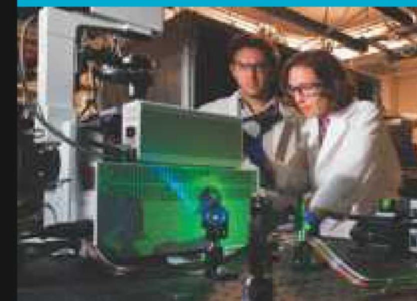
SAND2018-2772C

Synthesis of γ'-Fe_4N, a new soft magnetic material for inductors and transformers



Tyler E. Stevens, Charles J. Pearce, Mark A. Rodriguez, Sara Dickens, Bonnie B. McKenzie, Stanley Atcitty, & Todd C. Monson

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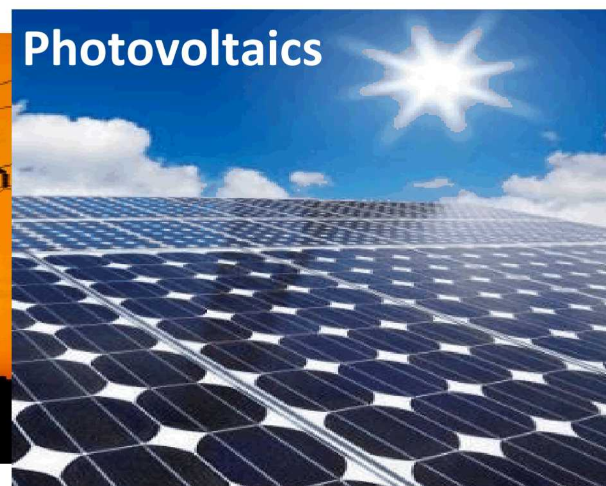
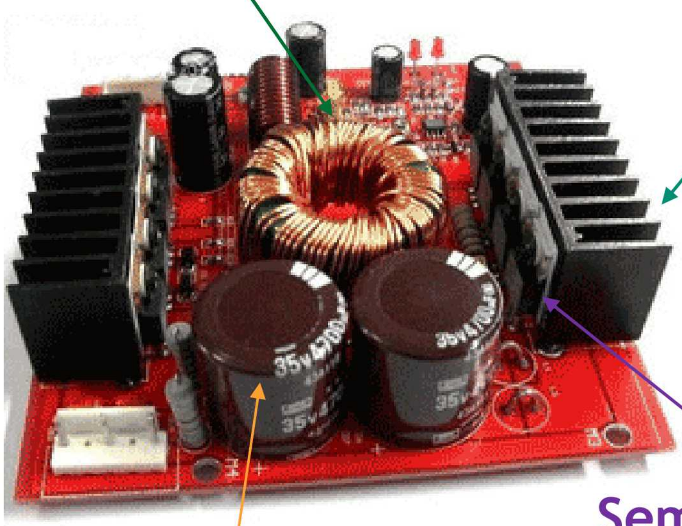
Sandia Collaboration to Develop New Power Electronics



Courtesy of Bob Kaplar

Magnetic Devices Impact Power System Volume and Weight

Power Electronics Components

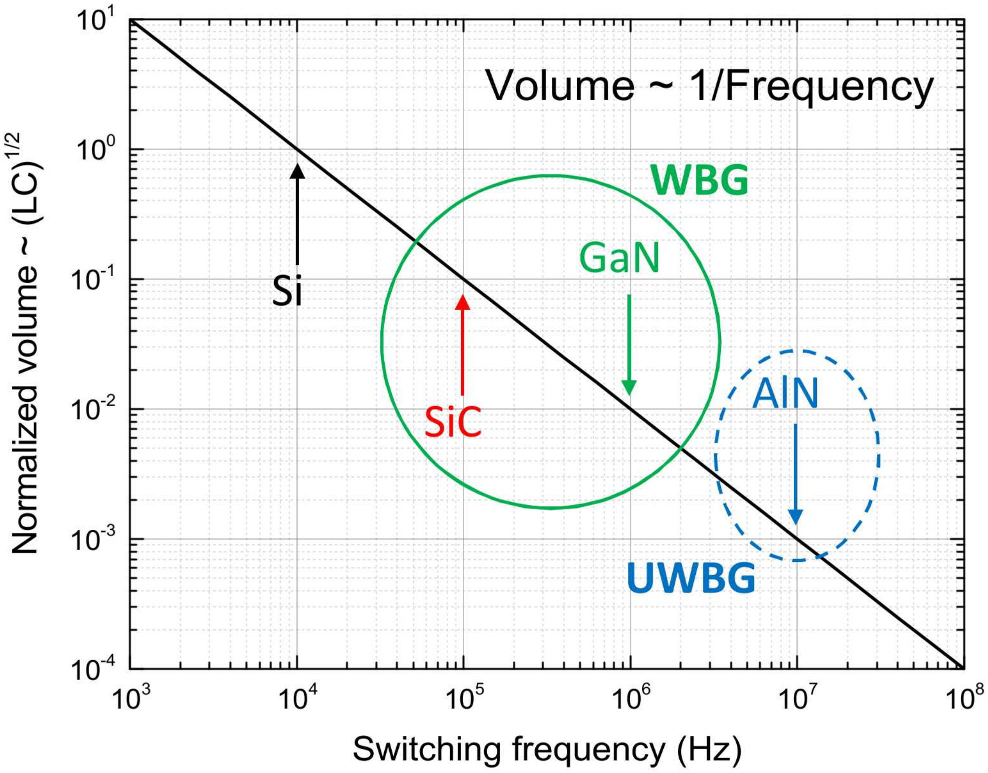


WBG/UWBG materials enable higher switching frequency and better thermal management

J. Neely, J. Flicker, B. Kaplar

- Passive elements and thermal management comprise the bulk of the volume and mass of a power converter

Higher Frequencies Decrease Inductance Requirements, However…

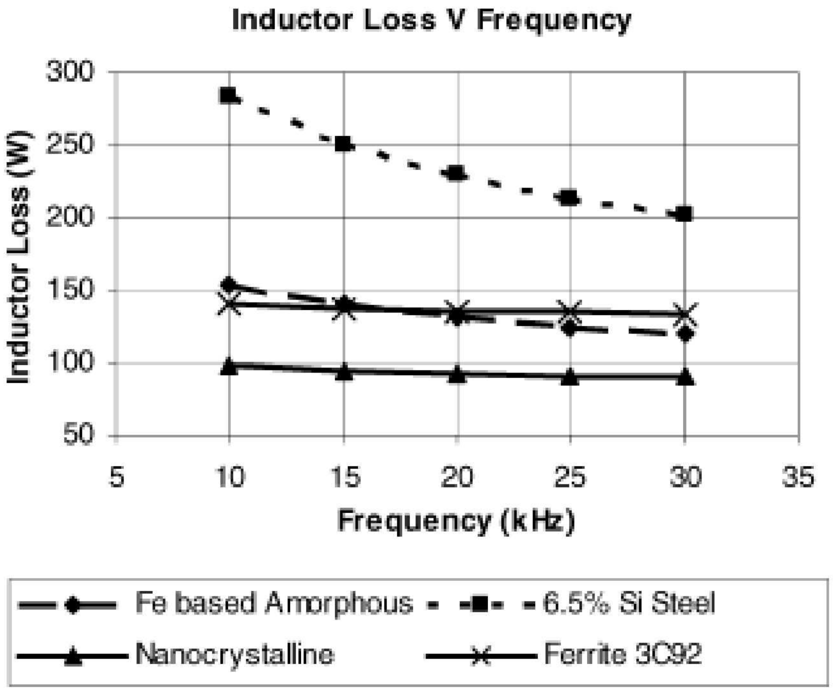


- **Higher switching frequency is enabled by scaling properties of WBG/UWBG materials**
- **Ideal 1/f SWaP dependence (true dependence likely weaker $1/f^n$ with $n < 1$ due to nonidealities)**
- **Other benefits of WBG/UWBG exist, e.g. higher voltage without series stacking of devices, and higher temperature operation**

Inductive core materials have essentially been an afterthought and new magnetic materials are needed

Adapted from Bob Kaplar

Inductor Loss Increases with Frequency

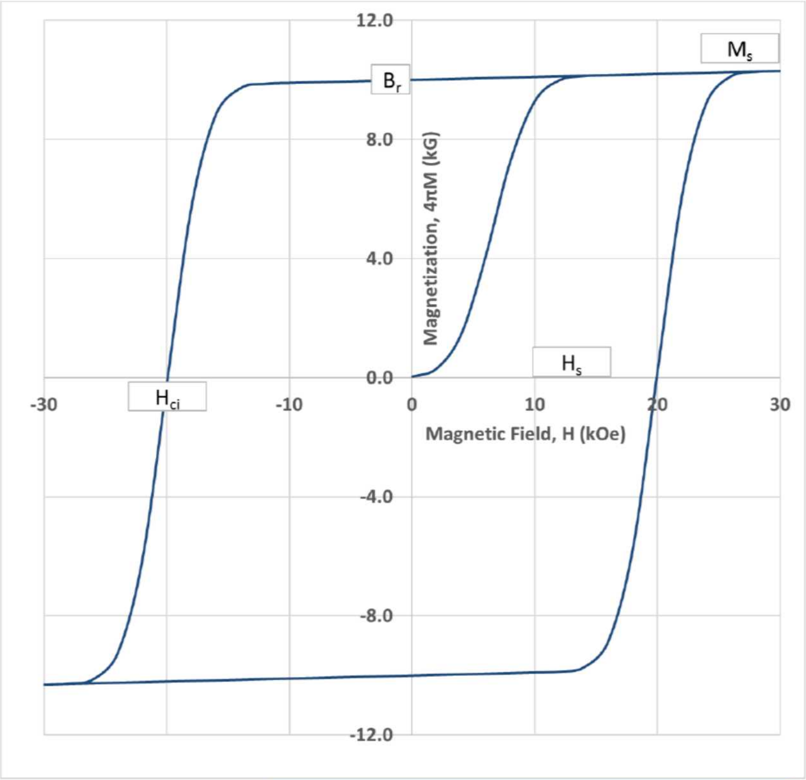


B.J. Lyons, J.G. Hayes, M.G. Egan, Magnetic Material Comparisons for High-Current Inductors in Low-Medium Frequency DC-DC Converters, *IEEE*, **2007**, 71.

<u>Requirements for a new magnetic material</u>

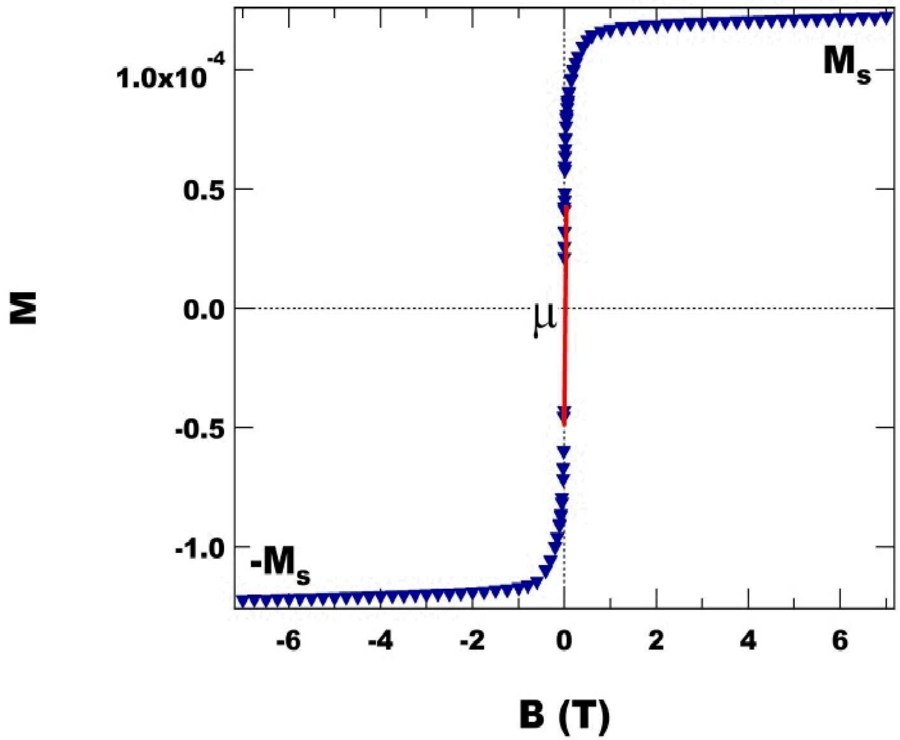
- Low loss in 10-200 kHz frequency range
- High permeability (low coercivity) and saturation magnetizations
- Low magnetostriction
- high temperature performance and scalable & affordable.

Hard (permanent)magnet



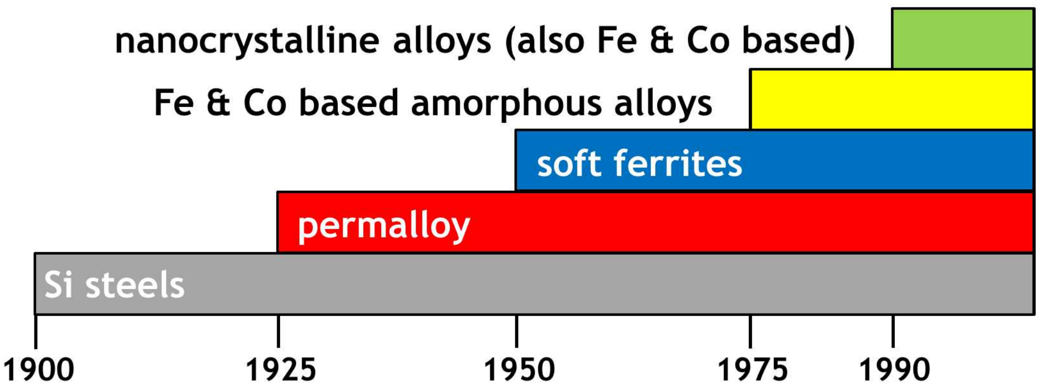
Source: Spontaneous Materials

Soft magnet

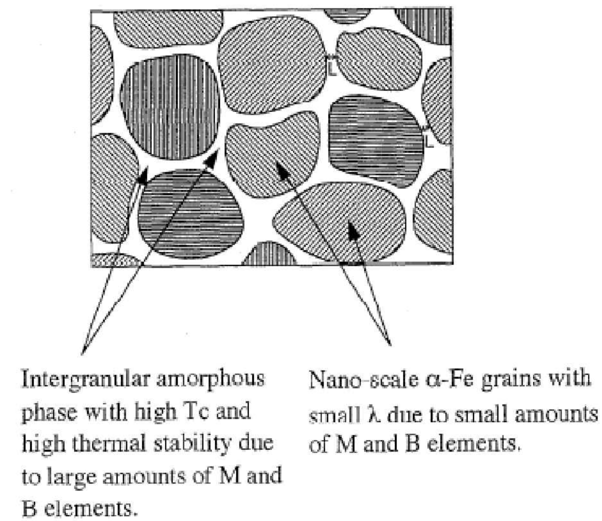


M_s = saturation magnetization, B_r = magnetic remanence
H_c = coercivity, μ = permeability

Development of Soft Magnetic Materials

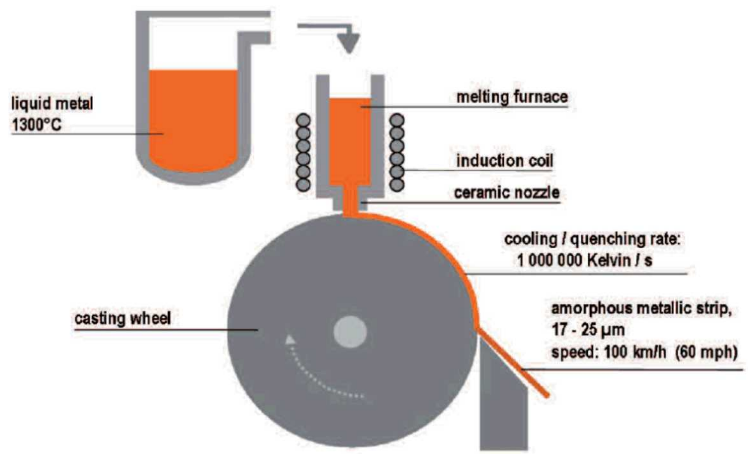


Adapted from: L.A. Dobrzański, M. Drak, B. Ziębowicz, Materials with specific magnetic properties, Journal of Achievements in Materials and Manufacturing Eng., 17, 37 (2006).



"NANOPERM"

A. Makino, et. al., Nanocrystalline Soft Magnetic Fe-M-B (M = Zr, Hf, Nb) alloys and their applications, Mat. Sci, and Eng., A226-228, 594 (1997).



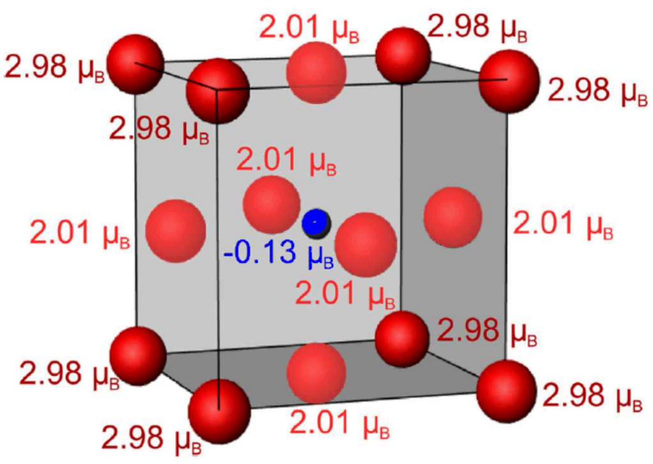
VITROPERM (Vacuumschmelze)

- **Complex stoichiometry including Fe, Co, and other inactive elements such as B, Zr, Hf, Nb, Cu, Mo, Si, C**
- **Time consuming and high temperature processing → costly!**
- **Substantial inactive material to form a low loss nanocrystalline structure**
- **Material produced in tapes and often combined with plastic laminations**

γ'-Fe_4N meets all of the requirements for a new soft magnetic material

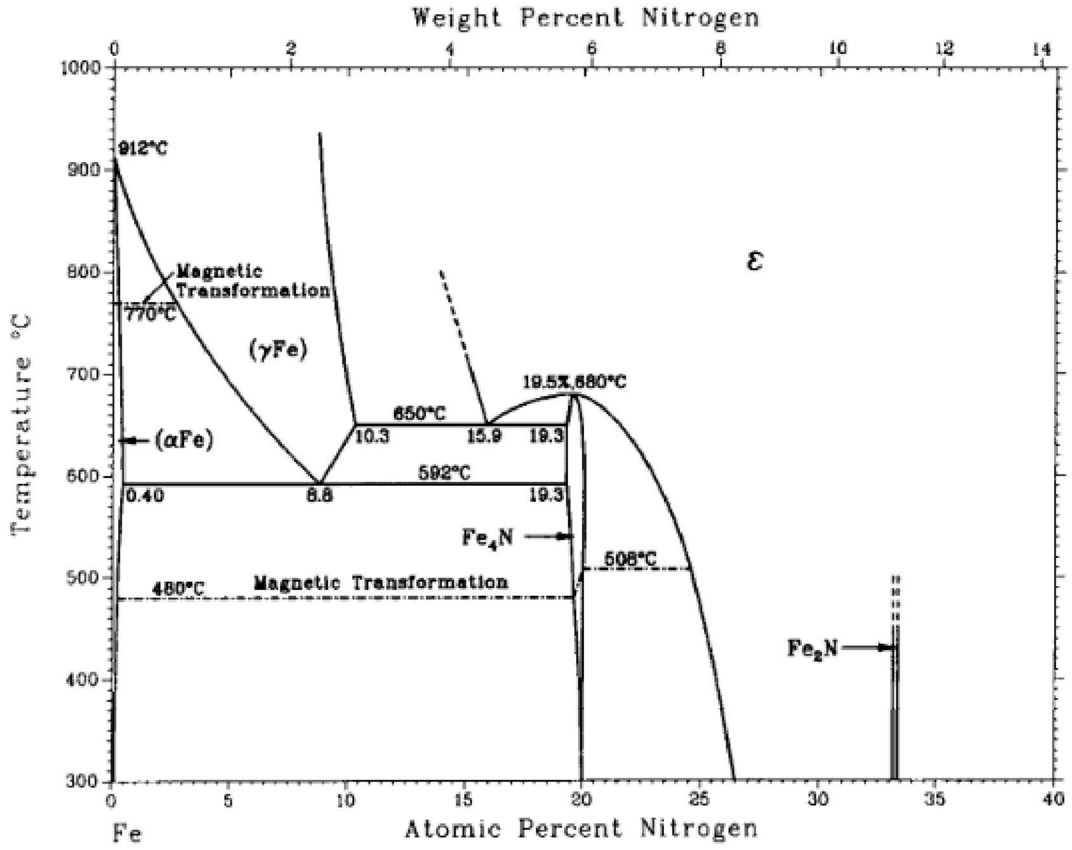
Magnetic Material	J_s (T)	ρ($\mu\Omega\cdot$m)	Cost
VITROPERM (Vacuumschmelze)	1.20	1.15	High
Metglas 2605SC	1.60	1.37	High
Ferrite (Ferroxcube)	0.52	$5x10^6$	Low
Si steel	1.87	0.05	Low
γ'-Fe_4N	1.89	> 200	Low

fcc γFe structure stabilized by interstitial nitrogen in the body center



G. Scheunert, et al., A review of high magnetic moment thin films for microscale and nanotechnology
Applications, Appl. Phys. Rev., 3, 011301 (2016).
J.M.D. Coey, *Magnetism and Magnetic Materials* (Cambridge University Press, Cambridge, UK, 2012).

Synthesis of γ'-Fe_4N is Challenging

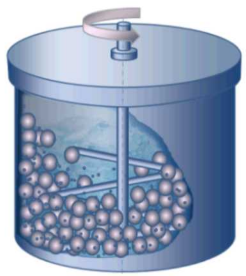


H.A. Wriedt, N.A. Gokcen, and R.H. Nafziger, 1987.

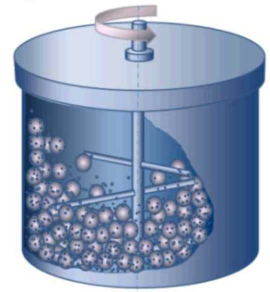
γ'-Fe_4N Synthesis and Processing

U.S. Patent Filed January 2015 (#62/105,918)

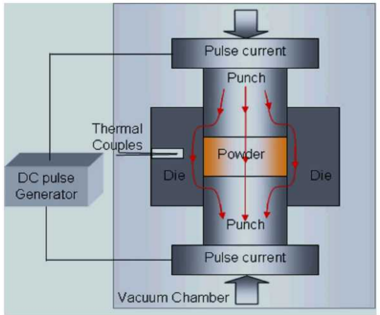
Synthesis of dense nanocrystalline iron nitrides using a two-step reactive milling and high pressure spark plasma sintering (SPS).



Liquid N_2



NH_3

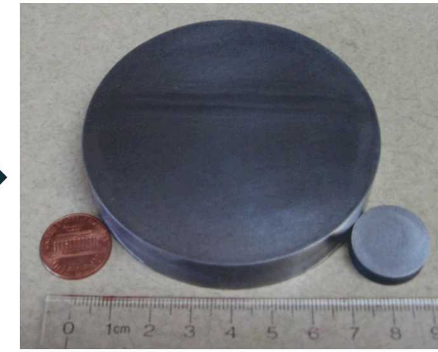


Spark Plasma Sintering (SPS)

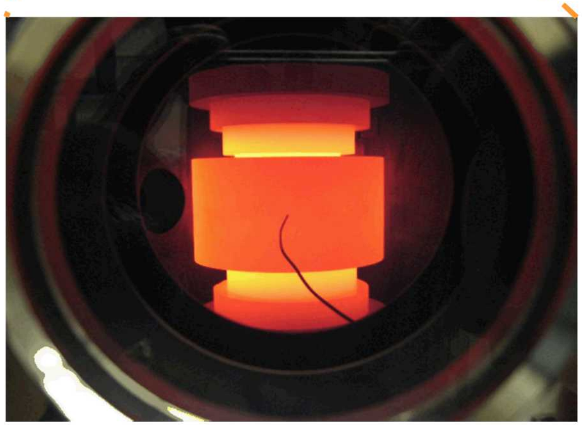
- **Cryomilling creates nanocrystalline Fe powder with large amounts of vacancies, grain boundaries, and dislocations**
- **Defects serve as fast diffusion pathways for nitrogen atoms from NH_3**
- **SPS quickly consolidates raw powders with a low sintering temperature**
  - **Excellent control over grain growth**
  - **Result: Improved magnetic properties**

Spark Plasma Sintering (SPS)

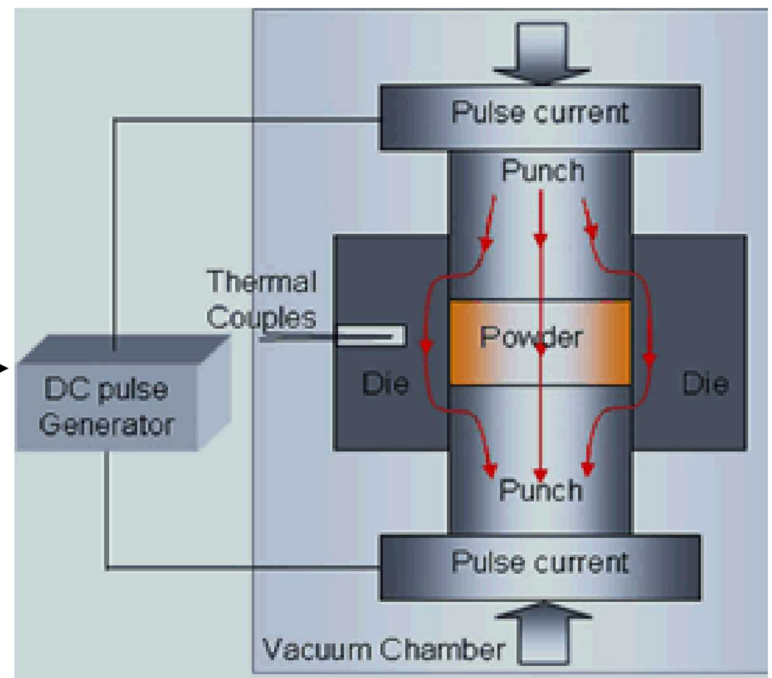
Starting Powder in Die



End Product

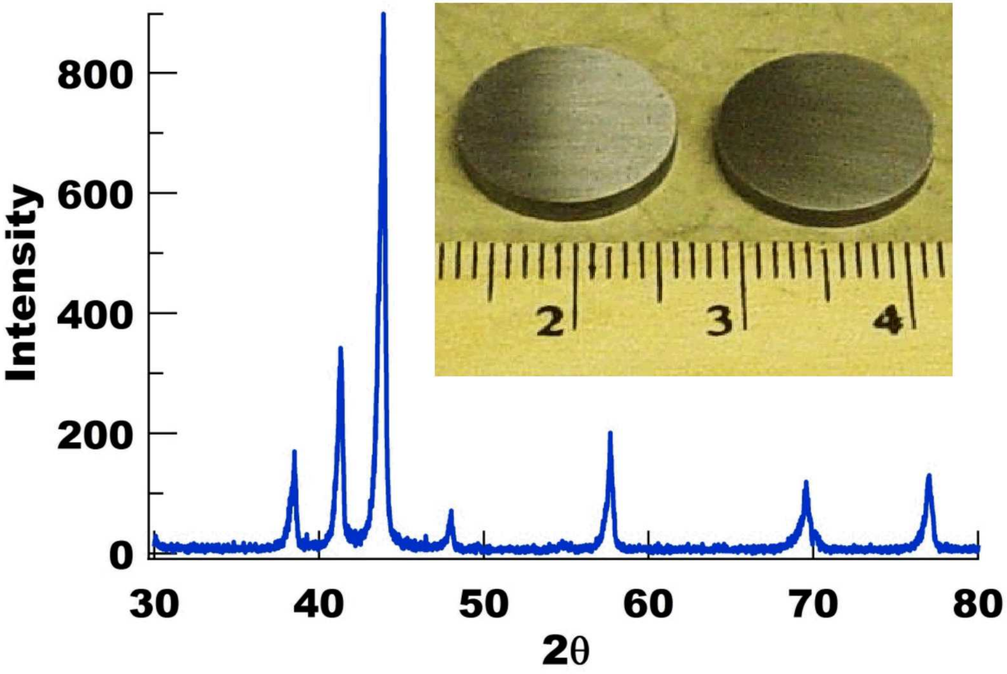


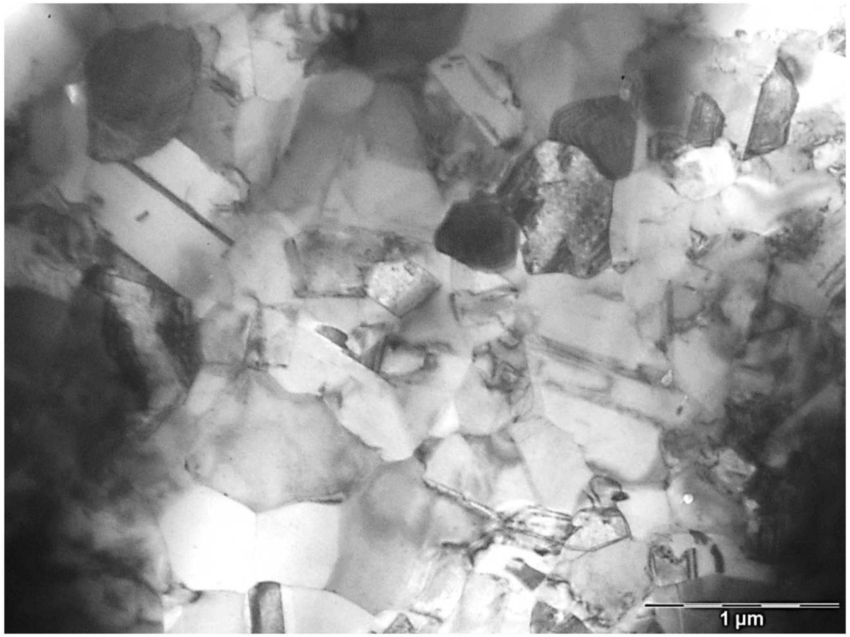
SPS Chamber



SPS consolidated Iron Nitride

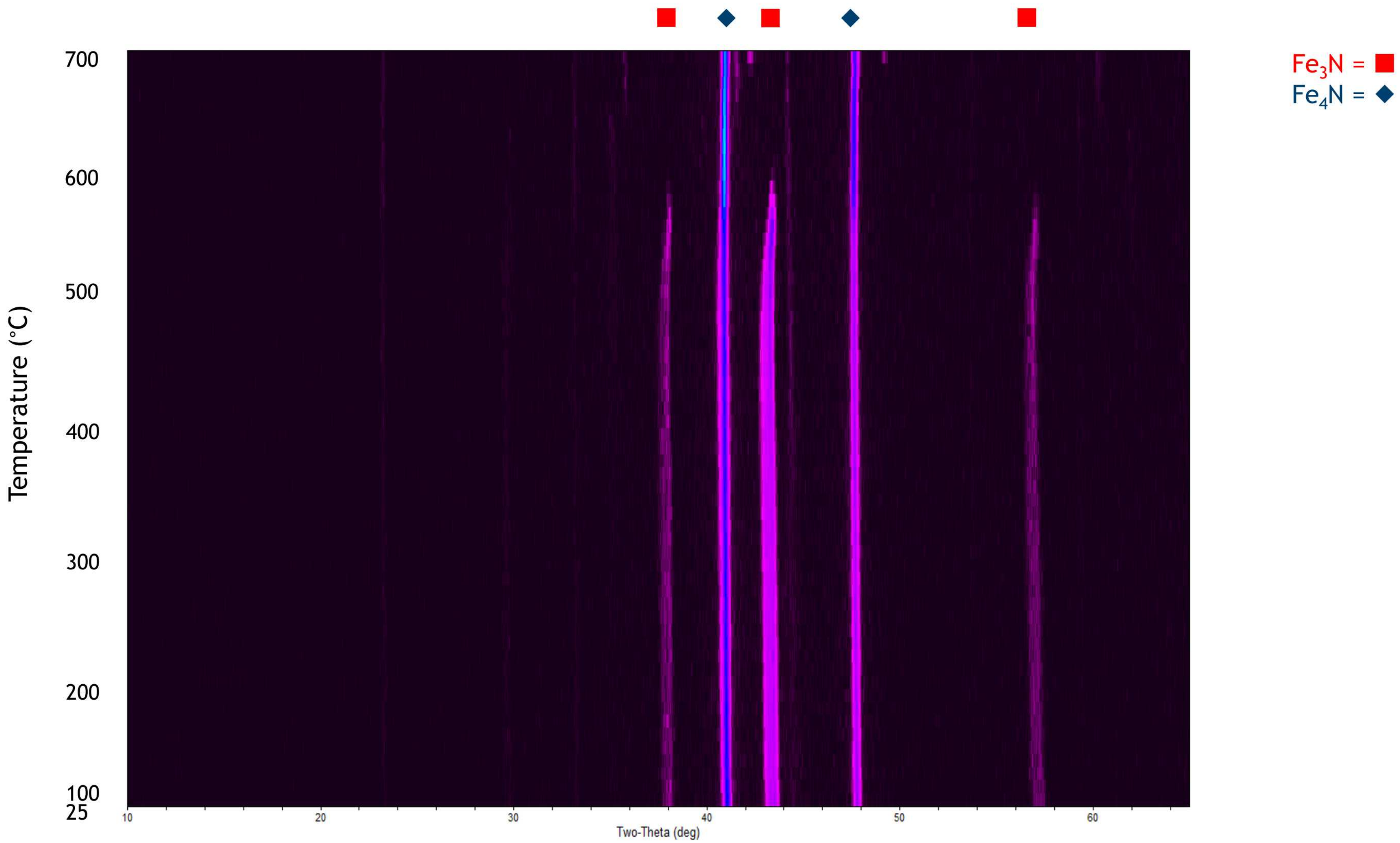
First ever bulk γ'-Fe_4N!





- **Fe nitride powders well consolidated with little porosity**
- **Grain sizes 200 nm – 1 µm → fine grain size = low loss**
- **γ'-Fe_4N primary phase**
- **Fe_3N secondary phase from mixed phase starting material**

Continued efforts to improve phase purity



Powder XRD, mixed phase iron nitride

Coercivity as a Function of Particle Size

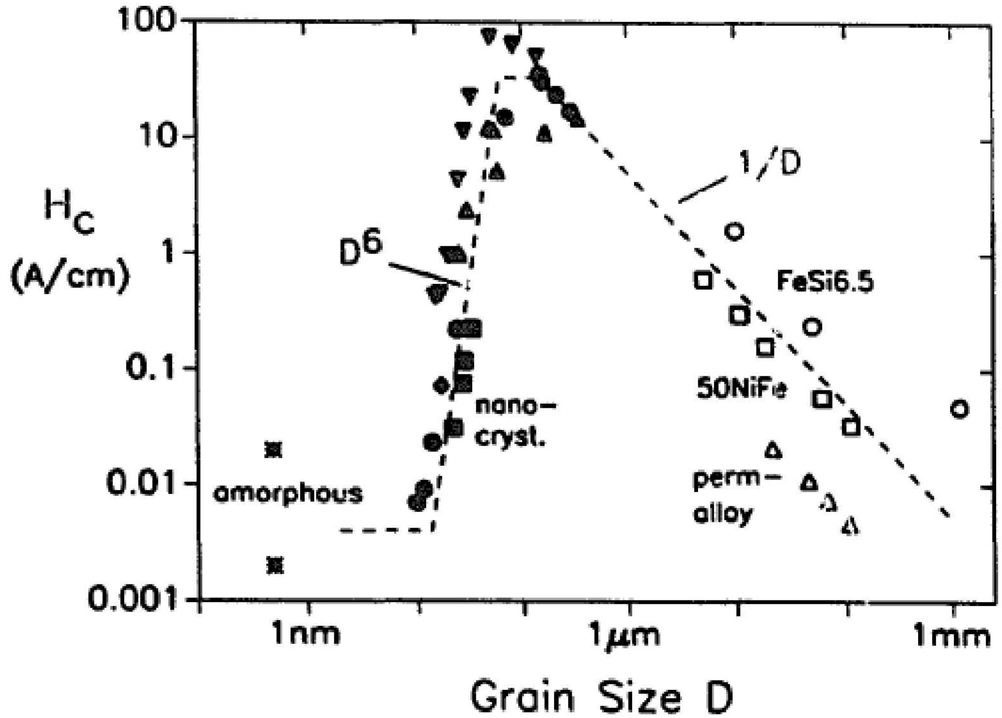
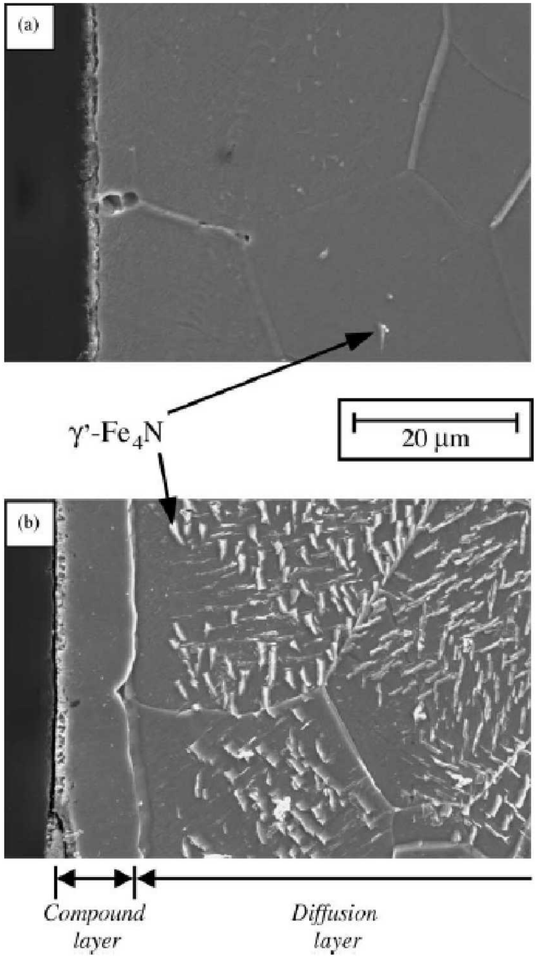


Fig. 2. Coercivity H_c vs. grain size for various soft magnetic metallic alloys. The data of the nanocrystalline material refer to (▲) FeNbSiB and (●) FeCuNbSiB [14], (◆) FeCuVSiB [15], (■) FeZrB [4] and (▼) FeCoZr [16].

G. Herzer, Nanocrystalline Soft Magnetic Materials, J. Magn. Mag. Mat., 112, 258 (1992).

Electrochemical Nitridation of Fe(0)

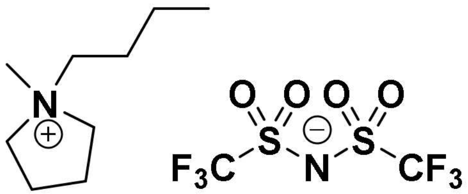
- **Growth of γ'-Fe_4N demonstrated in LiCl-KCl eutectic melt**
- **γ'-Fe_4N Formed at surface of Fe(0) electrode**
  - **Li_3N nitride source**
- **Demonstrates electrochemical synthesis of iron nitride possible**
- **Requires >700 K**
- **Is a lower temp approach possible?**

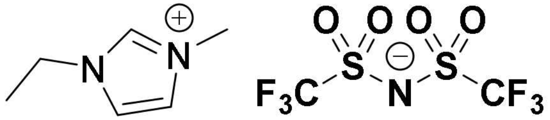


Ito *Journal of Alloys and Compounds* **2004**, *376*, 246.

Low Temperature Approach- Li_3N Solubility is Poor in ILs

- Two Ionic liquids considered
  - Both have excellent electrochemical stability





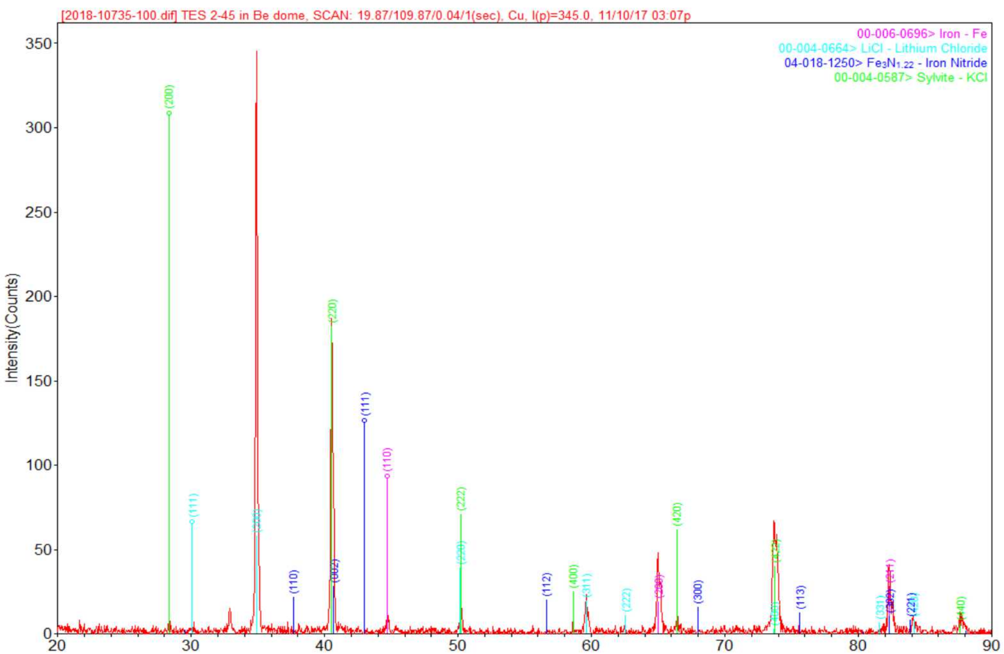
Lewandowski *Electrochemica Acta* **2006**, *51*, 5567.

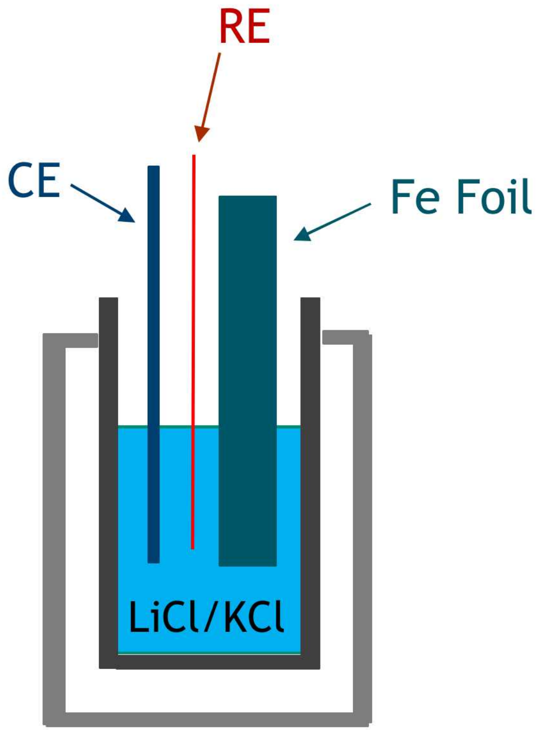
- Even with the use of crown ethers, we have yet to observe solubility of Li_3N

Electrochemical Nitridation of Fe(0)- LiCl/KCl

- Conditions similar to Ito
    - 450 ºC LiCl/KCl
    - 1 mol % Li_3N

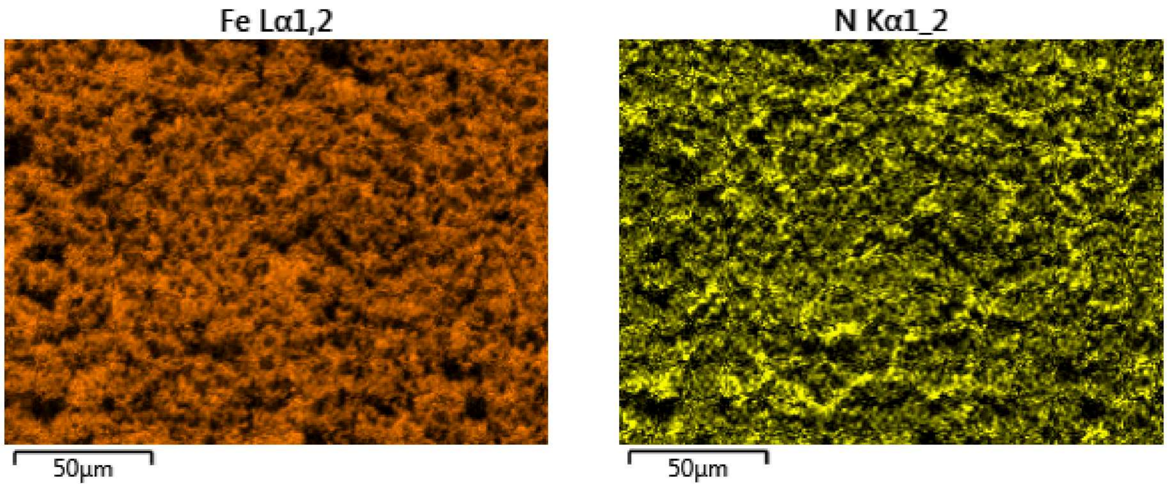
- Non-trivial to reproduce results
- Initial attempts did not produce nitride layer

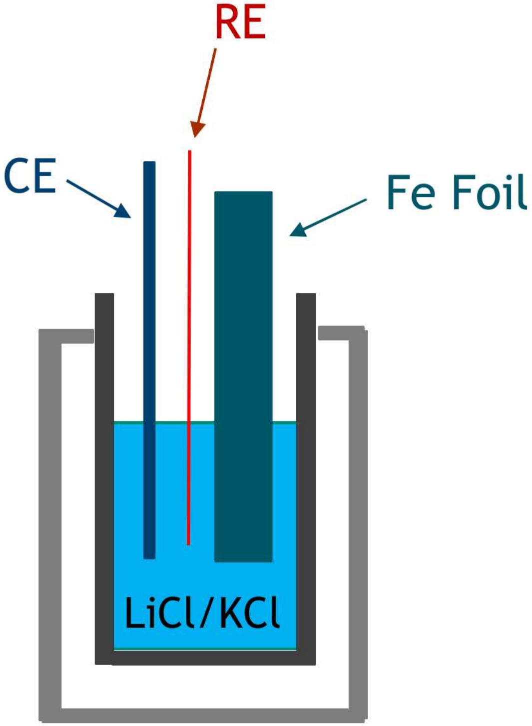




Electrochemical Nitridation of Fe(0)- LiCl/KCl

- Subsequent attempts have produced nitride layer
  - Longer reaction time
  - Addition of Li_3N in portions

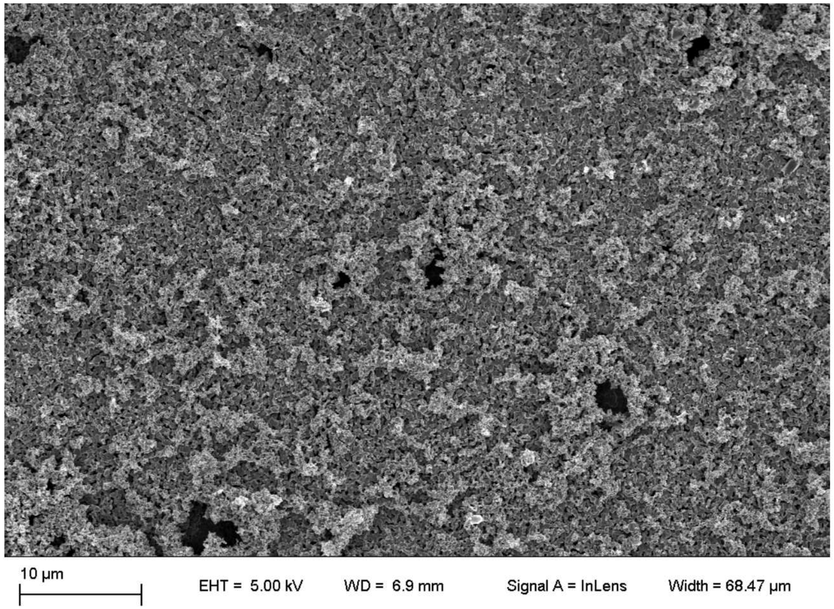


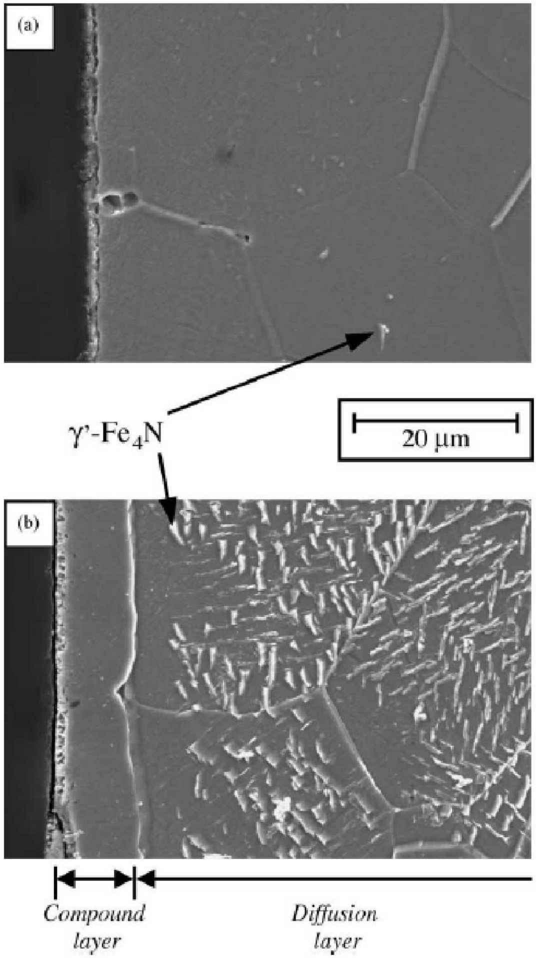


Electrochemical Nitridation of Fe(0)- LiCl/KCl

- Subsequent attempts have produced nitride layer

  However, surface appears very different



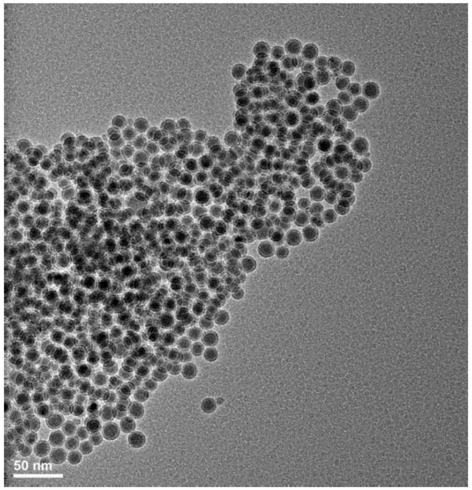


Ito *Journal of Alloys and Compounds* **2004**, *376*, 246.

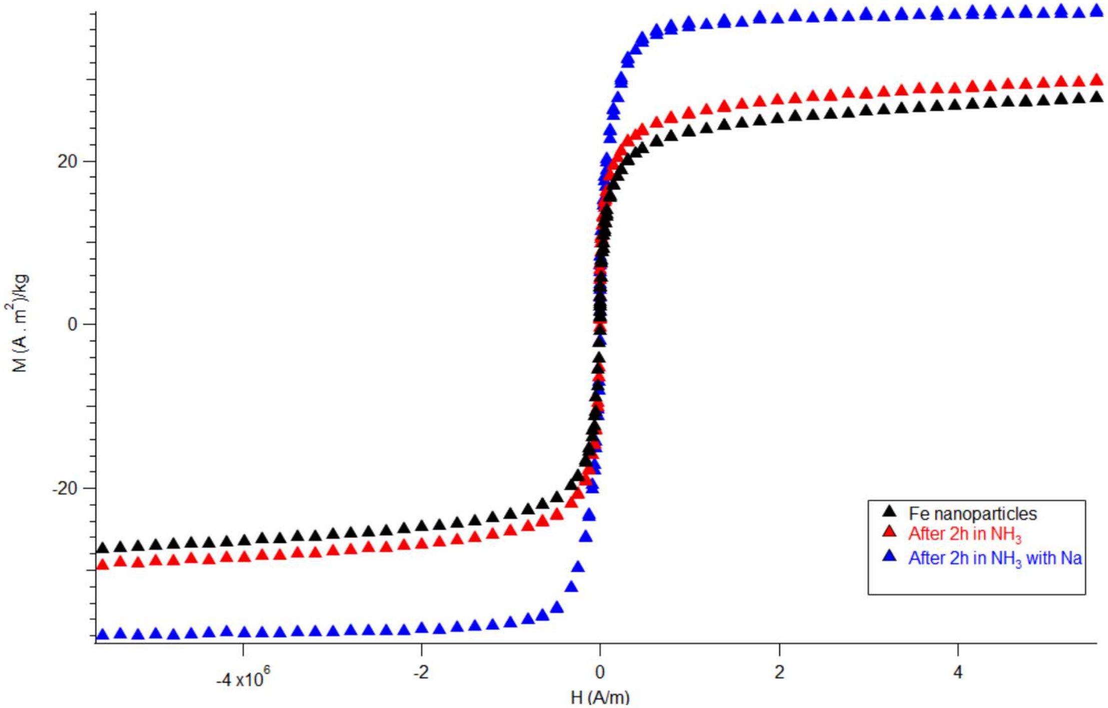
- A phase evolution vs. temperature (XRD) suggests FeN is the predominant phase

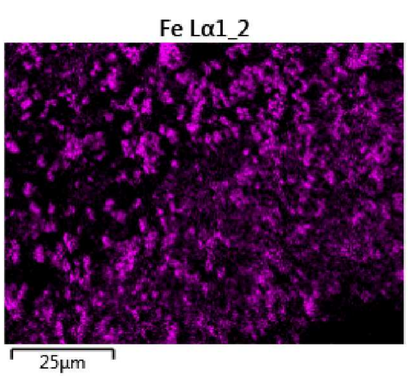
Nitridation of Iron Nanoparticles

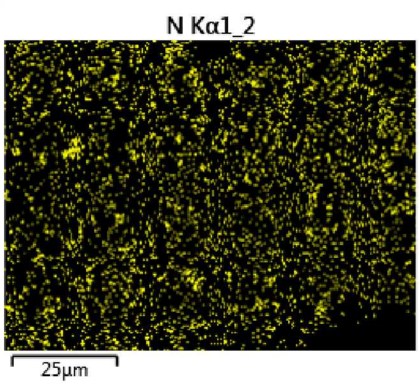
- Synthesis from well defined precursors
- Huber reported large-scale iron nanoparticle synthesis (Huber, 2011 *US Patent* #7,972,410)
  - Size control by reversible magnetic agglomeration



Magnetic Data Supports Formation of New Material

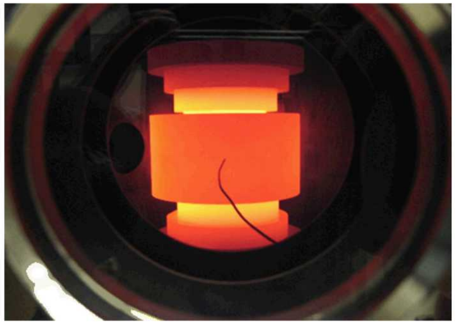


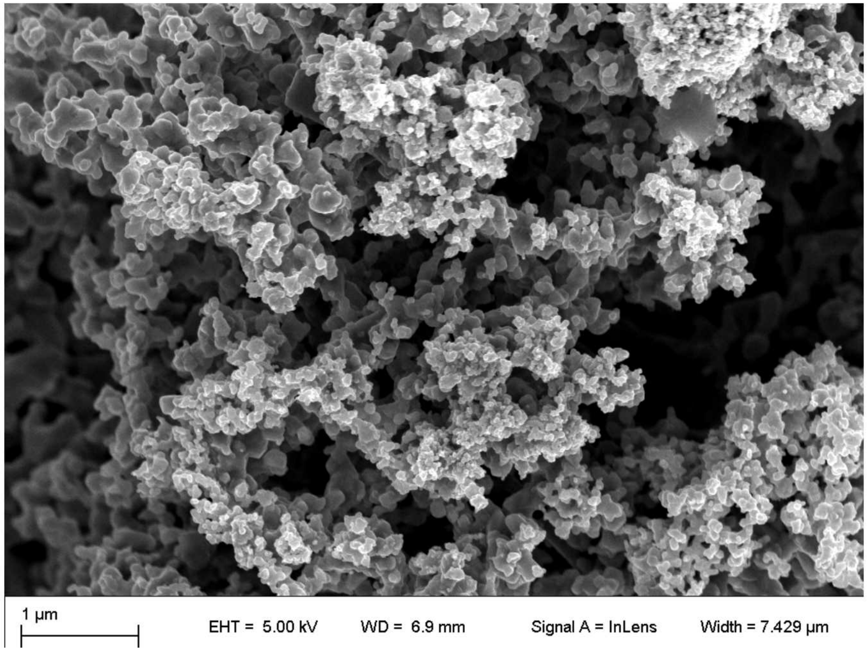




Summary

- γ'-Fe_4N is a promising low cost magnetic material for use in inductors and transformers
  - SPS consolidation has successfully been used to fabricate bulk γ'-Fe_4N
  - Nitridation of iron foil
    - Access to materials with high resistivity
  - Nitridation of iron nanoparticles
    - Fine control over size and morphology





Acknowledgements

The authors acknowledge support for this work from Dr. Imre Gyuk and the Energy Storage Program in the Office of Electricity Delivery and Energy Reliability at the US Department of Energy