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Making Every Microgram Count: Nanocalorimetry for Nuclear and Ultra-Rare Materials

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Changing the World's Energy Future

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Idaho National Laboratory

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Making Every Microgram Count: Nanocalorimetry for Nuclear and Ultra-Rare Materials

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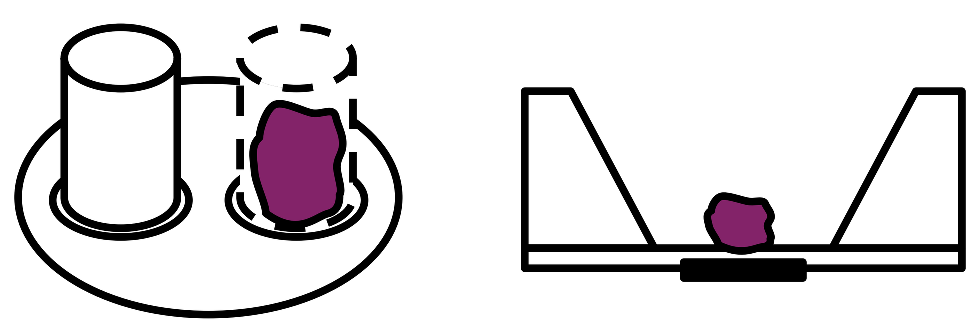
Arizona State University & INL Thermophysical Properties (U230)

Background

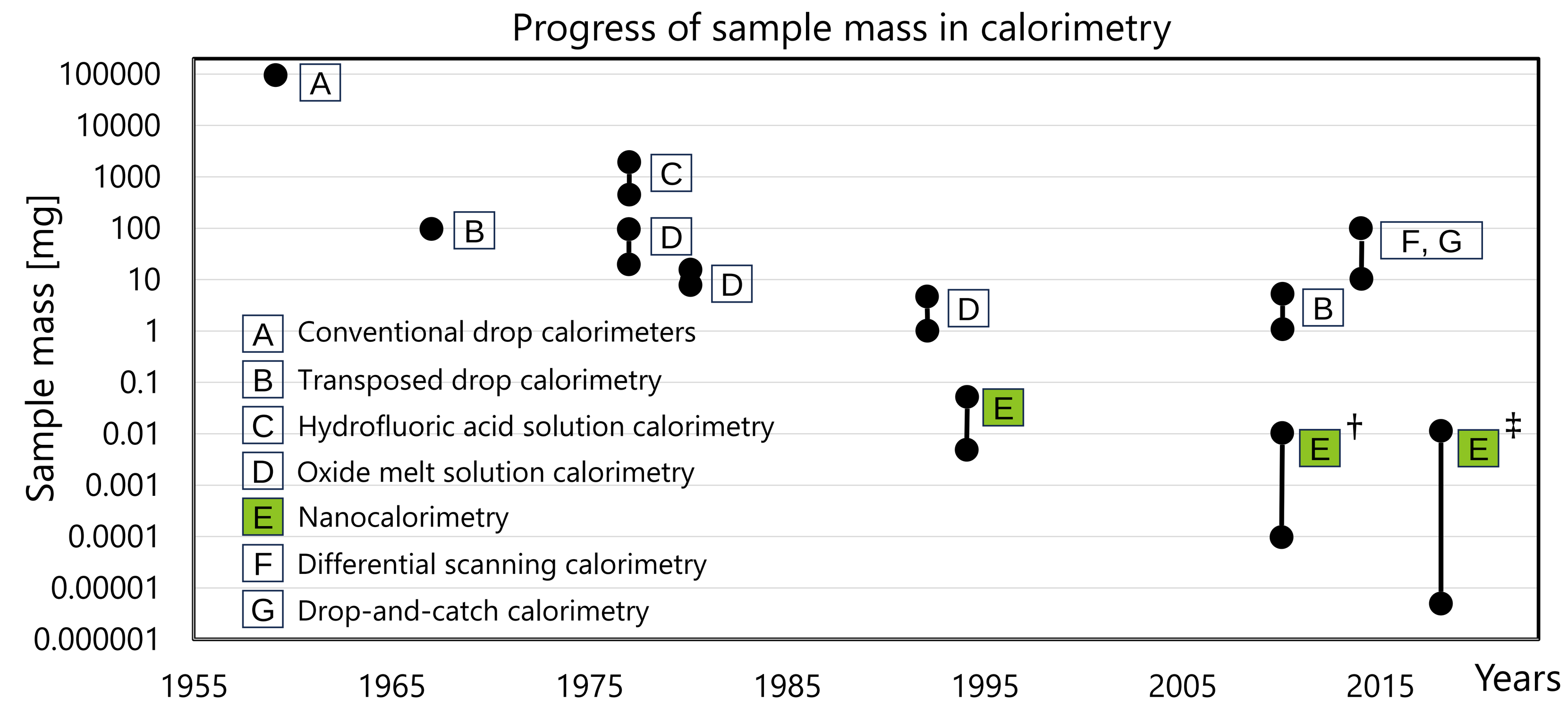
- Thermal analysis of materials (e.g., heat effects, heat capacity, radiation damage) plays important role in nuclear energy, planetary, environmental and technological settings
- **Traditional DSCs** (differential scanning calorimeters): **measurement capabilities restricted** when studying limited ultra-rare accessory minerals or extreme conditions materials
- **Nanocalorimeters**: recent commercial availability (last 15 years), possibility of measuring extreme heating and cooling rates, **measurements with small amounts of material**
- We investigate the usage of **nanocalorimetry for the energetic analysis of nuclear or ultra-rare materials**, such as irradiated materials and high-pressure phases, with an emphasis on radiation damage and heat capacity measurements



Traditional DSC — Crucibles — ~10^1 to 10^2 mg

Nanocalorimetry — MEMS — ~10^{-6} to 10^{-3} mg

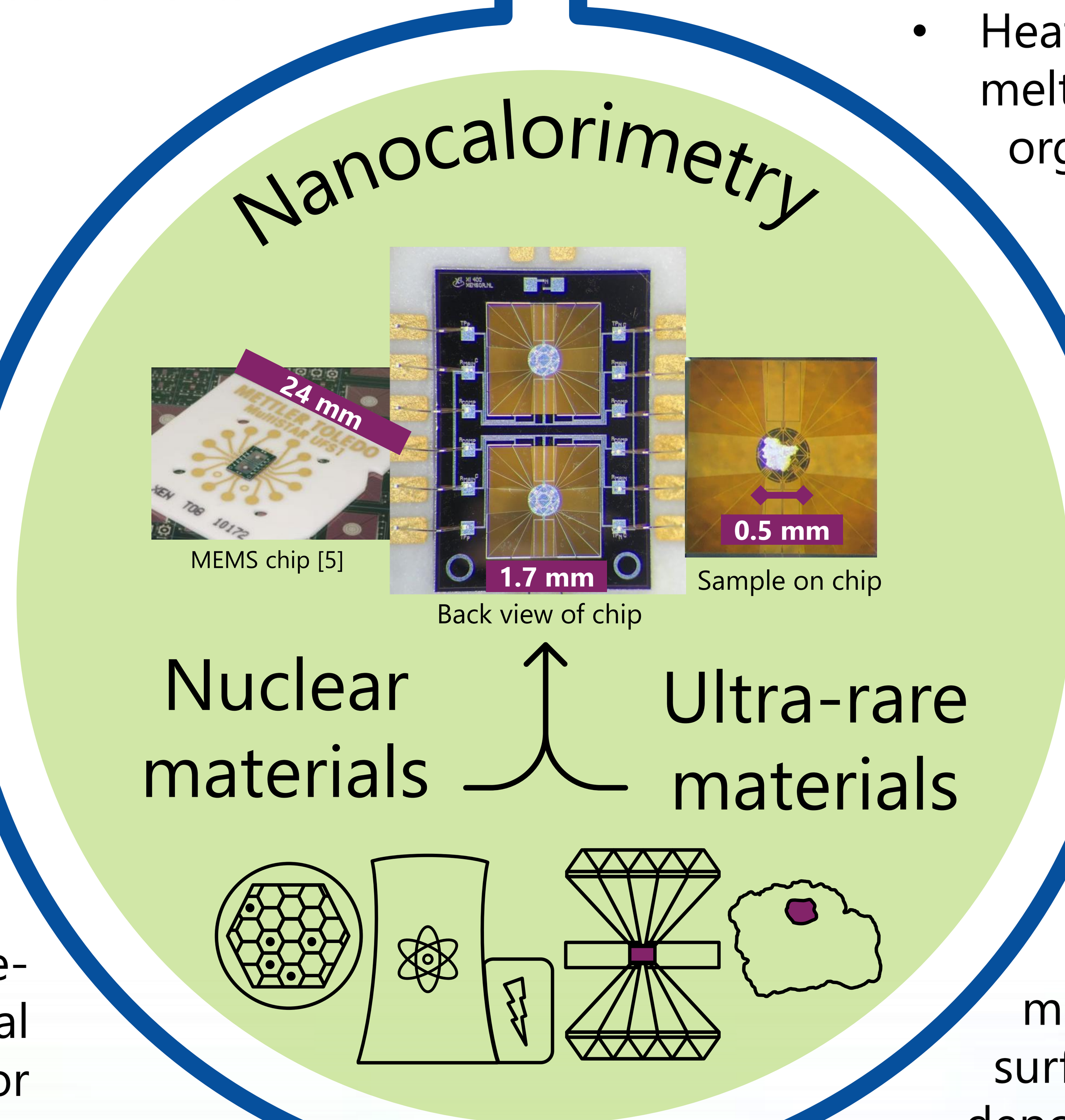
Results



† First (Flash DSC 1) and ‡ second (Flash DSC 2+) commercial nanocalorimeters by Mettler Toledo
All values are from sources [1-8]

Summary of nanocalorimetric measurements reported:

- Heat capacity, phase transitions, precipitation, melting, and nucleation of mostly polymers, organic metals, and alloys
- Combination with in-situ techniques: XRD, TEM, micro-Raman, and time-of-flight mass spectrometry
- Nanocalorimetry for ion-irradiated Si and radiation signature in PTFE



Objective & Methods

Objective:

Write a literature review evaluating the state-of-the-art of nanocalorimetry and its potential usage for thermochemistry of nuclear and/or ultra-rare materials

Methods:

1. Identify what can potentially be done with nanocalorimetry through a **literature search** about:
   - Thermochemistry of materials in the nuclear fuel cycle, including metallic fuels, irradiated materials, uranium oxides/silicides/carbides/nitrides, $(U/Pu)_xO_y$ mixed-oxides, silicon carbide, molten salts, fluorite structured materials and pyrochlores
   - Nanocalorimetry for thermal analysis of small amounts of samples at ultra-fast rates
2. Conduct **Flash DSC2+ measurements** for initial data of nanocalorimetry for radiation damage

Outlook & Future Work

Outlook: gaps in the literature consist of quantitative measurements of heat events in materials, which do not have a controlled surface contact on the chip (e.g., no melting, no deposition on chip or FIB sectioned), including irradiated oxides and metals

Future work:

1. Discuss **beneficial outcomes** in safety and environment
2. List **challenges** and propose advances necessary in nanocalorimetry techniques
3. Layout **possible directions for future experiments** of heat capacity and radiation damage
4. After installation of the Flash DSC2+, conduct **experiments with irradiated ceramics** for quantitative measurements of radiation damage energetics

Mettler Toledo Flash DSC2+ [9]

[1] Navrotsky, A. (1977). *Phys Chem Miner* **2**, 89-104; [2] Navrotsky, A. (1979). *Annu Rev Earth Pl Sc* **7**, 93-115; [3] Denlinger, D. W., Abarra, E. N., Allen, K., Rooney, P. W., Messer, M. T., Watson, S. K. & Hellman, F. (1994). *Rev Sci Instrum* **65**, 946-959; [4] Navrotsky, A. (1997). *Phys Chem Miner* **24**, 222-241; [5] van Herwaarden, S., Iervolino, E., van Herwaarden, F., Wijffels, T., Leenaers, A. & Mathot, V. (2011). *Thermochimica Acta* **522**, 46-52; [6] Navrotsky, A. (2014). *J Am Ceram Soc* **97**, 3349-3359; [7] Ushakov, S. V., Shvarev, A., Alexeev, T., Kapush, D. & Navrotsky, A. (2017). *J Am Ceram Soc* **100**, 754-760; [8] Mettler Toledo Flash DSC2+ Promotional Brochure (2021); *[9] https://www.mt.com/us/en/home/products/Laboratory_Analytics_Browse/TA_Family_Browse/Flash_DSC.html accessed on 07/13/2023.*

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