CARNEGIE MELLON UNIVERSITY
DEPARTMENT OF
MATERIALS SCIENCE AND ENGINEERING

Statistics of Grain Size Distributions

Sean Donegan

27-750 Texture, Microstructure, and Anisotropy

Monday, April 28, 2014

Outline

- Why grain size distributions?
- Statistical considerations of distributions
  - The log-normal distribution
  - PDFs and CDFs
- Theoretical approaches to grain size distributions
  - Hillert distribution
  - Mullins distribution
- Analysis of real microstructures
  - Visualizations
    - Histograms
    - eCDFs
    - Probability plots
  - Sampling
  - Extreme value theory

Why grain size distributions?

- **Grain size has a measurable effect on material properties**

Hall-Petch:

$$\sigma_y \propto \frac{C}{\sqrt{D}}$$

σ_y = yield stress
C = constant
D = 'grain size'

Creep:

$$\dot{\epsilon} \propto D^n$$

$\dot{\varepsilon}$ = strain rate
n = creep exponent
D = 'grain size'

- **Real grain sizes exhibit dispersion, which leads to a *grain size distribution***
- **So why only one 'grain size' in the phenomenological relationships?**

Why grain size distributions?

- Answer: no one likes to deal with statistics
- Another answer: it's hard
- Kurzydlowski attempted to incorporate grain dispersion into Hall-Petch by defining a constant size of grains (CSG) polycrystal
- Berbenni extended Kurzydlowski by defining a size-dependant constitutive equation for elasto-viscoplastic behavior
- Both these approaches assume log-normal distributions of grains; but are grain size distributions really log-normal?

K.J. Kurzydlowski, *Scr. Metall. Mater.*, vol. 24 pp. 879-884 (1990)
Berbenni et al., *Int. J. Plasticity*, vol. 23 pp. 114-142 (2007)

CARNEGIE MELLON UNIVERSITY

DEPARTMENT OF
MATERIALS SCIENCE AND ENGINEERING

Log-normal distribution

The log-normal distribution describes a random variable whose natural logarithm follows the normal distribution. The *cumulative distribution function* (CDF) of a log-normal distribution is:

$$F_{\mu,\sigma}(x) = \frac{1}{2}\operatorname{erfc}\left[-\frac{\ln(x)-\mu}{\sigma\sqrt{2}}\right] = \Phi\left[\frac{\ln(x)-\mu}{\sigma}\right]$$

The *probability density function* (PDF) of a log-normal distribution is:

$$f_{\mu,\sigma}(x) = \frac{1}{x\sigma\sqrt{2\pi}}\exp\frac{-(\ln x-\mu)^2}{2\sigma^2}$$

PDFs and CDFs

- The PDF of a random variable defines the probability of that variable taking a particular value (think 'bell curve' e.g. normal distribution)

$$\int_a^b f(x)\mathrm{d}x = 1$$

The integral of a PDF along its domain must equal 1 (i.e., if discrete, all probabilities sum to 1)

- The CDF of a random variable defines the probability that a value of the variable will be found <= x (think 's-curve')

$$F(x) = \int_{-\infty}^{x} f(t)\mathrm{d}t$$

The CDF can be defined as the integral of the PDF up to x

Theoretical approaches: Hillert

- **Hillert derived a limiting grain size distribution based on a presumed growth equation**
- **Assume grain boundary velocity is proportional to the local curvature:**

$$v = M\Delta P = M\sigma\left(\frac{1}{\rho_1} + \frac{1}{\rho_2}\right)$$

V = velocity
M = mobility
P = pressure

σ = grain boundary energy
ρ = radii of curvature

M Hillert, *Acta Metall.*, vol. 13 pp. 227-238 (1965)

Theoretical approaches: Hillert

- The rate of change should be equivalent to the integral of the velocity around the grain boundary surface
- This allows us to rewrite the velocity as a rate of change for the grain size:

$$\frac{\mathrm{d}R}{\mathrm{d}t} = \alpha M \sigma \left(\frac{1}{R_{cr}} - \frac{1}{R} \right)$$

R = circle/sphere equivalent radius
R_{cr} = critical radius

M Hillert, *Acta Metall.*, vol. 13 pp. 227-238 (1965)

Theoretical approaches: Hillert

- By the n-6 rule, the rate of change for grain size in 2D becomes:

$$\frac{\mathrm{d}R}{\mathrm{d}t} = \frac{M\sigma}{R}\left(\frac{n}{6} - 1\right)$$

n = number of sides

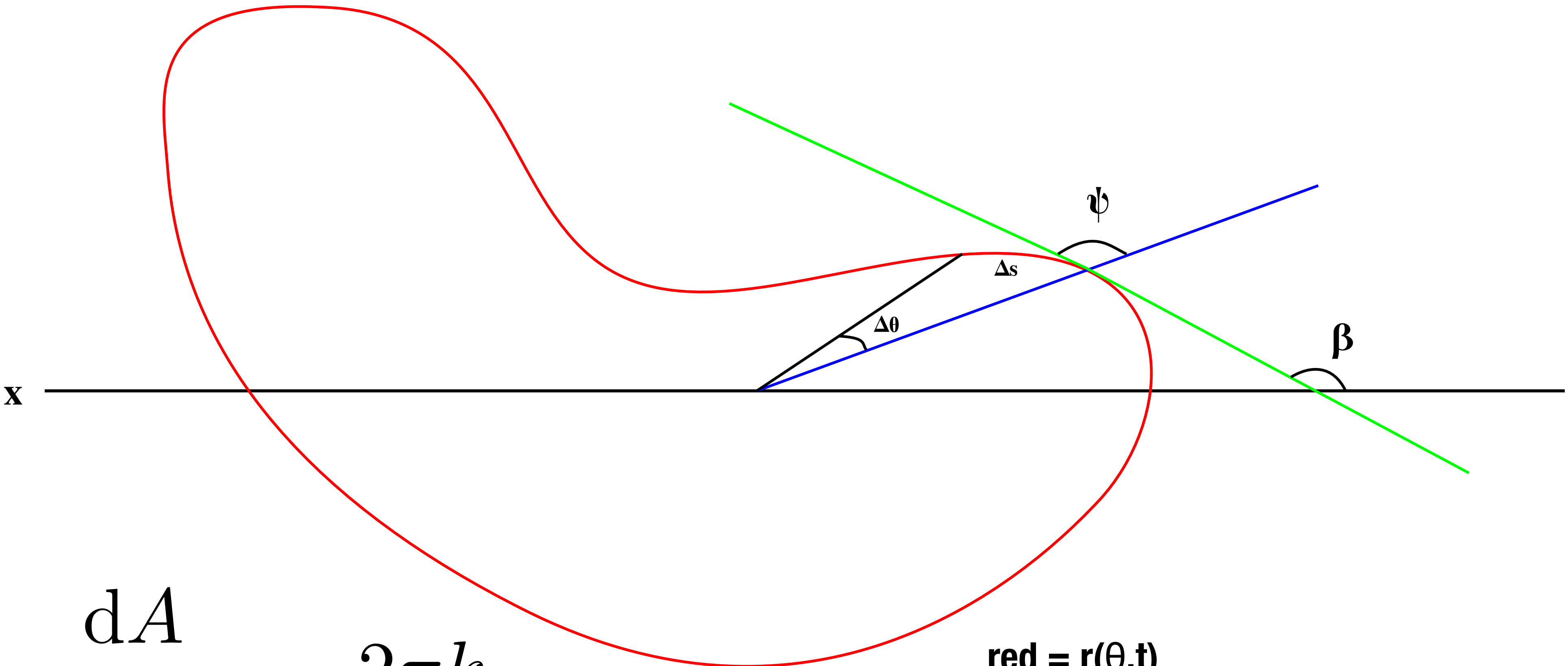
- But where does n-6 come from? Originally derived by von Neumann for soap froths, extended by Mullins

J. von Neumann, *Metal Interfaces*, ASM, Cleveland (1952)
W.W. Mullins, *J. Appl. Phys.*, vol. 27 pp.900-904 (1956)
M Hillert, *Acta Metall.*, vol. 13 pp. 227-238 (1965)

CARNEGIE MELLON UNIVERSITY
DEPARTMENT OF
MATERIALS SCIENCE AND ENGINEERING

n-6 rule

Consider a plane curve r(θ,t)



$$\frac{\mathrm{d}A}{\mathrm{d}t} = -2\pi k$$

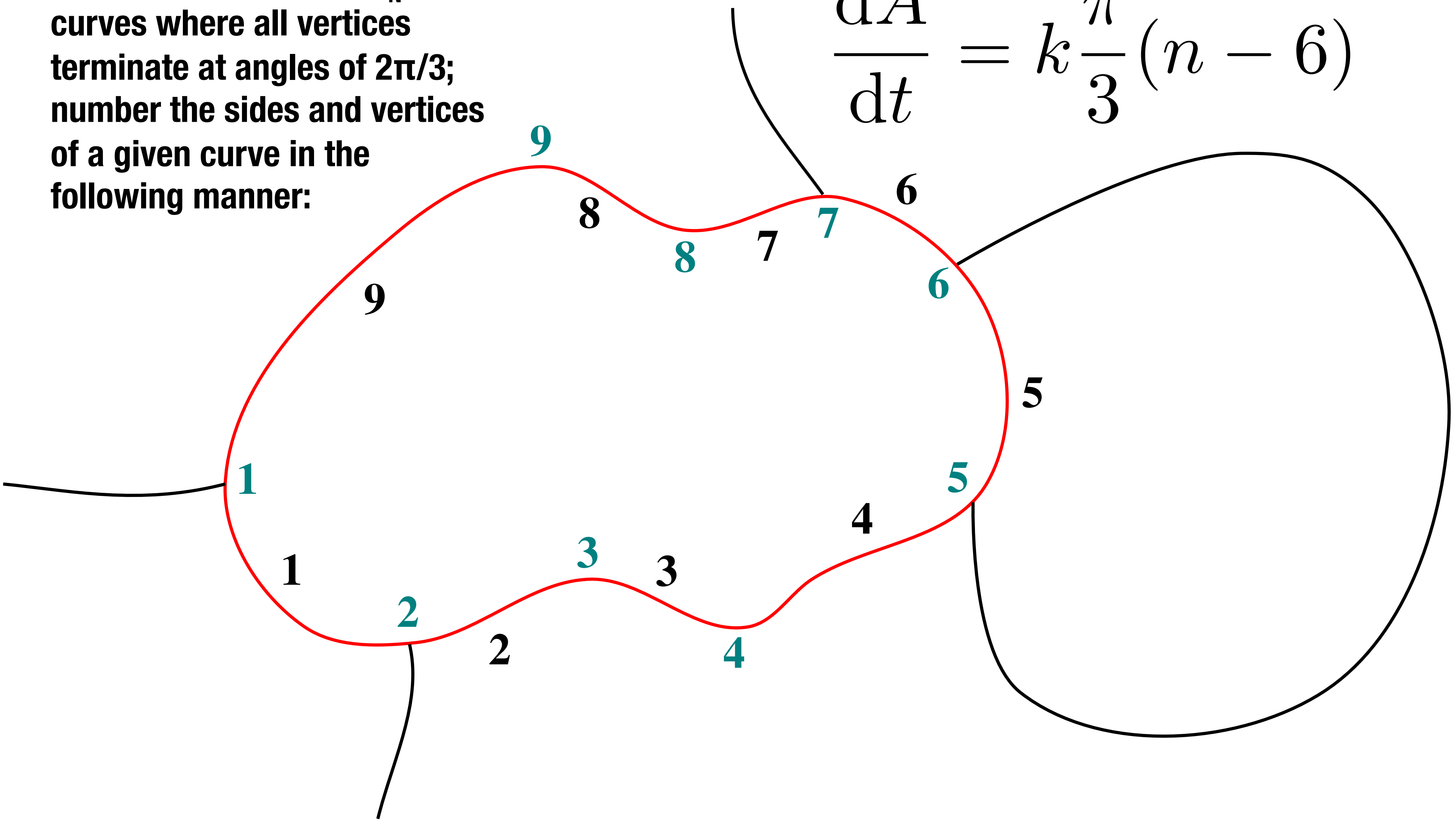
red = r(θ,t)
blue = polar vector
green = (directed) tangent
k = Mσ

J. von Neumann, *Metal Interfaces*, ASM, Cleveland (1952)
W.W. Mullins, *J. Appl. Phys.*, vol. 27 pp.900-904 (1956)

n-6 rule

Consider a network of $r_N(\theta,t)$ curves where all vertices terminate at angles of $2\pi/3$; number the sides and vertices of a given curve in the following manner:

$$\frac{dA}{dt} = k\frac{\pi}{3}(n-6)$$



J. von Neumann, *Metal Interfaces*, ASM, Cleveland (1952)
W.W. Mullins, *J. Appl. Phys.*, vol. 27 pp.900-904 (1956)

Theoretical approaches: Hillert

Now determine the average number of sides:

$$n = 6 + 6\alpha\left(\frac{R}{R_{cr}} - 1\right) \longrightarrow$$

$$\bar{n} = 6 + 6\alpha\left(\frac{\bar{R}}{R_{cr}} - 1\right) \longrightarrow$$

$$\bar{R} = R_{cr} \qquad \alpha = \frac{1}{2}$$

M Hillert, *Acta Metall.*, vol. 13 pp. 227-238 (1965)

Theoretical approaches: Hillert

Finally, after some calculus, arrive at the growth equation:

$$\frac{\mathrm{d}u^2}{\mathrm{d}\tau} = \gamma(u-1) - u^2$$

$u = R/R_{cr}$
$\gamma = 2\alpha M\sigma(dt/dR_{cr}^2)$
$\tau = \ln R_{cr}^2$

M Hillert, *Acta Metall.*, vol. 13 pp. 227-238 (1965)

Theoretical approaches: Hillert

The goal is now to arrive at a PDF for the limiting grain size distribution. After some (more) calculus:

$$P(u) = \frac{\beta u}{(2-u)^{2+\beta}} (2e)^{\beta} \exp \frac{-2\beta}{2-u}$$

Is it a PDF?

$$\int_0^2 P(u) \mathrm{d}u = 1$$

YES!

M Hillert, *Acta Metall.*, vol. 13 pp. 227-238 (1965)

Theoretical approaches: Mullins

- Mullins derived a more general form for the limiting grain size distribution that can extend up to ∞ (as opposed to just 2)
- The distribution requires a function of the number of sides of a grain (s(x)) and a function G(x) that is conceptually describes whether grains of a particular size will grow or shrink:

$$G(x) = x - \frac{d}{P}\int_x^{\infty} P(x')\mathrm{d}x'$$

x = R/<R>
d = dimensionality
P = PDF

NB: the integration need not be taken to ∞!

W.W. Mullins, *Acta Mater.*, vol. 46 pp. 6219-6226 (1998)

Theoretical approaches: Mullins

Inverting G(x) yields an expression for the PDF:

$$P(x) = \frac{d}{x - G} \exp[-\int_0^x \frac{d}{x' - G(x')} \mathrm{d}x']$$

Not all G(x) *necessarily* yield a true PDF!

If s(x) is defined (in 2D) as a linear function of the number of sides, then Mullins is degenerate to Hillert!

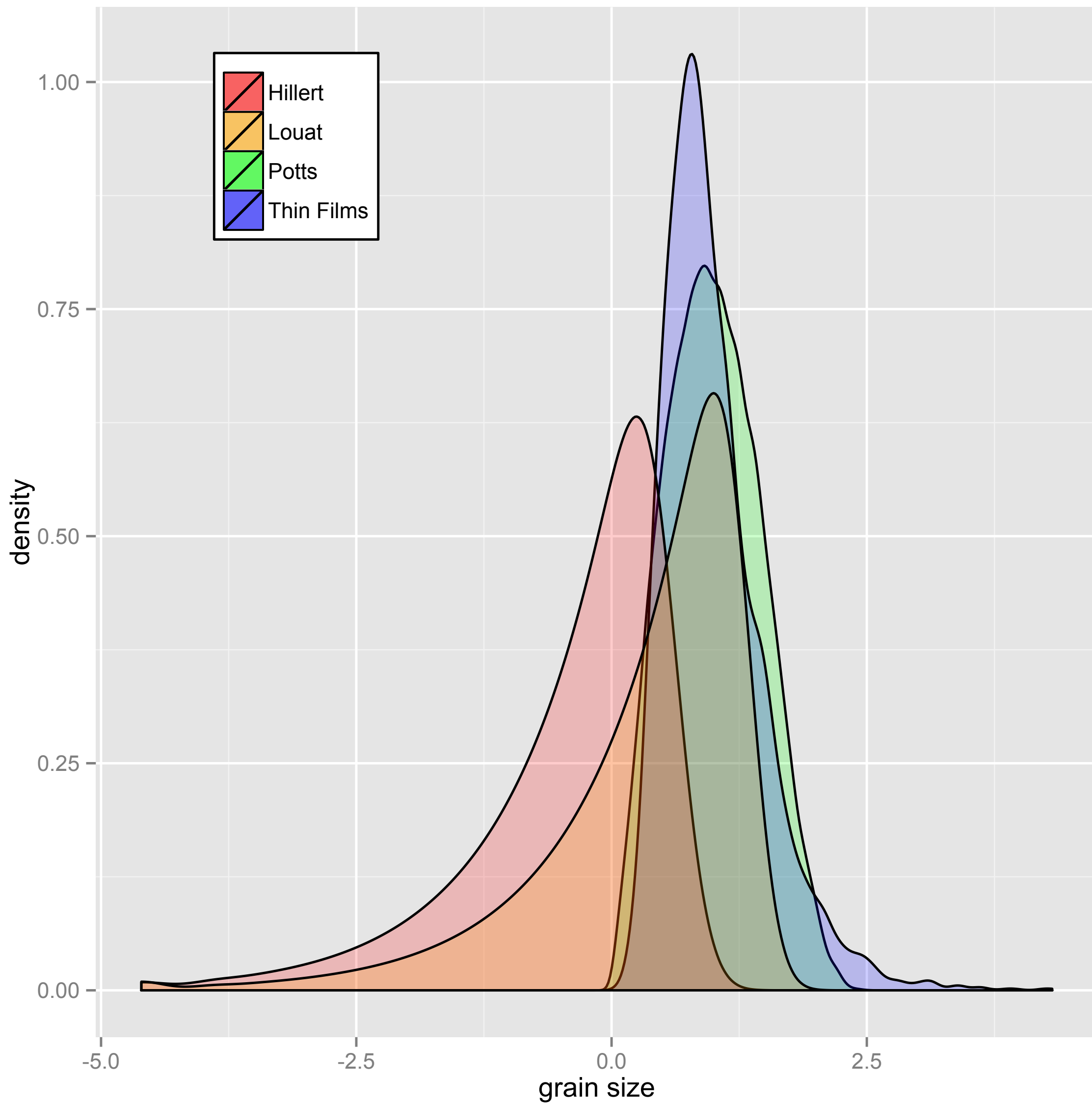
There is no (closed-form) analytical solution in 3D since we lack a well-defined n-6 rule in higher dimensions

W.W. Mullins, *Acta Mater.*, vol. 46 pp. 6219-6226 (1998)

CARNEGIE MELLON UNIVERSITY

DEPARTMENT OF
MATERIALS SCIENCE AND ENGINEERING

Are they log-normal?



Answer: not really… (they fail the standard tests)

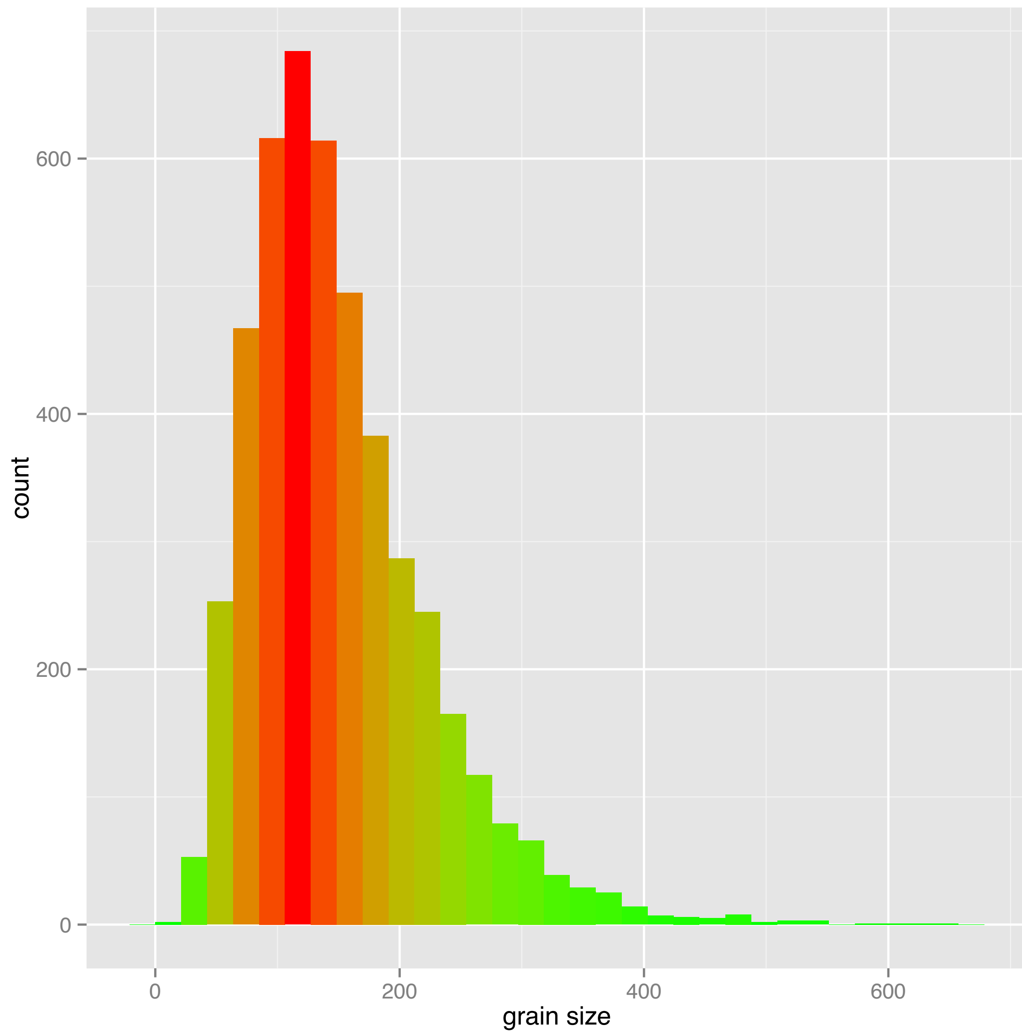
They are not really close to real grain size distributions (or simulations) either!

So where do we go from here?

CARNEGIE MELLON UNIVERSITY
DEPARTMENT OF
MATERIALS SCIENCE AND ENGINEERING

Visualizing grain size

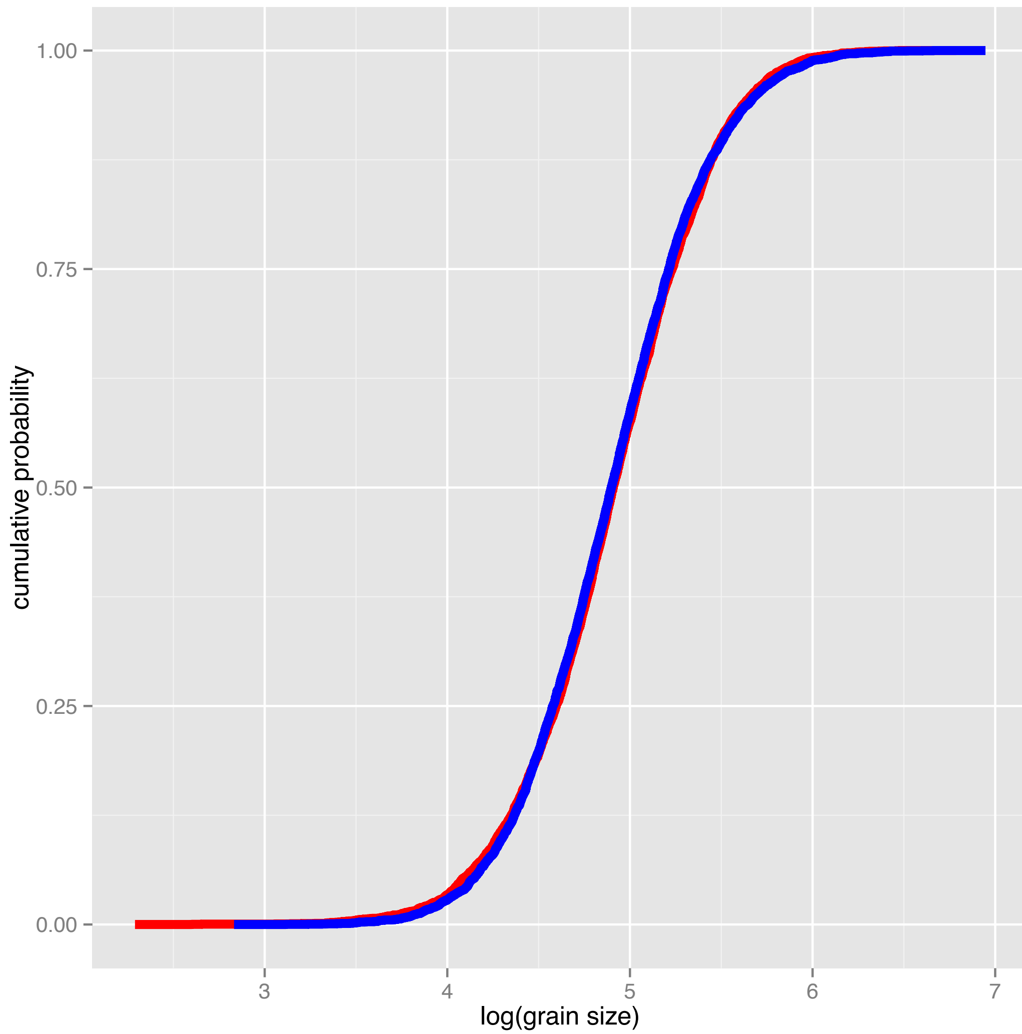
Histograms provide a way to visualize the PDF



A histogram discretizes the data by separating it into *bins* (x axis). The y axis is then the total number of data points that fall in each bin

Visualizing grain size

Empirical CDFs (eCDF) provide a way to visualize the CDF

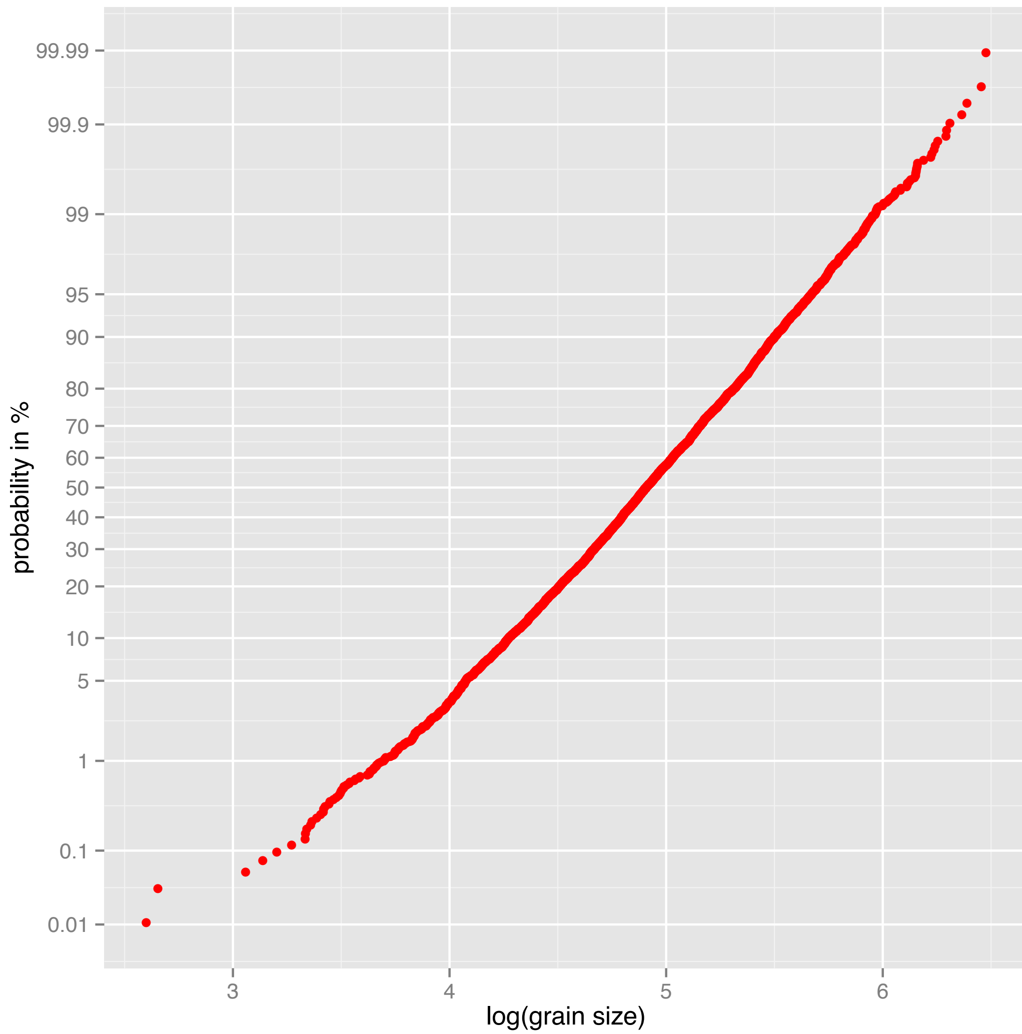


The eCDF is a *step function* that jumps by 1/x for each of the x data points

red = actual data
blue = sampled from ideal normal

Visualizing grain size

Probability plots compare empirical data to a theoretical distribution



Probability plots may have different types of axes (quantiles or probabilities)

The shape of the curve on a probability plot determines the shape of the underlying distribution

Sampling

- Actual problem: how does one sample data points from a given PDF?
- One answer: *inverse transform sampling*
- Inverse transform sampling requires knowing the *quantile function*, which is the inverse of the CDF:

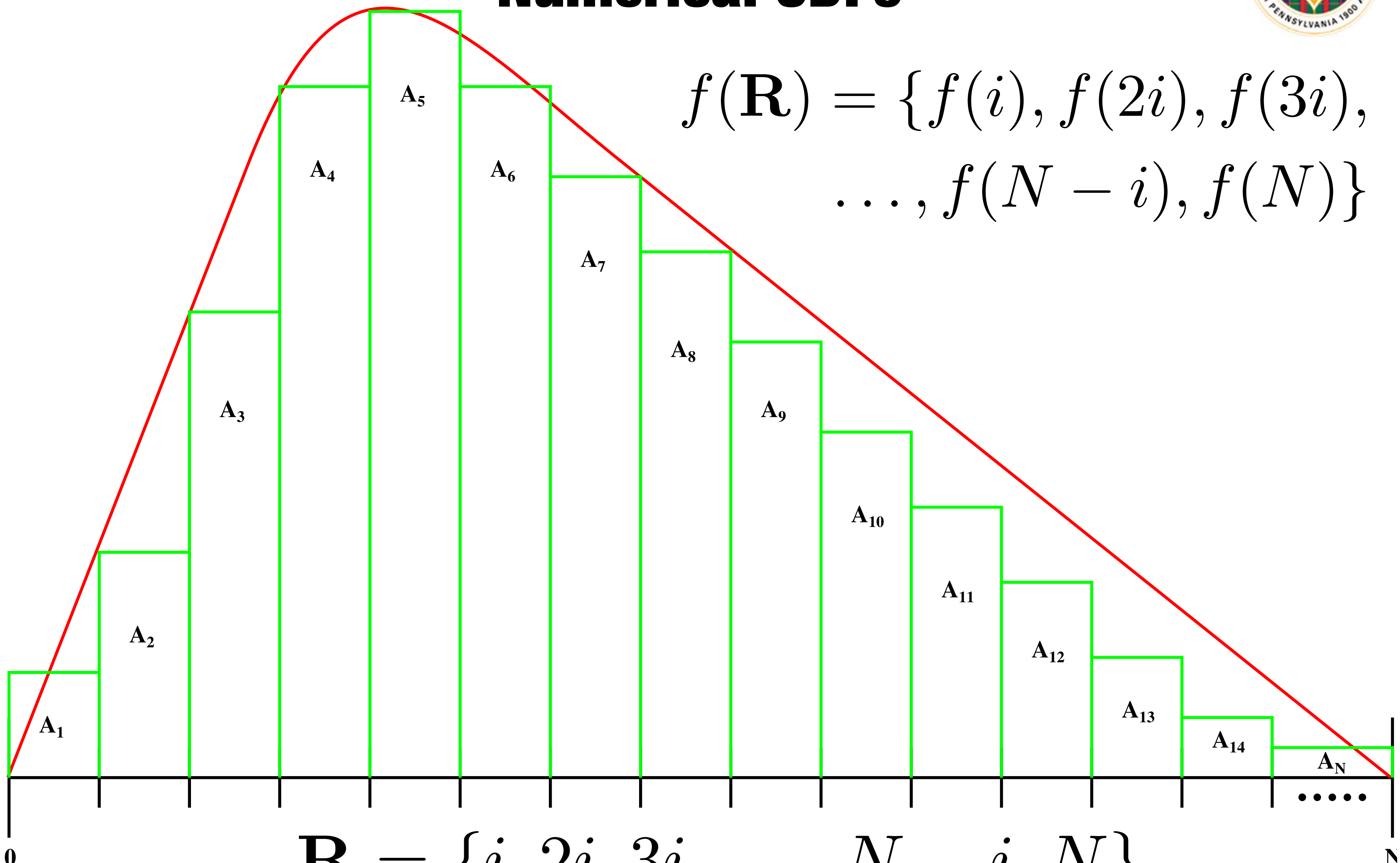
$$Q(p) = F^{-1}(x) = \inf\{x \mid F(x) \geq p, 0 < p < 1\}$$

- Unfortunately, not all CDFs can be expressed in terms of elementary functions, and thus cannot be inverted (not even the normal distribution); this is the case for the general Mullins, but not for the Hillert

CARNEGIE MELLON UNIVERSITY
DEPARTMENT OF
MATERIALS SCIENCE AND ENGINEERING

Numerical CDFs

$$f(\mathbf{R}) = \{f(i), f(2i), f(3i), \ldots, f(N-i), f(N)\}$$

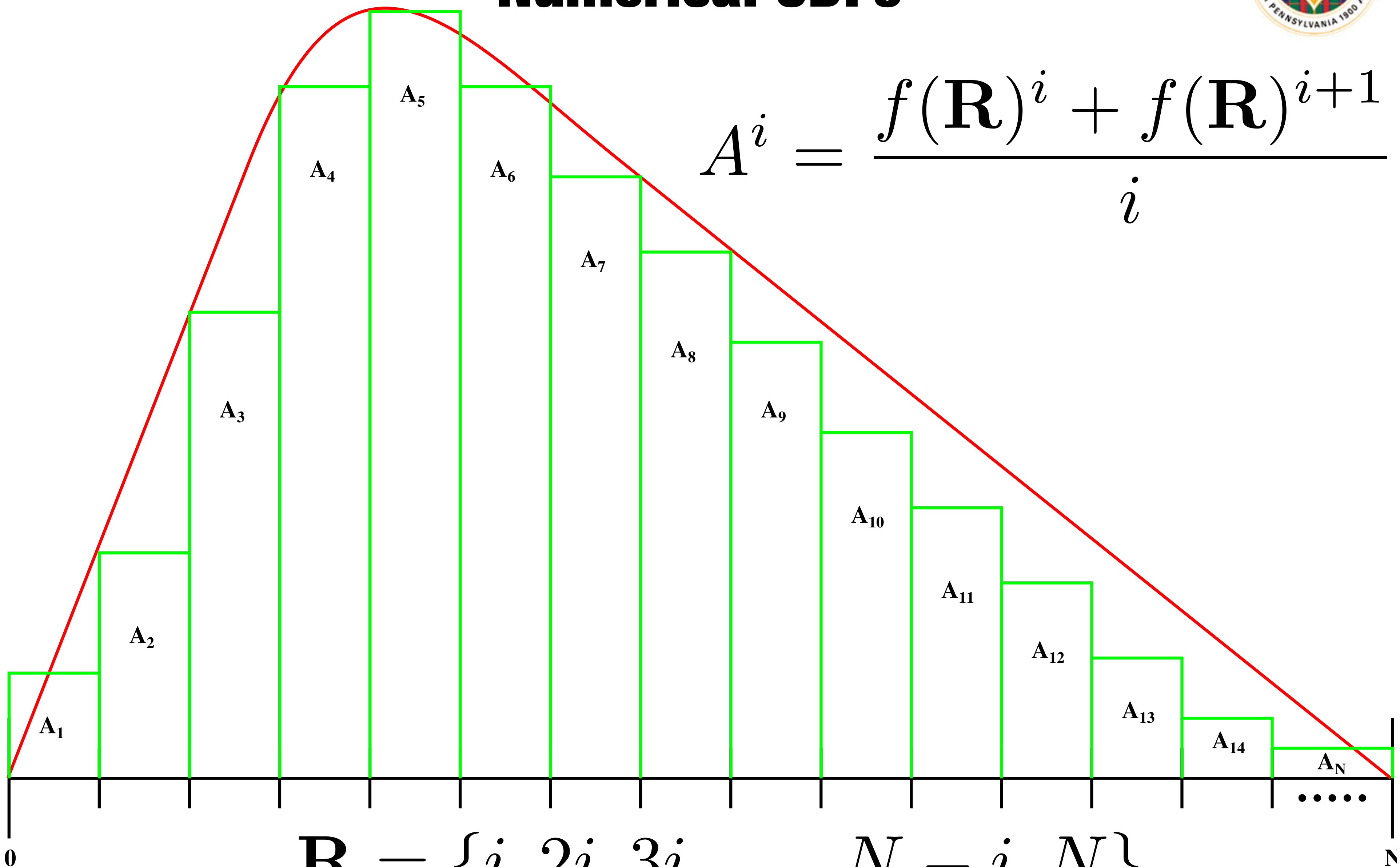


$$\mathbf{R} = \{i, 2i, 3i, \ldots, N-i, N\}$$

CARNEGIE MELLON UNIVERSITY

DEPARTMENT OF
MATERIALS SCIENCE AND ENGINEERING

Numerical CDFs



$$A^i = \frac{f(\mathbf{R})^i + f(\mathbf{R})^{i+1}}{i}$$

$$\mathbf{R} = \{i, 2i, 3i, \ldots, N-i, N\}$$

CARNEGIE MELLON UNIVERSITY

DEPARTMENT OF
MATERIALS SCIENCE AND ENGINEERING

Numerical CDFs

Construct a numerical CDF by computing the cumulative sum of the areas:

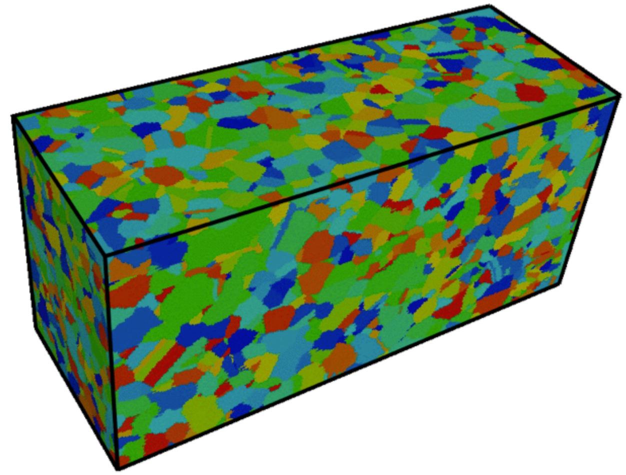
$$\mathbf{P} = \{A^i, A^i + A^{2i}, A^i + A^{2i} + A^{3i}, \ldots\}$$

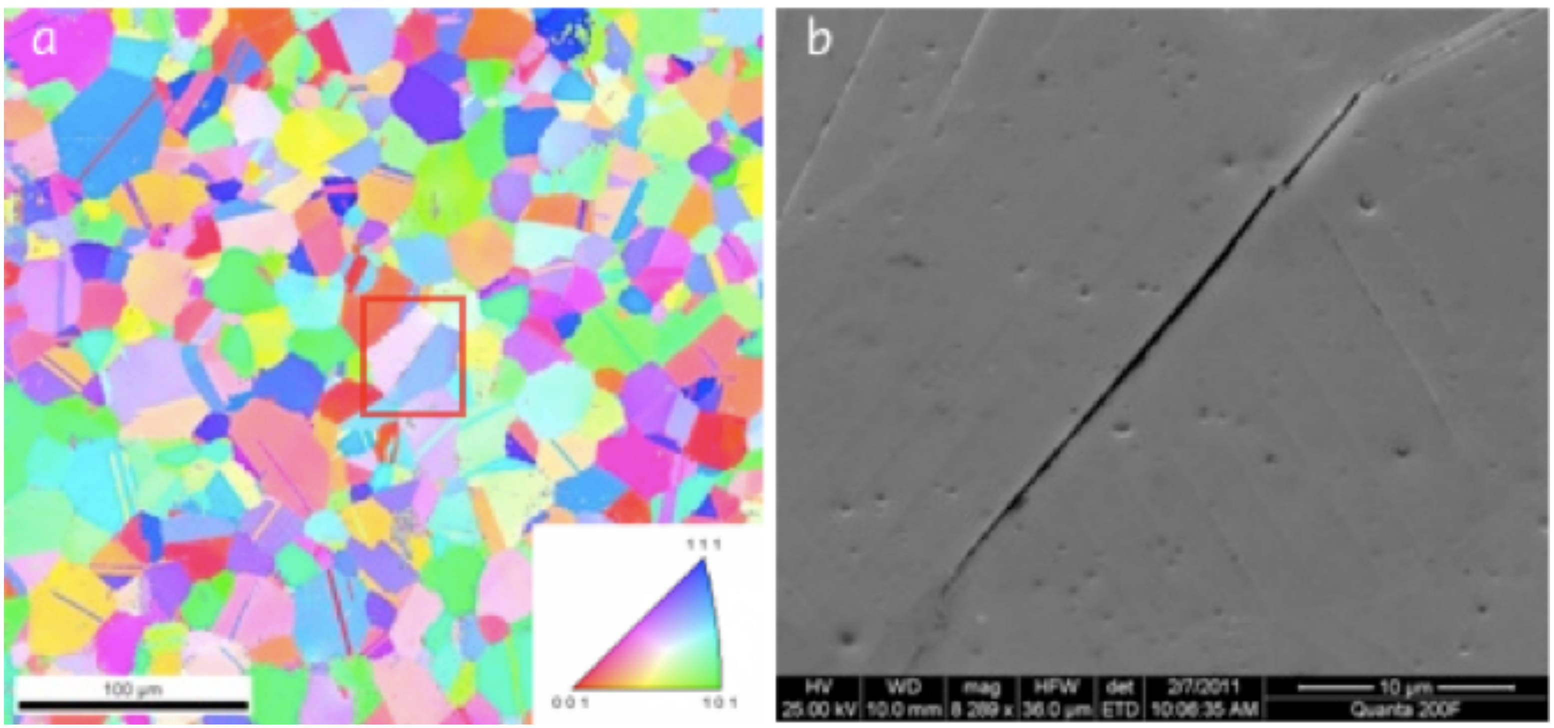
A grain size can now be sampled by finding a random (real) number, Q, on the interval [0,1], and comparing it to the set P:

$$R_{size} = \min\{\mid P^i - Q^i \mid\}$$

Extreme value theory

In some material systems, large grains ("as-large-as", or ALA) play an important role in failure, since they often serve as the nucleation site for fatigue cracks

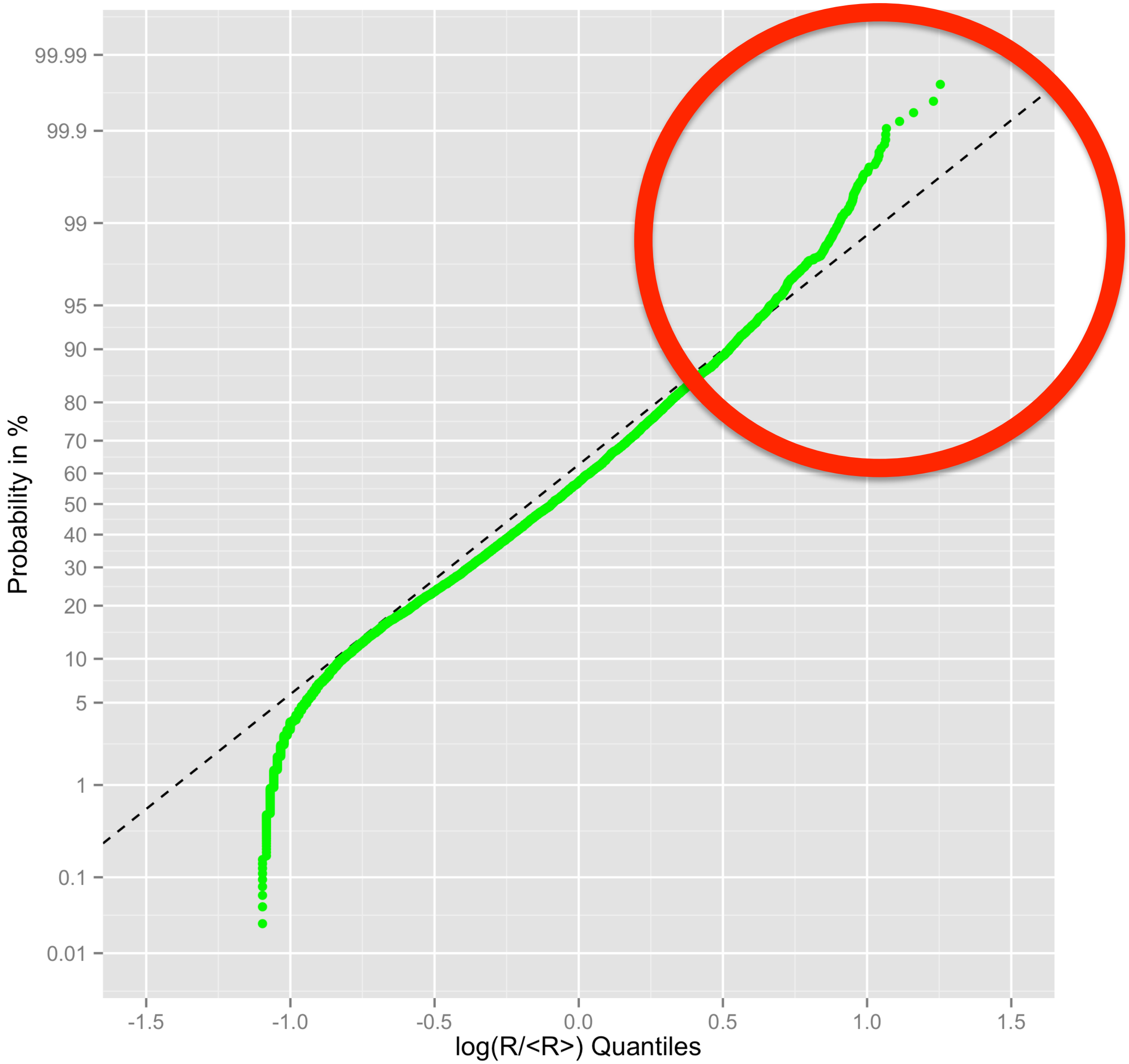




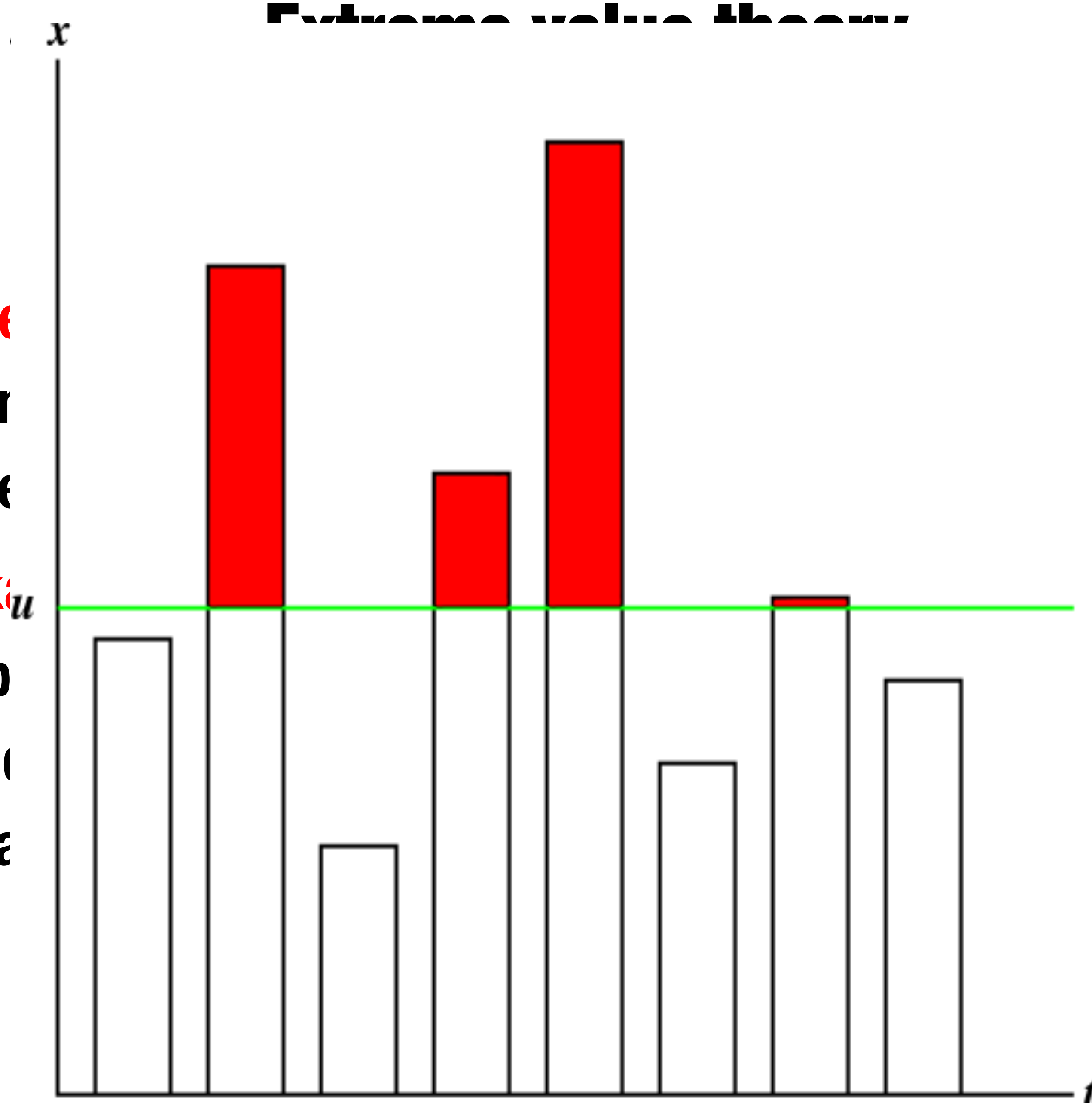
CARNEGIE MELLON UNIVERSITY
DEPARTMENT OF MATERIALS SCIENCE AND ENGINEERING

Extreme value theory

IN100



Extreme value theory



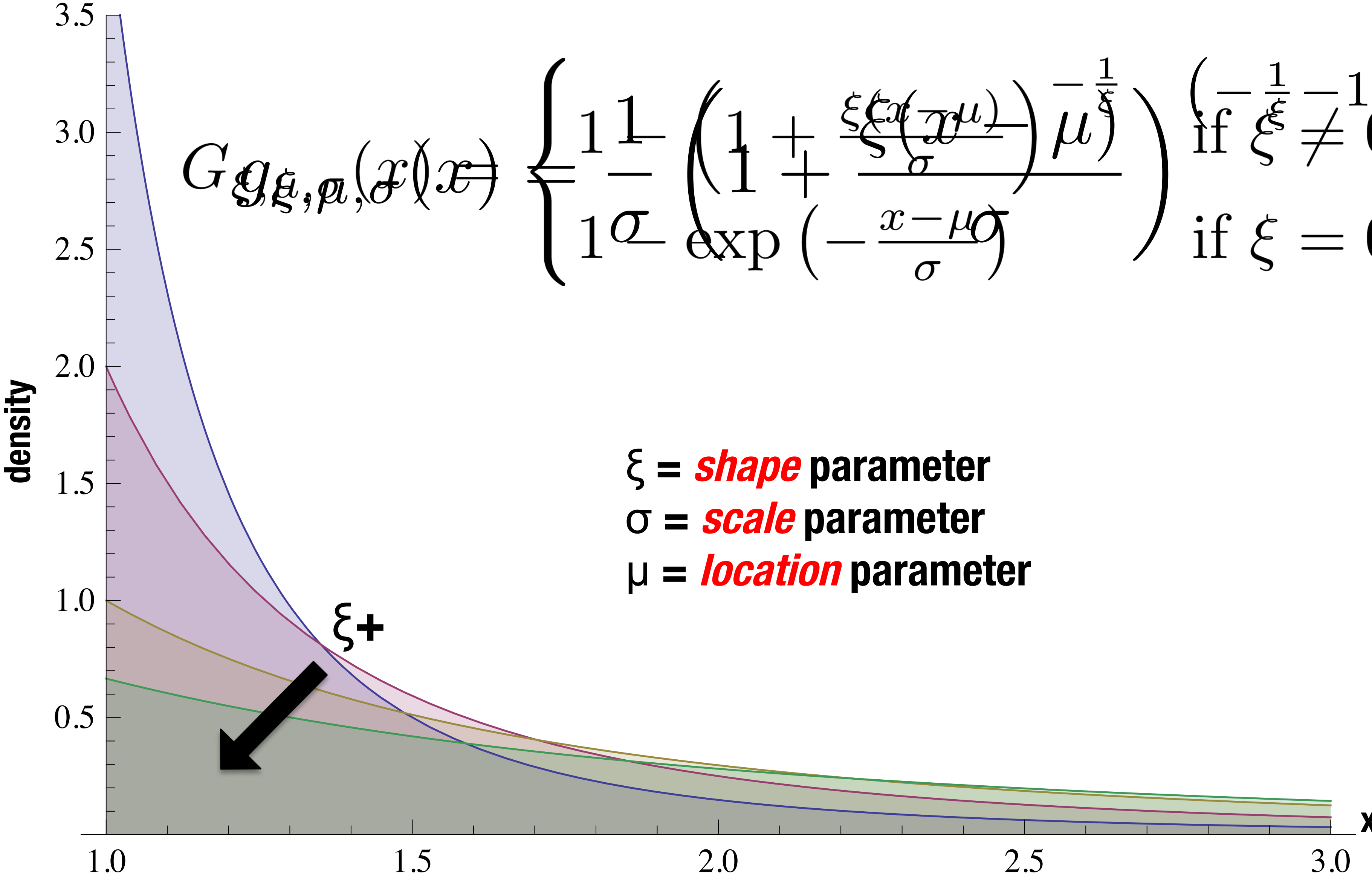
- **Fishe** **of a**
  randor **he**
  Gumbe
- **Picks** **known**
  distrib **values**
  beyon **to the**
  genera

R.A. Fisher and L.H.C. Tippett, *Mathematical Proceedings of the Cambridge Philosophical Society*, vol. 24, pp. 180-190, 1927
J. Pickands, *Annals of Statistics*, vol. 3, pp. 119-131, 1975

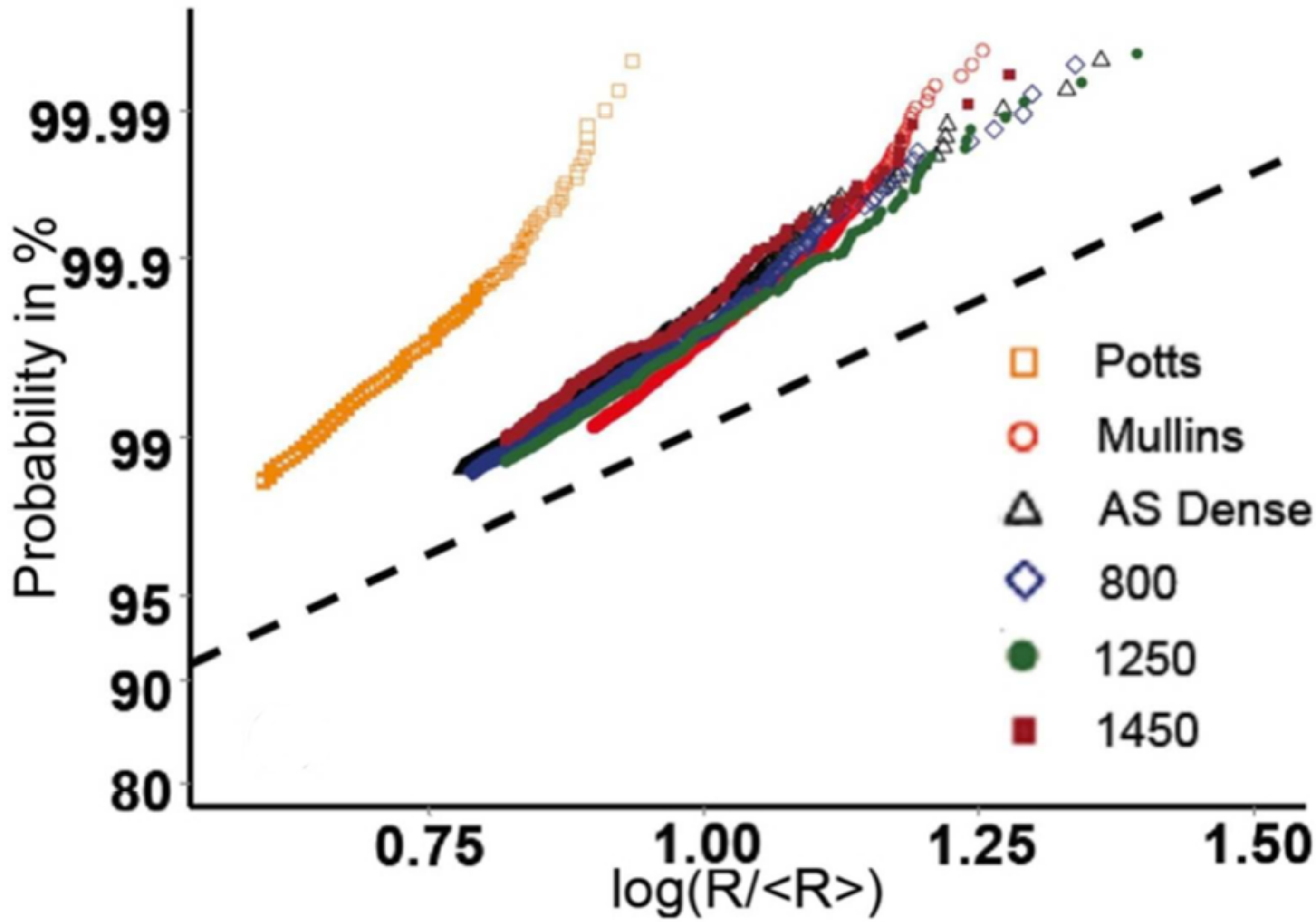
Extreme value theory

$$g_{\xi,\mu,\sigma}(x) = \begin{cases} \frac{1}{\sigma}\left(1+\frac{\xi(x-\mu)}{\sigma}\right)^{\left(-\frac{1}{\xi}-1\right)} & \text{if } \xi \neq 0 \\ \frac{1}{\sigma}\exp\left(-\frac{x-\mu}{\sigma}\right) & \text{if } \xi = 0 \end{cases}$$

ξ = ***shape*** **parameter**
σ = ***scale*** **parameter**
μ = ***location*** **parameter**



Extreme value theory



The tails of different grain size distributions can be quantitatively compared (given suitable normalization)

Q. Liu et al., *J. Amer. Cer. Soc.*, in press

Extreme value theory

- **The shapes of the upper tails of grain size distributions appears correlated to grain growth kinetics: upper tails become longer (more akin to log-normal) as the microstructure stagnates**
- **Analytical approaches to plane curve evolution indicate that the limiting (self-similar) size distribution is *uniquely* determined by the initial tail distribution**
- **"Analytical approaches" means application of *mean curvature flow* to a collection of disjoint plane curves**

Donegan et al., *Acta Mater.*, vol. 61 pp. 5595-5604 (2013)
M. Elsey, *Self-similarity in two-phase curvature flow*