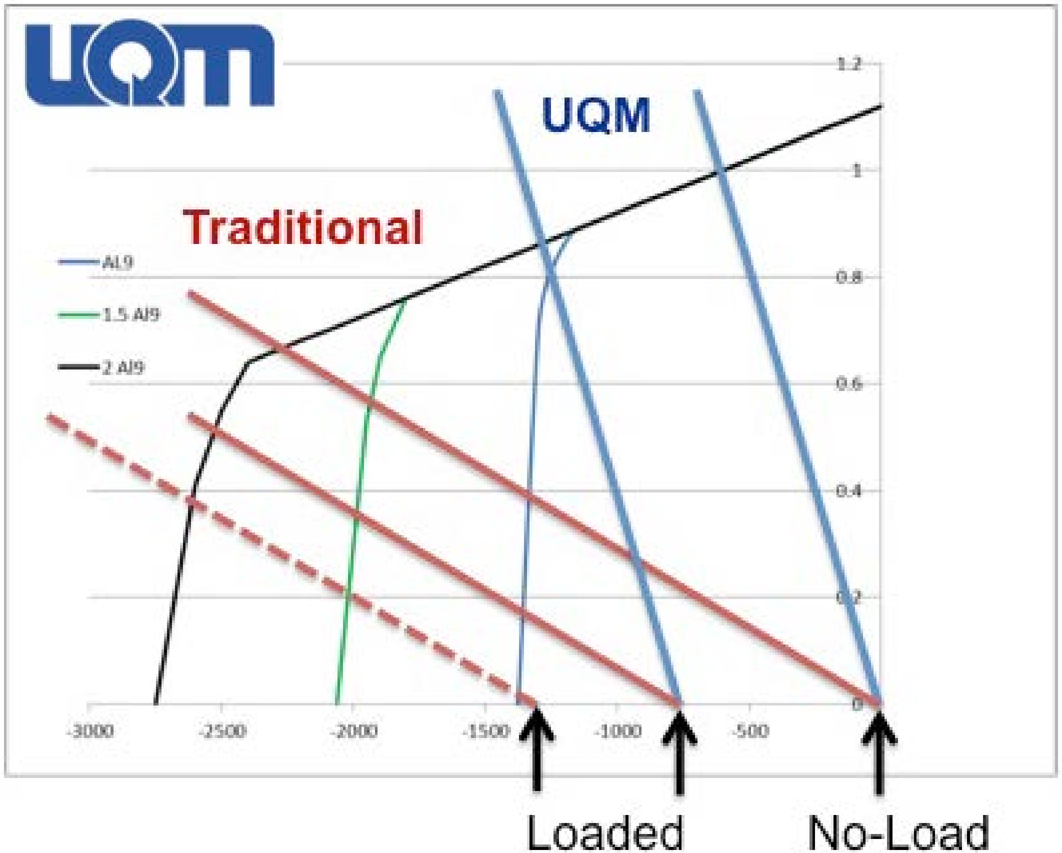
Aim alloy: 32.3Fe-38.0Co-13.0Ni-7.3Al-6.4Ti-3.0Cu (wt.%) ≈ alnico 8H
Analyzed: 32.4Fe-38.1Co-12.9Ni-7.3Al-6.4Ti-3.0Cu (45-75μm powder sample)

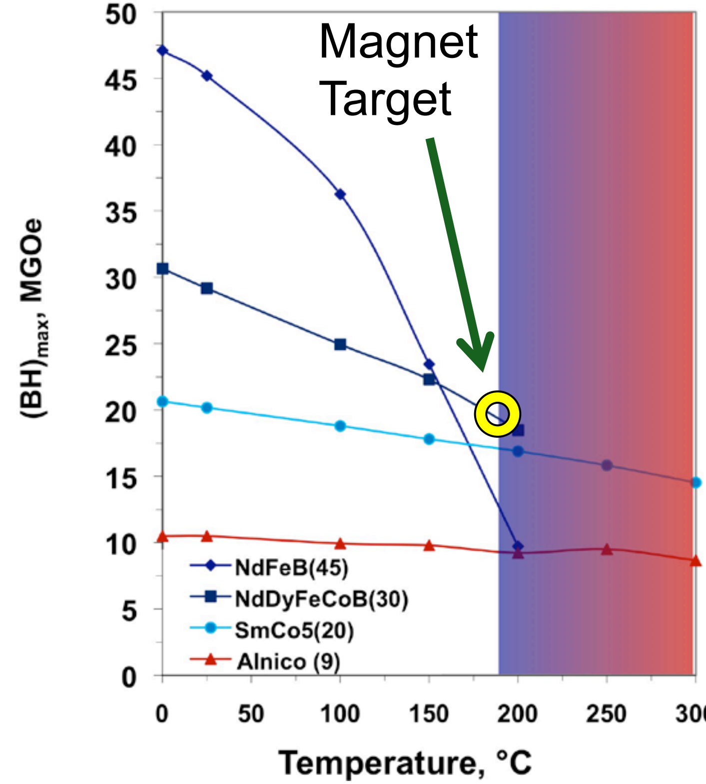
Interstitial impurities (ppmw): C=66, N=<10, O=420, S=30 (<20μm powder sample)

First attempt shows promise for new approach.

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

Outlook for Enhanced Alnico in Advanced PM Motor

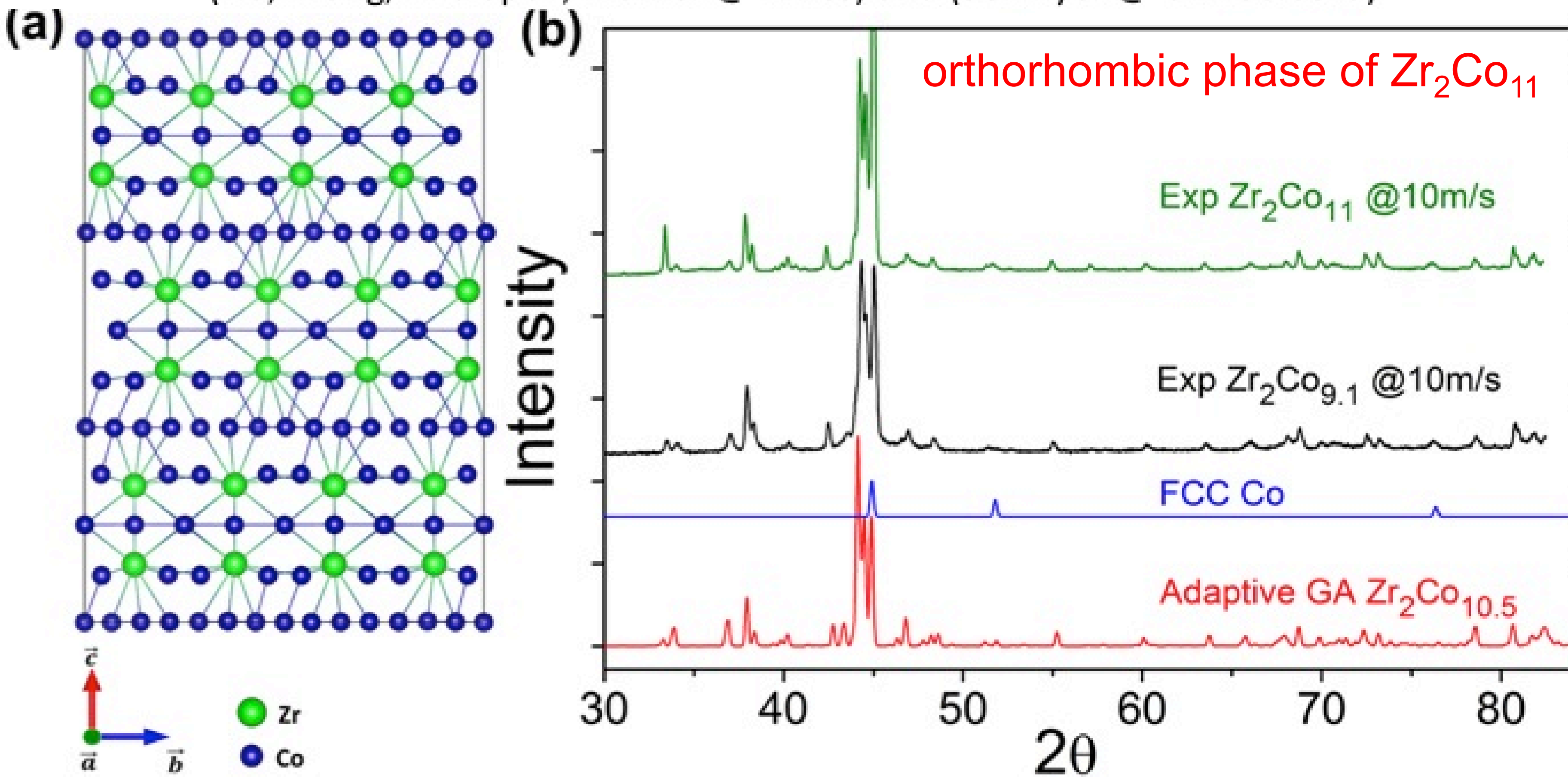




- "Operating load line of the UQM (motor) innovation is substantially higher than existing permanent magnet motors, allowing the use of lower coercivity magnets that support high flux densities." (e.g., alnico 9+)
- Possibility of improvement of alnico 8 (or 8/9) to useful range by powder processing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

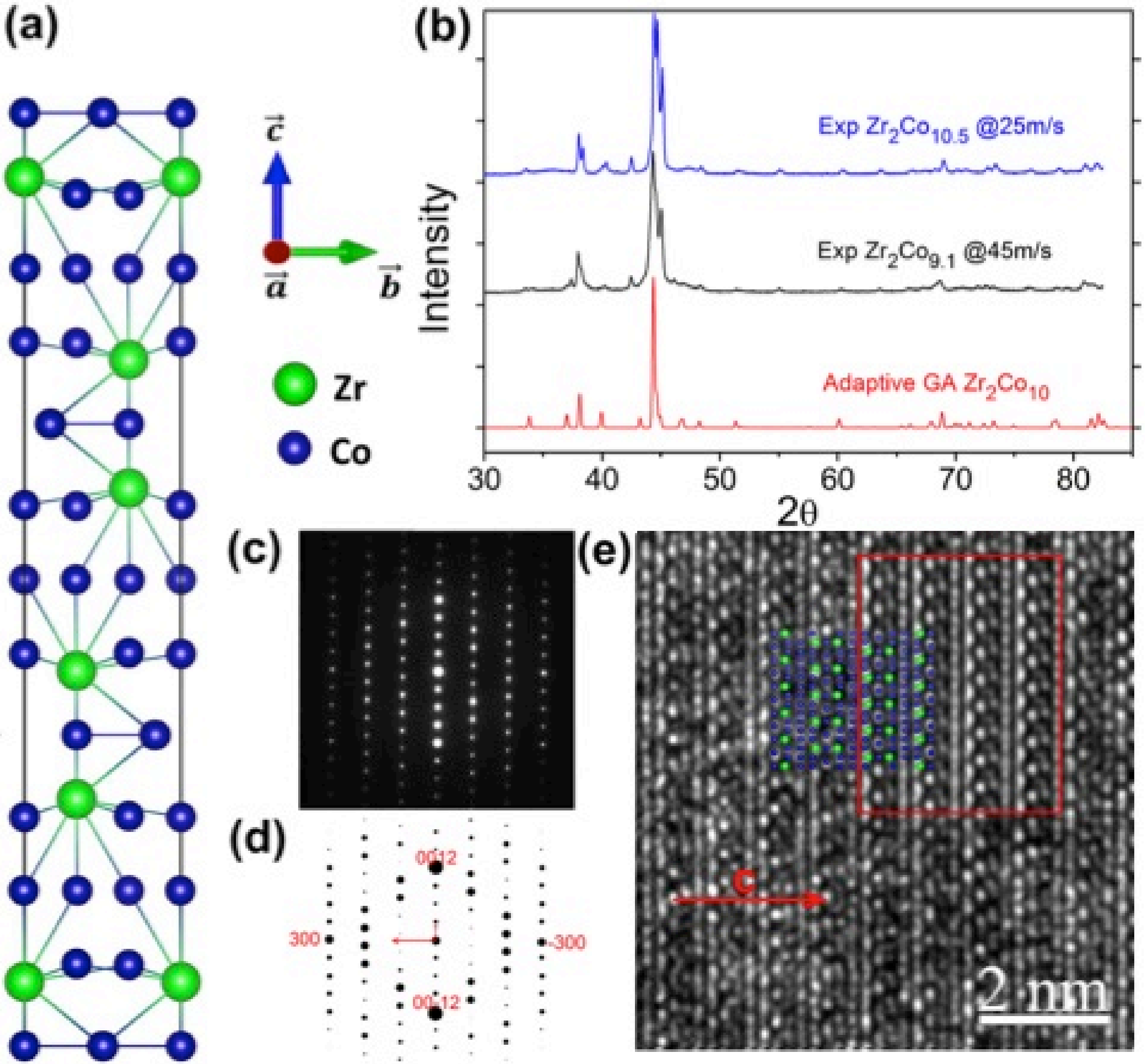
Long-term Co(Fe)-X candidate: Unraveling the structural mystery of Zr_2Co_{11} phases

(Ho, Wang, Antropov, Kramer @ Ames) and (Sellmyer @ U. Nebraska)



The unit cell contains 150 atoms. The simulated XRD spectrum and TEM diffraction patterns from the structure model predicted by the GA search agreed well with those from experiment.

Structure of the rhombohedral phase of Zr_2Co_{11} from GA search



(a) Rhombohedral model

(b) Simulated XRD in comparison with Experiment

(c) Measured SAED pattern along [010] direction

(d) Simulated SAED pattern along [010] from our model

(e) HREM image along the [010] zone axis. Inset, within the **red box** is the simulated image from our model.

❑ Magnetic properties calculation

Structure	Moment (μB/Co atom)	K (μeV/atom)	K (MJ/m3)	T_c (K)
*ZrCo5.25	1.05 [1.07]	50 [42]	0.64 [0.54]	950
ZrCo5 (Ni5Zr type)	1.09[1.12]	~ 0	~ 0	1063
Rhombohedral (R32)	0.92[1.01]	81 [111]	**1.04 [1.42]**	709
Hexagonal (P-62c)	0.94[1.01]	103 [104]	**1.32 [1.33]**	688

*Not uniaxial. The calculation is done on the structure with unit cell containing 50 atoms
[ ] GGA results; **K**: magnetic anisotropy; **T_c**: Curie Temperature

❑ Results

- Predicted unit cells of the Rhombohedral and Hexagonal phases are within experimental error---both phases can form.
- The magnetic moment of Co atoms in Zr_2Co_{11} is about $0.9\mu_{B;}$
- K ~ 1.3 MJ/m^3; significant magnetic anisotropy over (hex.) Co
- T_C ~ 700 °C; should have operating temperature limit >> 200˚C

Ames Lab Collaborations and Partnerships, 2013

Collaborators:

- 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)
- 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.

BREM Project (VT) Partners (subcontractors):

- ORNL (Theory), Univ. Maryland (Synthesis, Characterization), Univ. Nebraska (Theory, Synthesis, Characterization), Brown Univ. (Synthesis), Arnold Magnetic Technologies (Synthesis).

UQM TECHNOLOGIES

imagination at work

ARNOLD® MAGNETIC TECHNOLOGIES

Summary of FY12-13 Accomplishments

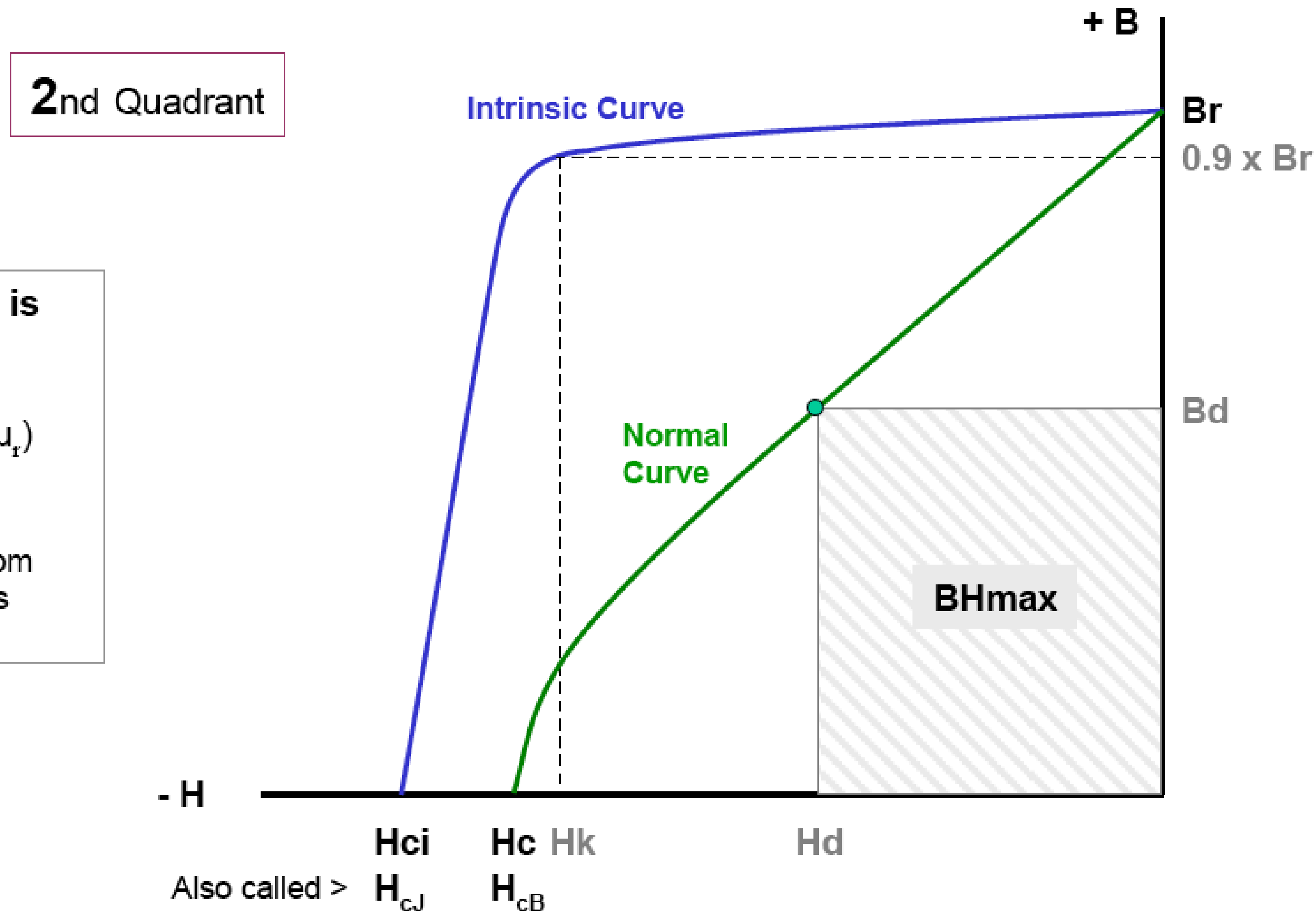
- Developed new single stage hot deformation (SSHD) method to fabricate fully dense anisotropic magnets from Zn-coated glassy $MRE_2(Fe,Co)_{14}B$ flake with reduced Dy.
- Completed initial analysis of commercial alnico types 5-7, 8, and 9.
  - Started investigation of alloy influences on completion of spinodal partitioning.
  - Began study of ultimate reduction of spinodal decomposition scale.
  - Characterized details of complex matrix phase in alnico 8 & 9 and differences with alnico 5-7 in Fe-Co phase pattern and shape anisotropy.
  - Analyzed grain boundaries and product phases in sintered (powder) alnico 8.
- Started modified heat treating of commercial alnico to generate variations in nano-structure (spacing, solute partitioning, and continuity)
- Performed additional theoretical analysis of alnico interface effects on anisotropy energy and direction (impacts on coercivity) and volume% effects on remanence.
- Produced first pre-alloyed alnico 8H gas atomized powder, consolidated it, and magnetically annealed samples for testing on hysteresis-graph.
- Used theoretical tools to expand investigation of potential new Fe-Co-X and Fe-Co-Z phases on clusters and supercomputers.
- Extended experimental investigations of Co-X, Fe-X, & Fe-Co-X systems (X=5d and 4d, e.g., Zr, W, Ta, Mo, Hf) with combinatorial synthesis, cluster deposition (with alignment field), and chemical synthesis
- Maintained WebEx® sharing of results and held Spring & Fall BREM workshops.

Outline of FY13 Plans

- Further develop SSHD method to fabricate fully dense anisotropic magnets from Zn-coated glassy $MRE_2(Fe,Co)_{14}B$ flake (blended with Al) to lower Dy content and verify high temperature stability.
- Provide detailed characterization results to allow improved selection of alnico compositions and development of bulk magnet processing approaches, emphasizing new powder metallurgy approach.
- Extend exploration with theoretical tools to investigate possible alnico interface/phase configurations and potential new Fe-Co-X phases on existing clusters and supercomputers (access from new INCITE award).
- Based on theoretical guidance, pursue promising regions of ternary Fe-Co-X systems for further investigation by full suite of experimental synthesis methods.
- Maintain WebEx contact and bi-annual workshops with team.

Technical Back-Up Slides

Permanent Magnet Key Characteristics



Energy product is related to Br

BHmax ~ $Br^2 / (4 \cdot \mu_r)$
$\mu_r \sim 1.05$

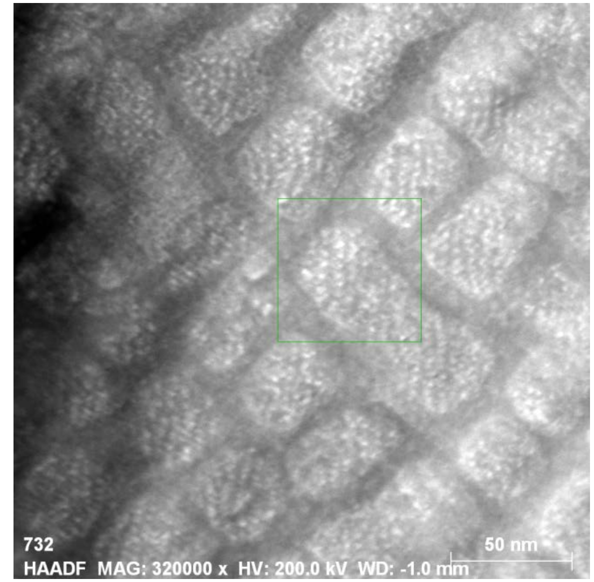
When Normal curve from Br to Operating Point is Linear

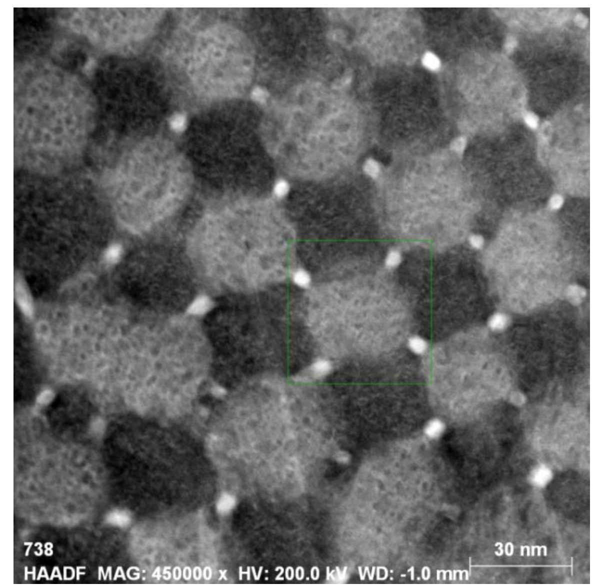
Alnico Theoretical Limits

		Alnico 5-7	Alnico 8	Alnico 9
aspect ratio		~ 5:1	~ 10:1	> 10:1
fraction bcc phase (f)		0.62	0.4	0.53
Fe:Co in bcc phase		0.74	0.58	0.60
mole % Fe+Co in bcc		0.92	0.90	0.91
~M_s (KG) for bcc based on Fe:Co		23.8	23.9	23.9
Fe:Co in intermetallic		0.44	0.37	0.27
mole % Fe+Co in bcc		0.31	0.51	0.40
B_r (KG)	measured	13.5	8.2	10.6
	calculated	13.6	8.6	11.5
Hc_i (Oe)	measured	740	1860	1500
	calculated	3105	4365	3715
BH_{max} (MGOe)	measured	7.5	5.3	9.0
	calculated	21.1	18.8	21.4

Spinodal Phases in Alnico

	Fe-Co			'Al-Ni'		
	bcc phase (at. %)			intermetallic phase (at. %)		
	5-7	8	9	5-7 B2	8 – $L2_1$	9 - $L2_1$
Fe	68.1	52.3	54.4	13.4	18.8	10.8
Co	24.2	37.6	36.5	17.4	32.3	28.7
Ni	2.6	3.2	3.5	33.0	15.8	20.6
Al	3.6	4.3	4.0	30.6	14.6	24.4
Cu	0.5	0.7	0.4	4.2	1.1	1.5
Nb	0.1			0.5		
Ti		1.4	0.5	0.3	16.8	12.9
Cr			0.1			0.9
Si	0.5	0.2			0.4	





Long-term Co(Fe)-X candidate: Unraveling the structural mystery of Zr_2Co_{11} phases

(Ho, Wang, Antropov, Kramer @ Ames) and (Sellmyer @ U. Nebraska)

- For 30 years, attention was focused on Zr_2Co_{11} as potential non-RE magnet material.
- However, atomic structure of this compound has not been resolved.
- Atomic structure needed for compound substitution to enhance magnetic properties.

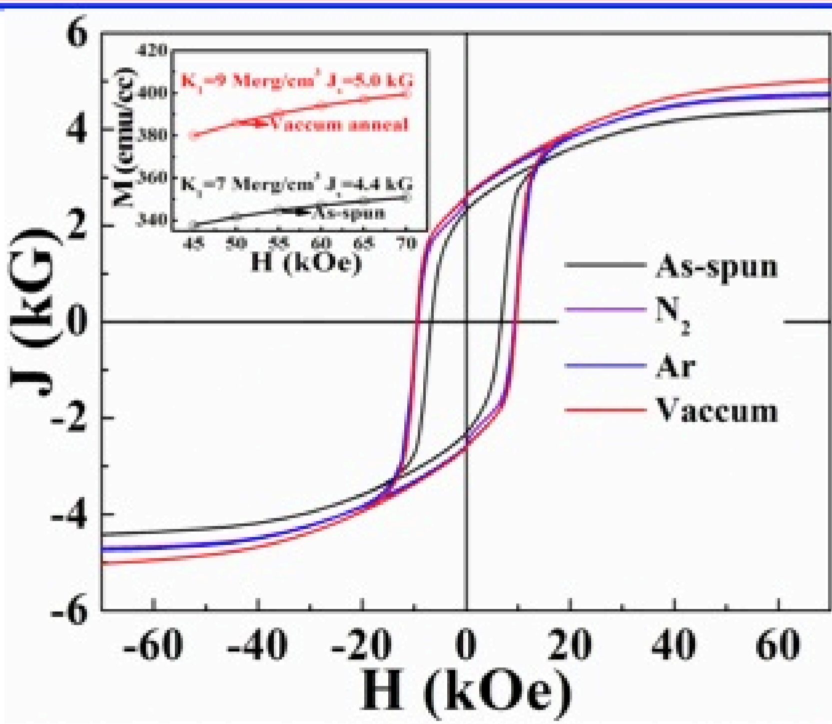
References	Proposed crystal structure
Demczyk et al. *J. Appl. Cryst.* **24**, 1023 (1991).	Orthorhombic (a=4.8Å, b=8.2Å, c=36.0Å)
Gabay et al. *J. Magn. Magn. Mater.* **236**, 37(2001).	FCC (a=6.7Å)
1. Ivanova et al. *J. Alloys Compd.* **432**, 135(2007);	Rhombohedral (a=4.76Å, c=24.2Å) **High-T** phase
2. Ivanova et al. *The Physics of Metals and Metallography*, **107**, 270 (2009);	Hexagonal (a=4.76Å, c=16.1Å)
3. Zhang et al. J. Phys. D: Appl. Phys. (in press)	Orthorhombic (a=4.71Å, b=16.7Å, c=24.2Å) **Low-T** phase

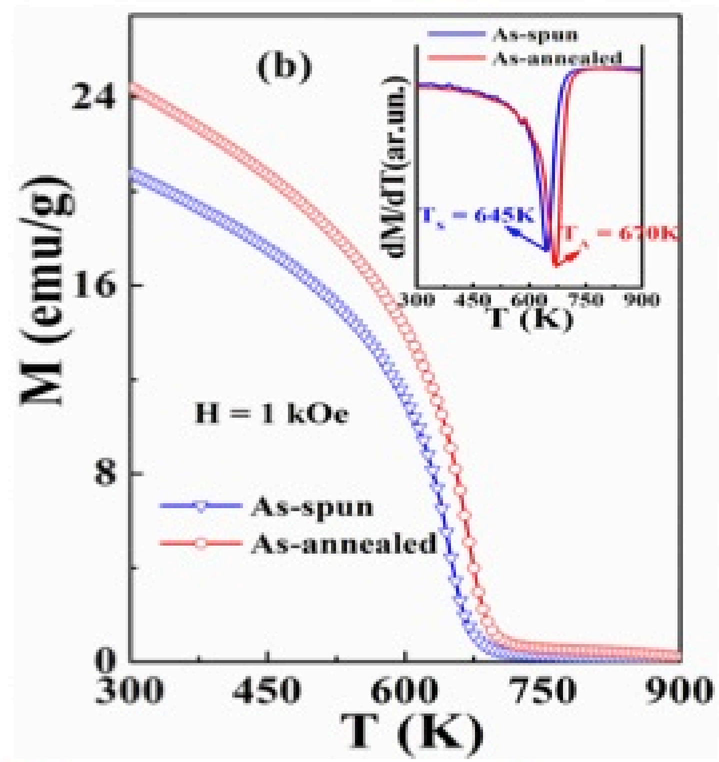
- **Difficulties** determining the crystal structure of Zr_2Co_{11}:

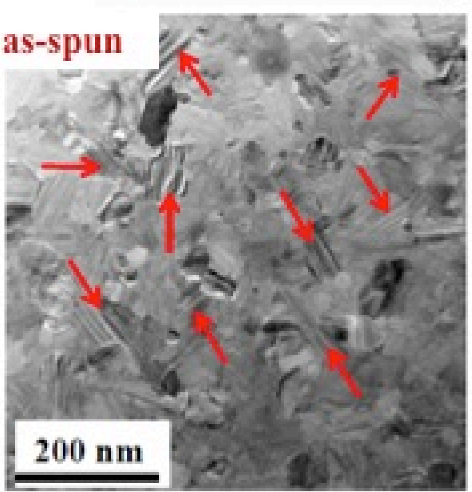
Small grain size samples; Multiple phases-some at high temp.; Composition uncertainty.

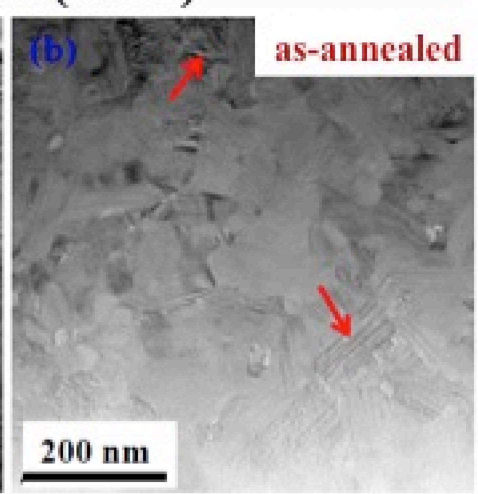
Annealing Effect on Magnetic Properties

$Zr_{16}Co_{78-x}Mo_xSi_3B_3$ (x = 4) spun at 16 m/s (Sellmyer, Shield @ UNL)









√ Annealing increases $_iH_c/T_c$ and slightly enhances J_s.

√ Annealing coarsens the grain size and decreases the number of the stripe defects indicated by red arrows.