Model Calibration Methods for Phase Diagram Determination

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SDRD
Starbucks Directed Research and Development

Coffee: Plain black coffee, brewed less than an hour ago.

Engineering Sciences & Applications Division

Materials Science & Technology

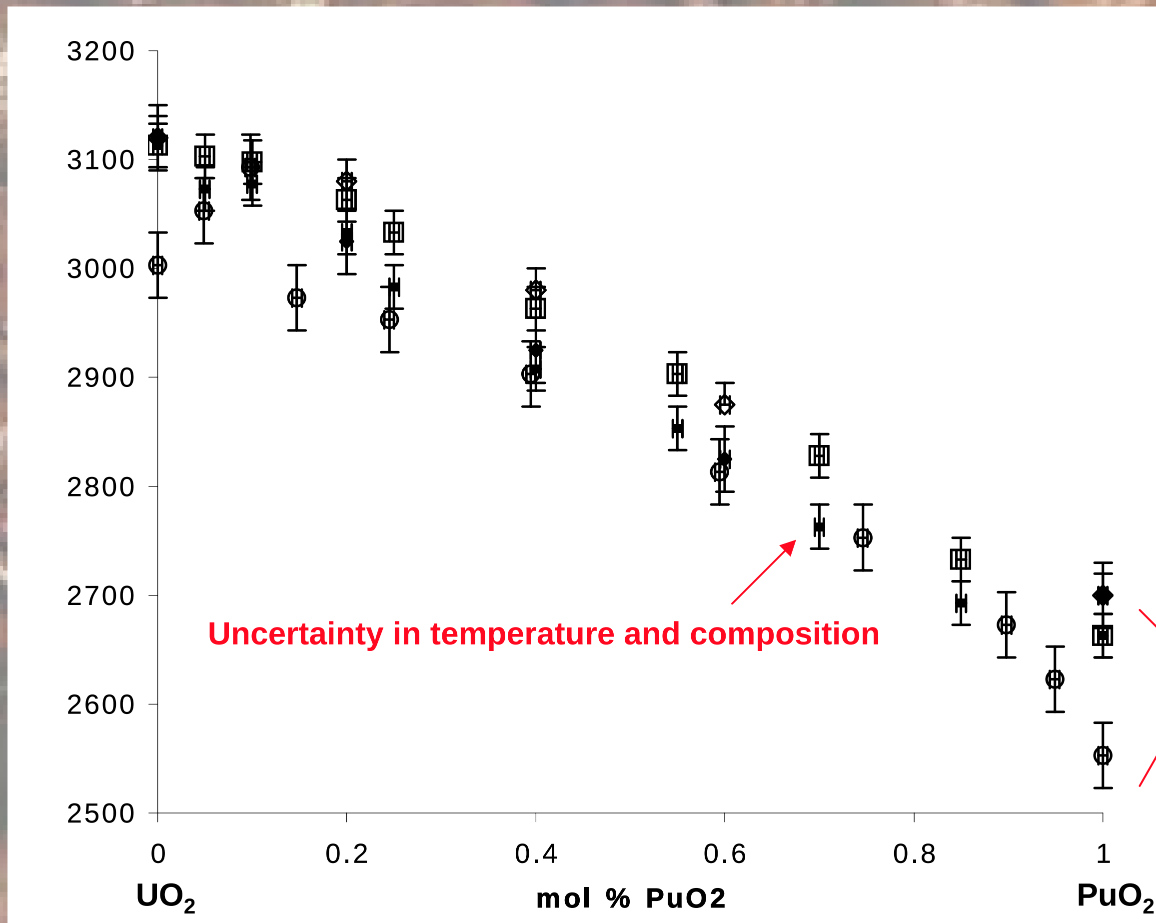
Why Model Phase Diagrams

- 5 to 10 component phase systems are often used in critical applications
  – Nuclear fuels
- One must predict phase transitions in these multi-component systems
  – melting points and eutectic compositions
  – volume changes
- However, it is not feasible to experimentally determine the entire phase diagram of a multi-component system
- Thus the need for modeling.

Latte: Espresso, steamed milk, and foam, not sweetened in any way unless you ask for syrup or sugar in it.

Uncertainty In Phase Diagrams

Where are the solidus and liquidus?



Many inconsistent data sources

Cappuccino: Like a latte, only much more foam.

The Uncertainty of the UO_2-PuO_2 Phase Diagram*

Collect available data

↓

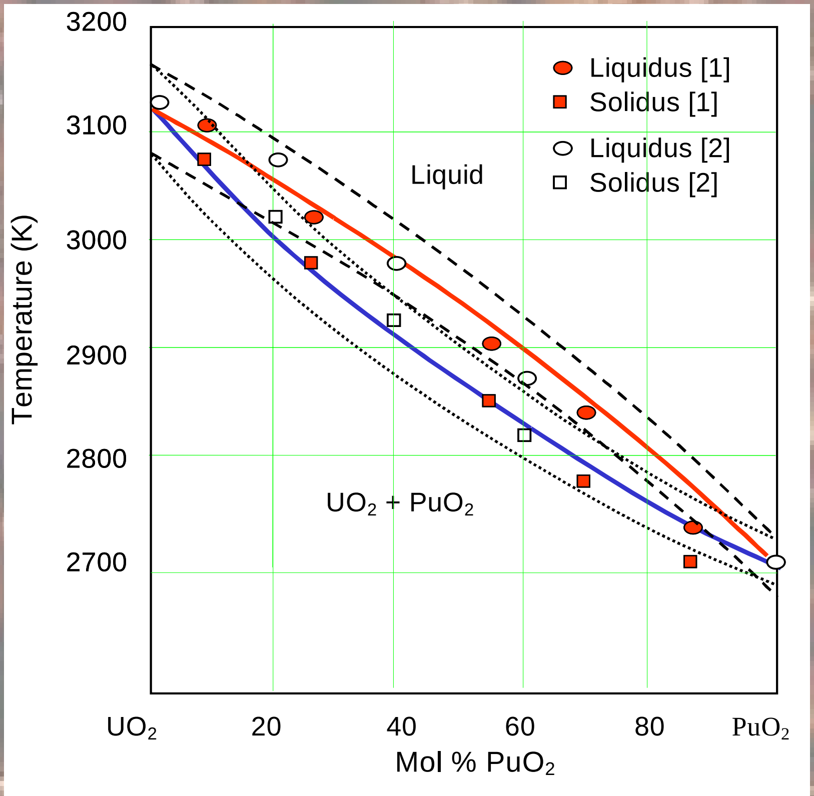
Bayesian Statistics and Genetic Algorithm

↓

Evaluate uncertainty

↓

Calculate diagram



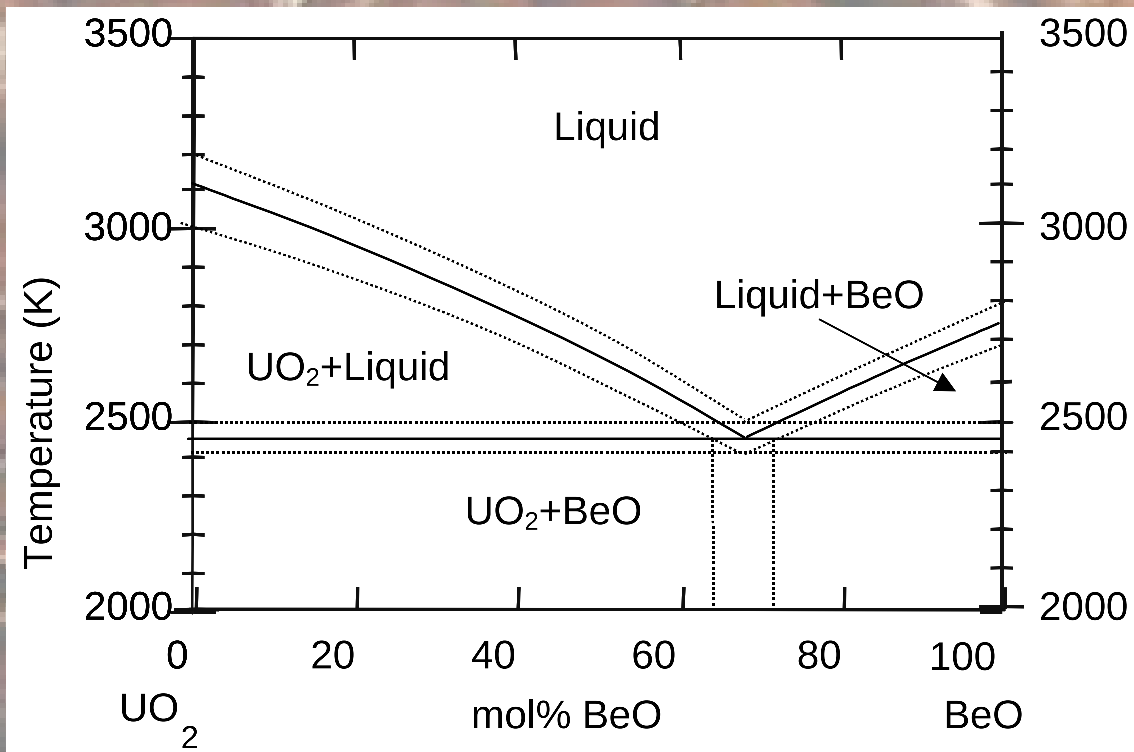
***M. Stan and B. J. Reardon, *CALPHAD*, 27 (2003) 319-323.**

[1] M. G. Adamson, E. A. Aitken, and R. W. Caputi, J. Nucl. Mater., 130 (1985) 349-365.

[2] T. D. Chikalla, J. Am. Ceram. Soc., 47 (1964) 309-309.

Americano: Espresso diluted with hot water until it's roughly the strength of regular coffee.

The Uncertainty of the UO_2-BeO Phase Diagram*



*M. Stan and B. J. Reardon, *CALPHAD*, 27 (2003) 319-323.

Experiemental by P. P. Budnikov, S. G. Tresvyatski, and V. I. Kushakovsky, Proc. 2nd U. N. Intern. Conf. Peaceful Uses At. Energy, Geneva, 1958, pp. 127.

Mocha: Espresso and steamed milk mixed with chocolate and served with whipped cream on top

Sources of Uncertainty

- Uncertainty in phase boundaries are due to:
  - Difficulty in measuring temperature
  - Difficulty in identifying the onset of a phase transition
  - Composition drift due to vaporization
- Many thermodynamic values of components are also uncertain
  - Melting Temperature (T^M)
  - Heat of Melting (ΔH^M)
- For these reasons, many authors report vastly different values for boundary positions

Which model should you use?

1) Ideal Solid Solution Law

$$x^{Liq}(T) = \frac{1 - \exp\left(\frac{\Delta H^M_{UO_2}}{R}\left(\frac{1}{T} - \frac{1}{T^M_{UO_2}}\right)\right)}{\exp\left(\frac{\Delta H^M_{PuO_2}}{R}\left(\frac{1}{T} - \frac{1}{T^M_{PuO_2}}\right)\right) - \exp\left(\frac{\Delta H^M_{UO_2}}{R}\left(\frac{1}{T} - \frac{1}{T^M_{UO_2}}\right)\right)}$$

$$x^{Sol}(T) = x^{Liq}(T) \cdot \exp\left(\frac{\Delta H^M_{PuO_2}}{R}\left(\frac{1}{T} - \frac{1}{T^M_{PuO_2}}\right)\right)$$

2) Polynomial in X

$$T_s(K) = a_s + b_s x + c_s x^2 + d_s x^3$$
$$T_l(K) = a_l + b_l x + c_l x^2$$

3) Polynomial in X

$$T_s(K) = a_s + b_s x + c_s x^2$$
$$T_l(K) = a_l + b_l x + c_l x^2$$

4) Polynomial in X

$$T_s(K) = T_{MUO_2} / \left(1 + b_s x + c_s x^2\right)$$
$$T_l(K) = T_{MUO_2} / \left(1 + b_l x + c_l x^2\right)$$

5) Model 1 & the eutectic UO_2-BeO system

$$x^{Liq}_{Liq+BeO}(T) = \exp\left(\left(\frac{-\Delta H^M_{BeO}}{RT}\right)\ln\left(\frac{T^M_{BeO}}{T}\right)\right)$$

$$x^{Liq}_{UO_2+Liq}(T) = 1 - \exp\left(\left(\frac{-\Delta H^M_{UO_2}}{RT}\right)\ln\left(\frac{T^M_{UO_2}}{T}\right)\right)$$

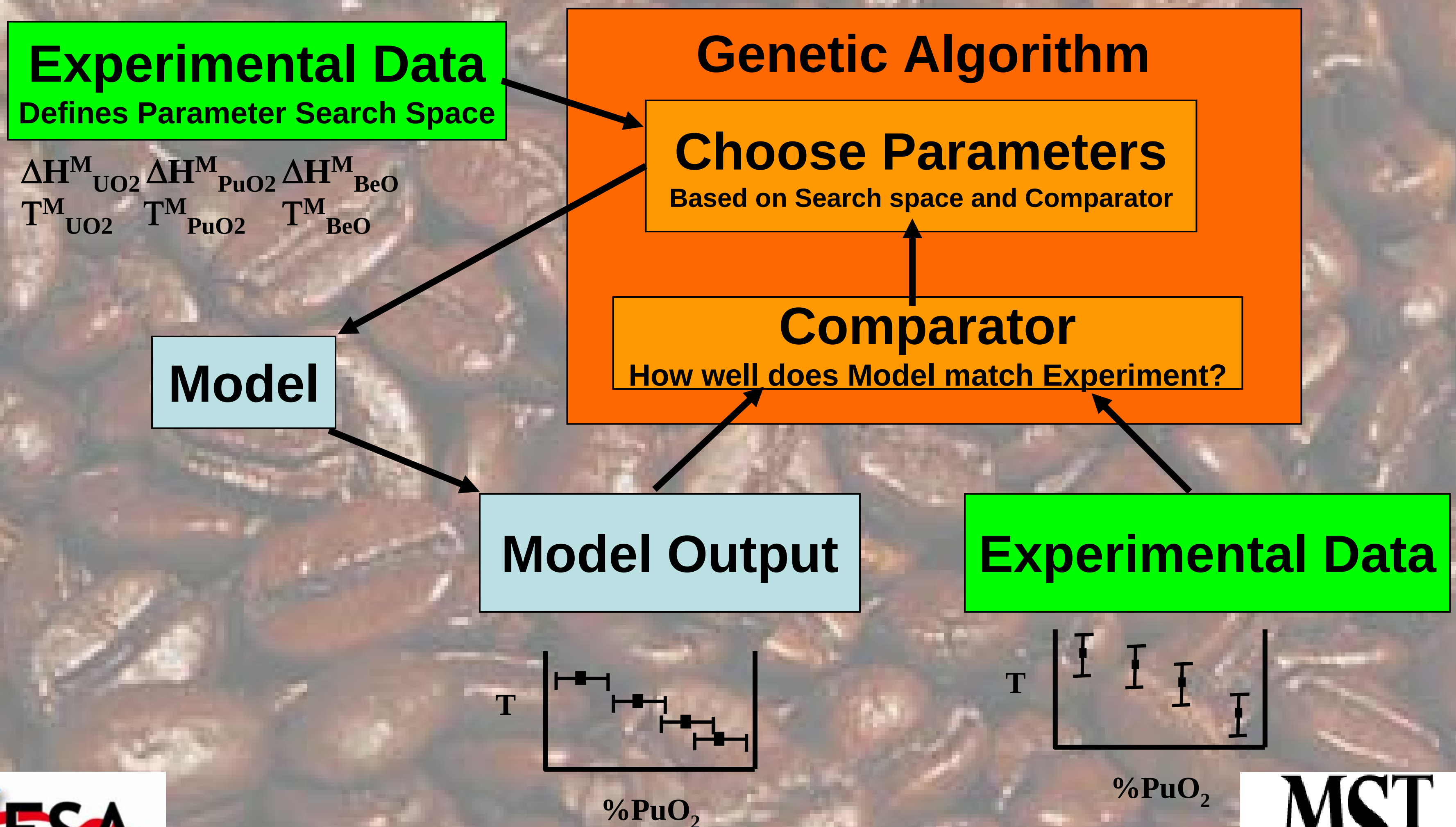
Mocha Valencia: A mocha with Valencia (orange) syrup and an extra espresso shot added, with whipped cream and orange sprinkles .

Pros and Cons of each Model

Model	Pros	Cons
1	Based on thermodynamics Model parameters applicable to other systems Model parameters experimentally accessible	Assumes ideal solution Model parameters uncertain Model: x(T), Data: T(x) - hard to fit
2,3,4	Model and data : T(x) - much easier to fit	Arbitrary functions and parameters not applicable to other systems Parameters not experimentally accessible
5	Same as Model 1 Incorporating another phase diagram constrains $\Delta H_{UO_2}^{M}$ and $T_{UO_2}^{M}$	Same as Model 1 Requires ΔH_{BeO}^{M} T_{BeO}^{M} Assumes perfect eutectic.

Cinnamon Spice Mocha: A mocha with cinnamon syrup added, served with foam and cinnamon on top rather than whipped cream.

The Calibration Problem



Why Use a Genetic Algorithm?

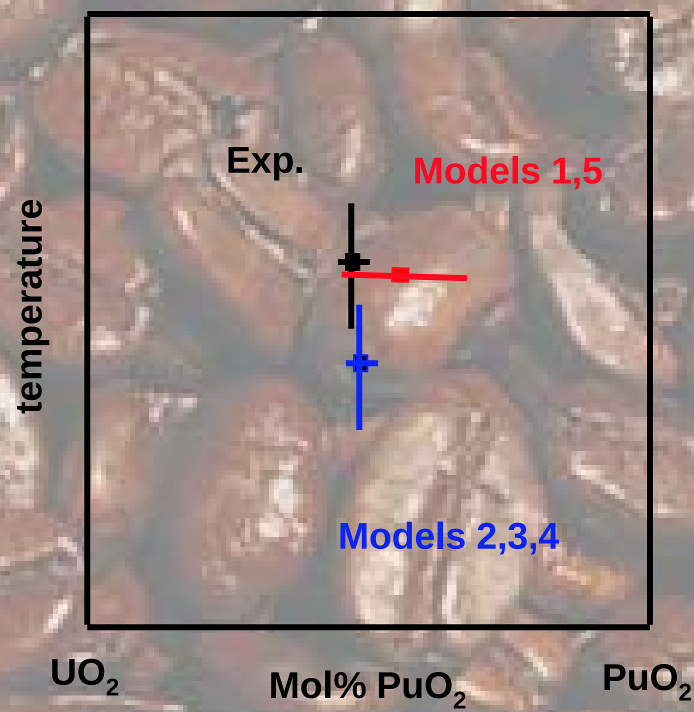
- **Robust to many classes of problems**
- **Does not assume distributional form of uncertainties**
- **Provides distributions and correlations of parameter values**
- **Using fuzzy rule set, the GA compares any number of experimental data points to model results**
- **The distribution of optimal solutions provides insight to experimental design*.**

B.J. Reardon, S. Bingert, *Acta Materialia*, 2000, 48(3), p.647-58

Mocha Macchiato: Espresso dropped into a cup of milk foam, and only foam

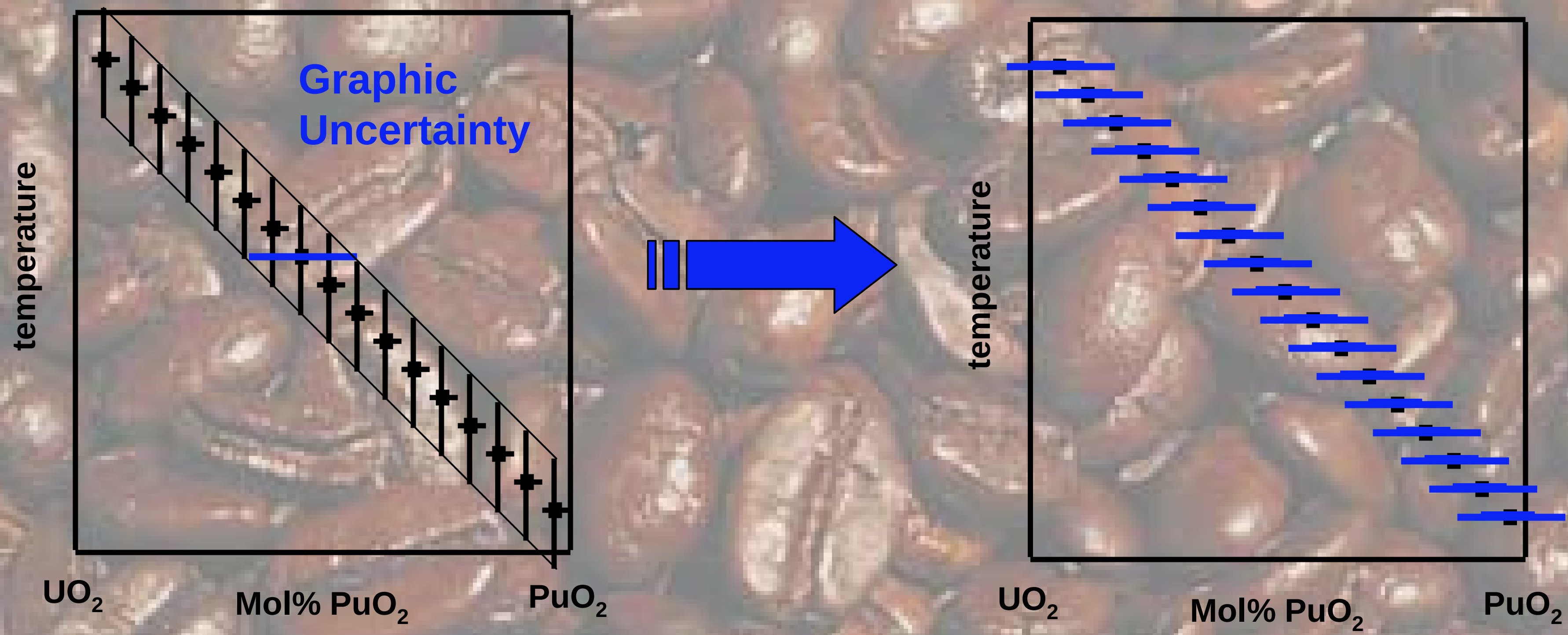
How to compare x(T) with T(x)?

Easy to compare:
- RMS
- Chi^2
- Kolmogorov-Smrinov
- Kullback-Liebler
- Jeffery's J
- Fuzzy rule set



Hard to compare:
- ?

Converting uncertainty in T(x) to x(T)



Short: 8 oz. The smallest size Starbucks offers

Applying the GA to this Calibration Problem

- Define model to be studied
- Define search range for model parameters
- Run GA
- Analyze the number and fitness of solutions
- Analyze how well each model was fit by the GA

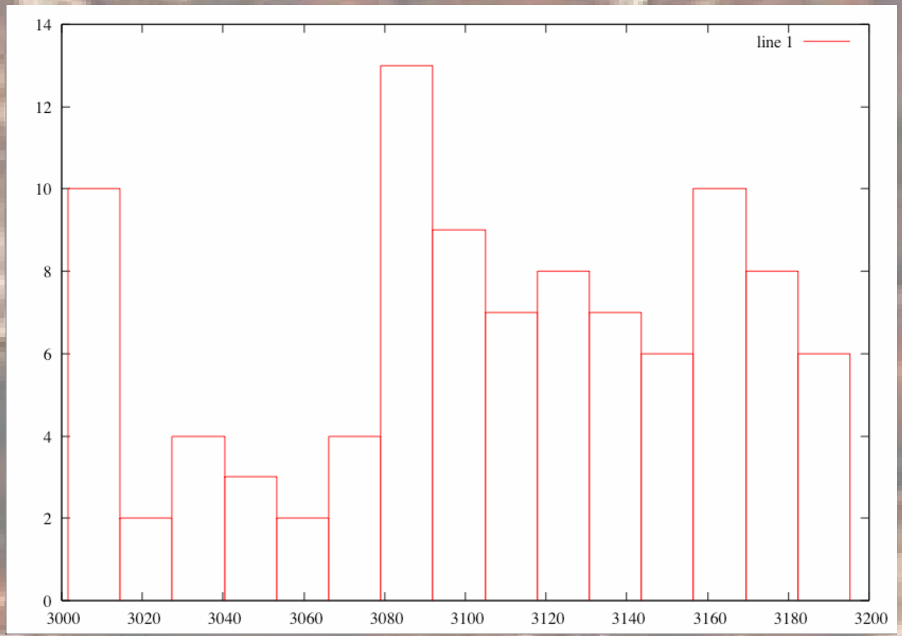
Results of Model 1 GA Calibration

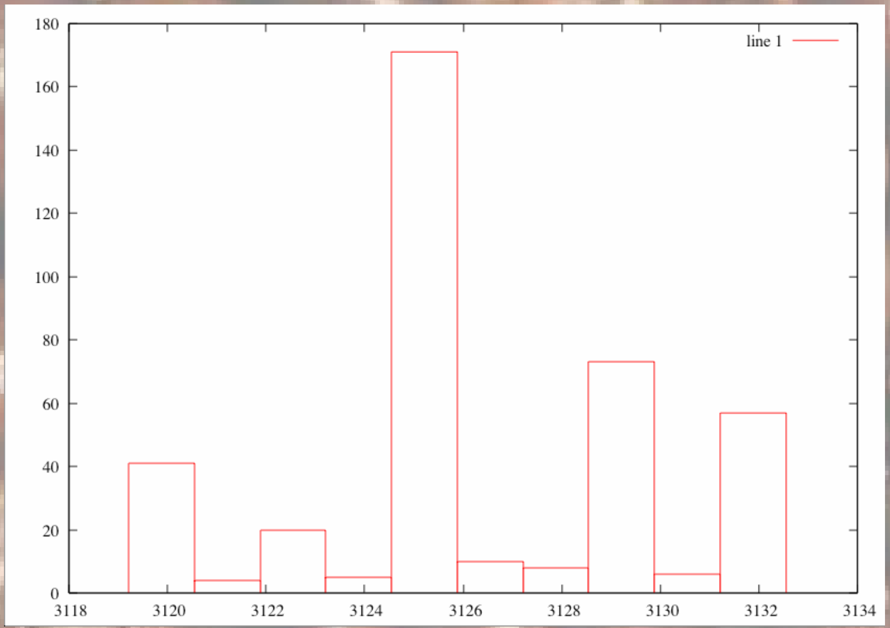
The results of optimizing model 1 against the available data sets.
C: Chikalla, L: Lyon and Baily, and A: Aitken and Evans.

	Model	Data Sets	# Sol	Fitness
1	1a	L	1	0.949274
2	1a	A	1	0.976892
3	1a	C	1	0.84066
4	1a	L+A	1	0.928932
5	1a	L+A+C	1	0.790519
6	1b	L	394	1
7	1b	A	1	0.989953
8	1b	C	1	0.887064
9	1b	L+A	1	0.99024
10	1b	L+A+C	1	0.874315

- **Model 1a vs. Model 1b**
  - **Model 1a: standard uncertainty in exp. X**
  - **Model 1b ‘Graphically driven’ uncertainty in exp. X**
- **# Solutions**
  - **number of solutions found**
  - **Lyon’s data best fits the thermodynamic model**
- **Fitness:**
  - **1 is the maximum - perfect fit**
  - **Goes up with ‘Graphically driven’ uncertainty**

Evolution of UO_2-T[M]





Initial

Final - 394 solutions after 72 generations

Venti: 20 oz. hot, 24 oz. cold. Pronounced "VENN-tee," and reportedly means twenty in Italian.

Results of Model 2 GA Calibration

The results of optimizing model 2 against the available data sets.
C: Chikalla, L: Lyon and Baily, and A: Aitken and Evans.

	Model	Data Sets	# Solutions	Fitness
11	2	L	5	0.998358
12	2	A	377	1
13	2	C	1	0.9591
14	2	L+A	14	0.995911
15	2	L+A+C	1	0.941159

- # Solutions
  - This model was originally developed for Aitken's data. This can be seen in the fact that a large number of solutions were found when using only Aitken's data set.
  - Unfortunately, this model can not be extended to any other phase system.

Decaf: Made with decaffeinated espresso, pulled from decaffeinated espresso beans.

Results of Model 3 GA Calibration

The results of optimizing model 3 against the available data sets.
C: Chikalla, L: Lyon and Baily, and A: Aitken and Evans.

	Model	Data Sets	# Solutions	Fitness
16	3	L	145	0.998775
17	3	A	291	1
18	3	C	502	1
19	3	L+A	82	0.993154
20	3	L+A+C	2	0.930555

- **# Solutions**
  - **This model was originally developed Chikalla's data. This can be seen in the fact that a large number of solutions were found when using only Chikalla's data set.**
  - **Unfortunately, this model can not be extended to any other phase system.**
  - **Also, it should be noted that the optimized parameters from each run (16-20) are significantly different.**

Results of Model 4 GA Calibration

The results of optimizing model 4 against the available data sets.
C: Chikalla, L: Lyon and Baily, and A: Aitken and Evans.

	Model	Data Sets	# Solutions	Fitness
21	4	L	3	0.983283
22	4	A	1	0.993808
23	4	C	449	0.960122
24	4	L+A	1	0.982623
25	4	L+A+C	1	0.920988

- # Solutions
  - This model does not fit any of the data sets well
  - Like the others, this model can not be extended to any other phase system.
  - The large number of solutions found when using Chikalla's data is not significant since the over all fitness of these solutions is so low.

Ristretto:. A normal shot of espresso takes about twenty seconds to pull; a ristretto shot is stopped at fifteen seconds.

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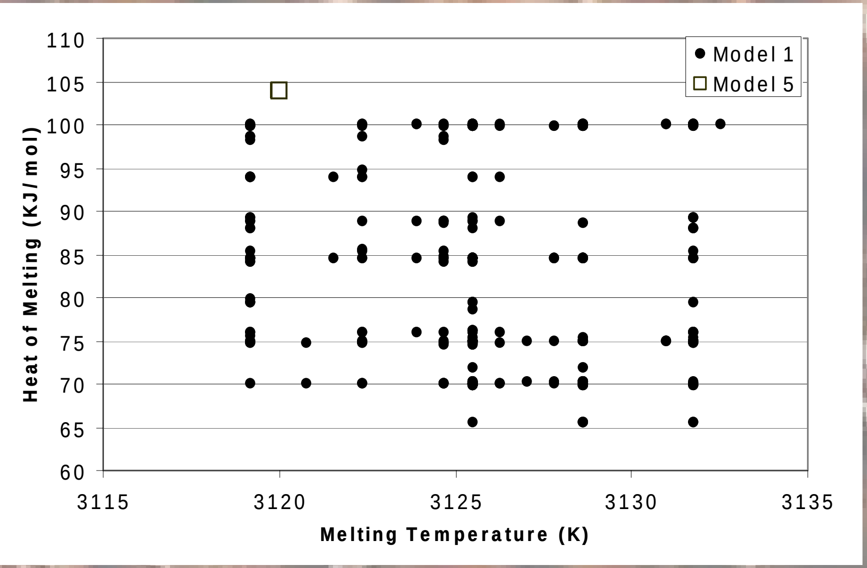
Results of Model 5 GA Calibration

The results of optimizing model 5 against the available data sets.
C: Chikalla, L: Lyon and Baily, and A: Aitken and Evans.

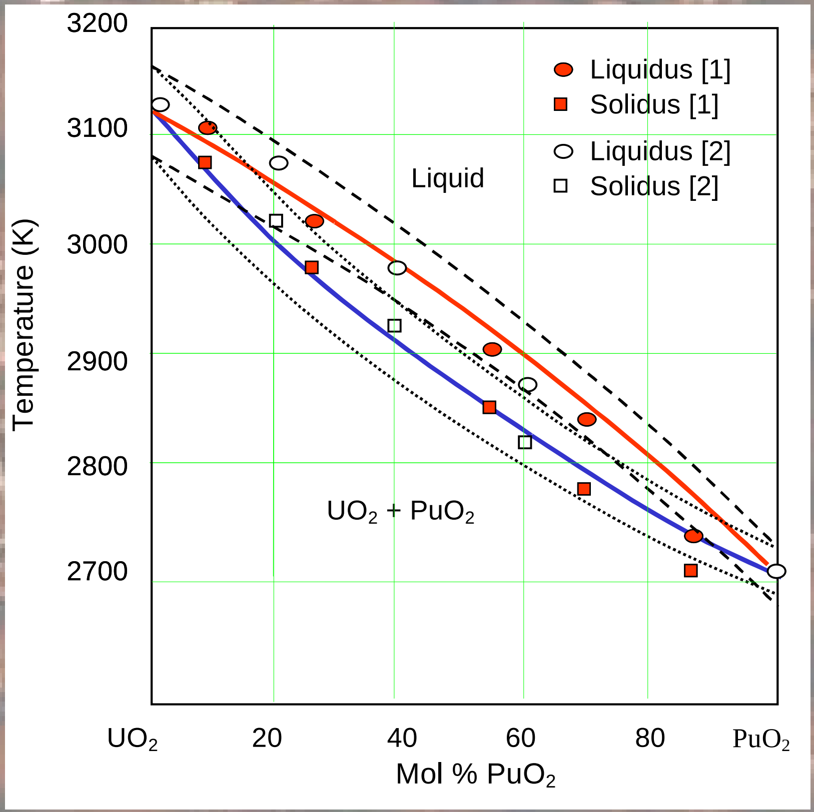
Test	Model	Data Sets	# Solutions	Fitness
26	5a	L	11	0.974211
27	5a	A	322	0.981022
28	5a	C	105	0.899736
29	5a	L+A	1	0.963978
30	5a	L+A+C	7	0.894648
31	5b	L	308	0.999815
32	5b	A	255	1
33	5b	C	395	0.933822
34	5b	L+A	43	0.995135
35	5b	L+A+C	127	0.930271



The final solution sets for the heats of melting and the melting points of UO_2 determined through the optimization of Model 1 (circle) and Model 5 (square).

Breve: Made with half and half instead of regular milk.

The Uncertainty of the UO_2-PuO_2 Phase Diagram*



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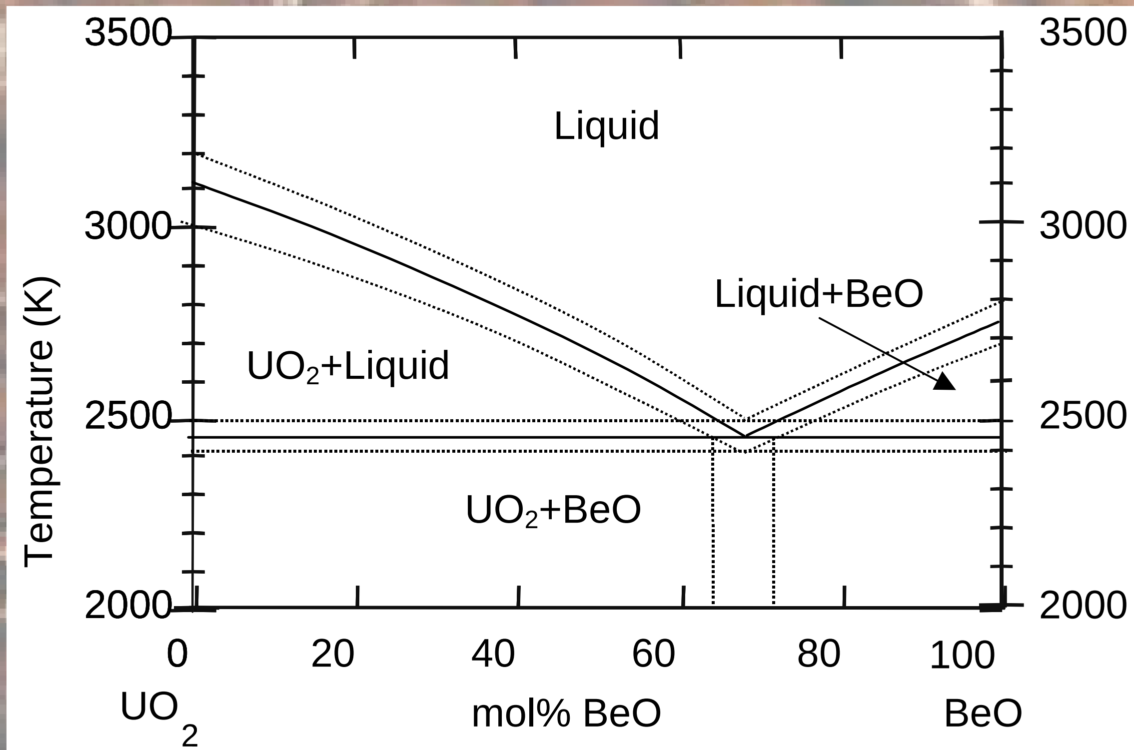
Misto: A drink consisting of half coffee, half steamed milk and a bit of foam.

The Uncertainty of the UO_2-BeO Phase Diagram*



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Kid's: By Starbucks rules, any drink that's going to be served to a child must be no hotter than 130 degrees.

Summary of Calibration Results

- **The 'graphic uncertainty' conversion was necessary to find a large number of solutions given the thermodynamic model**
- **While the polynomial models fit some of the data sets well they**
  - **Do not handle all the data well**
  - **Can not be extended to other systems**
- **Some data sets are more thermodynamically self consistent than others**

Conclusions

- GAs calibrate models using disparate, sparse, uncertain data sources.
- This calibration provides the overall predictive credibility of the models.
- The phase boundary uncertainties of the UO_2-PuO_2 and UO_2-BeO systems have been determined by accounting for:
  - –the available phase boundary data
  - –the accepted models of the phase boundaries
  - –the thermodynamic data used in the models.
- The net result is an internally self-consistent reduction in uncertainty of the values of the thermodynamic data as well as the phase boundaries.
- Modern heuristic optimizers such as GAs were crucial to this work since they are both robust and require no assumptions about the uncertainty distributions.