Analysis of a moment-based inverse problem solution technique for breakage kernel identification

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Abstract

Breakage kernels using power-law rate kernels and self-similar daughter distributions exhibit similarity solutions. The authors have exploited this to develop a matrix-inversion technique for extracting model parameters from the moments of measured product distributions. By specifying the daughter distribution parameters and the exponent in the power law, a mapping of daughter distribution moments into product distribution moments was produced. Regression resulted in a set of linear equations giving the product moments as functions of the daughter moments and the power-law rate exponent. Given a set of measured product moments and a measured value of the power-law rate exponent, the daughter moments are obtained, and from them, the daughter distribution parameters.

In this paper, the possible sources of error in the resulting models are evaluated including (a) potentially ill-conditioned matrices in solution of the linear equation set and (b) regression error propagation. A third source of error, mismatch between the model and the actual physics, can only be assessed when one is confident that the numerical errors have been minimized. These analyses lead to an alternative method proposal. A program for developing this alternative more fully is given as the logical next step.

Introduction

The development starts from the population balance equation shown below:

$$\frac{\partial n(V,t)}{\partial t} = \Gamma_o \left[\int_V^\infty \Phi^\eta b(V;\Phi) n(\Phi,t) d\Phi - V^\eta n(V,t) \right]$$

$$b(V;\Phi) = \frac{\beta(z)}{\Phi}; \quad z \equiv \frac{V}{\Phi}$$

where the breakage rate is power-law in size and the daughters are self-similar. The source term describes breakage of parent particles of volume Φ into daughter particles of volume V. The rate is first order in the particle concentration ($n(\Phi),\Phi>V$), and proportional to the product of a size-dependent rate kernel ($\Gamma(\Phi)$) and a conditional daughter distribution ($b(V;\Phi)$) describing the fraction of daughters that are size V from a parent of size Φ. In power-law breakage the size-dependent rate kernel is assumed to take the form $\Gamma(V) = \Gamma_o V^\eta$, with η the order of the breakage rate in particle size. For self-similar daughters, the self-similar part of the daughter distribution ($\beta(z)$) only depends on the ratio of daughter-to-parent size (z). This function β is divided by the parent size to assure mass conservation when applying b. The sink term gives the breakage rate of particles of size V into all possible daughters. The parameters of the model are Γ_o, η, and any parameters needed to describe β.

In previous papers ([1], [2]), the authors proceed by:
(a) developing moment models in both physical and scaled spaces,
(b) showing that they have similarity solutions,
(c) developing a recursion expression for the scaled moments of the similarity solution in terms of the rate exponent η and certain moments of the daughter distribution, but requiring the η-order moment of the similarity solution as a seed, and
(d) developing an analytical expression for the η-order moment of the similarity solution in the cases where η is a reciprocal integer.

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Therefore, if a daughter distribution and its parameters are specified as well as the rate exponent η, the moments of the similarity solution can be recovered for any reciprocal integer value of η. These results are summarized below:

$$\text{Moment definition:} \quad M_j = \int_0^\infty V^j n(V)dV$$

$$\text{Moment scaling:} \quad \gamma_j = \frac{M_j}{M_0 V_n^j} \quad \text{with} \quad V_n \equiv \frac{M_1}{M_0}$$

$$\text{Daughter moment definition:} \quad b_j = \int_0^1 z^j \beta(z)dz$$

$$\text{Similarity solution:} \quad \gamma_{(k+1)\eta} = (p-1)^k \gamma_\eta^{k+1} \prod_{m=1}^k \left(\frac{m\eta - 1}{1 - b_{m\eta}} \right); \quad k \geq 1$$

$$\text{Seed expression:} \quad \gamma_\eta = (p-1)^{-1} \left[\prod_{m=1}^{n-1} \left(\frac{m/n - 1}{1 - b_{m/n}} \right) \right]^{-1/n}; \quad \eta = \frac{1}{n}$$

It was shown that at the similarity solution, the rate exponent η and rate coefficient Γ_o could be extracted from the slope and intercept of a ln-ln plot of measured mean particle size versus time. What remains is to discover a means for daughter distribution extraction from size distribution trajectory data.

A very general daughter distribution is based on the Hill-Ng power-law product distribution ([3]) (generalized into a beta distribution by the authors ([4])). This has been derived from a specific joint multivariate distribution for p daughters (with $p \geq 2$) as described in [3]. In multi-term form, this becomes:

$$\beta(z) = \sum_i w_i \left(\frac{q_i + r_i}{q_i} \right) \frac{z^{q_i - 1} (1-z)^{r_i - 1}}{B(q_i, r_i)} \quad \text{with} \quad p_i = \frac{q_i + r_i}{q_i}$$

where B in the denominator is the beta function. Each term has three parameters: its weight, w_i, and two exponents q_i and r_i. The mass balance constraint requires the sum of the term weights to be unity, so for an n-term distribution there are only n-1 independent weights. The number of daughters corresponding to each term (p_i) is related to the two exponents as shown above. This means that only one of the exponents (say q_i) is independent of the number of daughters. The independent exponent can be thought of as a sharpness parameter. As discussed in [1] and [2], small values of q_i correspond to mechanisms that generate daughters of disparate size (erosion, or fracture with fines) while large values represent mechanisms generating daughters of near equal size (cleavage or fragmentation). As q_i tends to infinity, the daughters approach identical size.

The moments of this daughter distribution are given by:

$$b_j = \sum_i w_i \left(\frac{q_i + r_i}{q_i} \right) \frac{B(q_i + j, r_i)}{B(q_i, r_i)}$$

In [1], a single term daughter distribution was studied (i.e., $w_1 = 1$, $q_1 = q$, $p_1 = p$). The analytical similarity solution given above was used to generate a large data set of product moments (γ_j) as a function of η, p and q. It was observed that for small q, the product moments were large and nearly independent of p while for large q they were small and nearly independent of q. It was hoped that a single moment (say γ_2) could be used to discriminate regimes, and that regime-dependent relations could be developed expressing that moment in terms of η and either p or q, with the other parameter arbitrarily set. Thus, at small γ_2, q would

be set to 10 while at small γ_2, p would be set to 2. This program was carried out as described in [1], and it was shown that a forward solution of the population balance problem using the quadrature method of moments (see [5] for a discussion of this method) reproduced the size and 2nd moment trajectories from which the model parameters were derived. Unfortunately, when the product distributions were reconstructed (following the method of the authors given in [6], [7]), the reconstructed distributions were not in good agreement with the data. It was concluded that greater detail was needed to describe the daughter distribution in order to capture the full system behavior.

In [2], the daughter distribution was expanded to a two-term form where the 1st term was of Peterson ([8]) form ($r_1=1$) and the 2nd term was of the general form:

$$\beta(z) = (1-w)\left(\frac{q_1+1}{q_1}\right)\frac{z^{q_1-1}}{B(q_1,1)} + w\left(\frac{q_2+r_2}{q_2}\right)\frac{z^{q_2-1}(1-z)^{r_2-1}}{B(q_2,r_2)}$$

The form of the similarity solution suggested that the following relationship between the daughter distribution moments and the similarity solution moments would be a good fit:

$$\ln \gamma_j = \begin{aligned} &A_j + \frac{C_j}{\eta} + \left(a_{0,j} + \frac{c_{0,j}}{\eta}\right)\ln\left(\left|b_0 - 1\right|\right) + \left(a_{1/3,j} + \frac{c_{1/3,j}}{\eta}\right)\ln\left(\left|b_{1/3} - 1\right|\right) \\ &+ \left(a_{2/3,j} + \frac{c_{2/3,j}}{\eta}\right)\ln\left(\left|b_{2/3} - 1\right|\right) + \left(a_{4/3,j} + \frac{c_{4/3,j}}{\eta}\right)\ln\left(\left|b_{4/3} - 1\right|\right) \end{aligned}$$

Again, a large set of values was generated using the analytical solution for reciprocal integer η. The coefficients in equations for the 1/3, 4/3, 5/3 and 2nd moments were obtained by multilinear regression. The resulting correlation coefficients were near unity in all cases, indicating excellent fits.

After first assuring the self-similarity had been reached, experimental similarity solution moments were obtained by fitting cumulative mass data to an expansion of the distribution function as a two-term modified gamma distribution as described in [1] and [2]. The values of Γ_o and η were obtained by the appropriate ln-ln plot of the mean size trajectory after reaching self-similarity. The daughter distribution moments in the above equation were then obtained by matrix inversion. The parameters of the daughter distribution were found via an interactive Newton-Raphson procedure. This completed solution of the inverse problem.

The matrix inversion process may be described as:

$$\mathbf{Ax = b} \quad \Rightarrow \quad \mathbf{x = A^{-1}b}$$

$$\mathbf{A} = \begin{bmatrix} a_{0,1/3}+\dfrac{c_{0,1/3}}{\eta} & a_{1/3,1/3}+\dfrac{c_{1/3,1/3}}{\eta} & a_{2/3,1/3}+\dfrac{c_{2/3,1/3}}{\eta} & a_{4/3,1/3}+\dfrac{c_{4/3,1/3}}{\eta} \\[2ex] a_{0,4/3}+\dfrac{c_{0,4/3}}{\eta} & a_{1/3,4/3}+\dfrac{c_{1/3,4/3}}{\eta} & . & . \\[2ex] a_{0,5/3}+\dfrac{c_{0,5/3}}{\eta} & . & a_{2/3,5/3}+\dfrac{c_{2/3,5/3}}{\eta} & . \\[2ex] a_{0,2}+\dfrac{c_{0,2}}{\eta} & . & . & a_{4/3,2}+\dfrac{c_{4/3,2}}{\eta} \end{bmatrix}$$

$$\mathbf{x} = \begin{bmatrix} \ln\left(\left|b_0 - 1\right|\right) \\ \ln\left(\left|b_{1/3} - 1\right|\right) \\ \ln\left(\left|b_{2/3} - 1\right|\right) \\ \ln\left(\left|b_{4/3} - 1\right|\right) \end{bmatrix} \qquad \mathbf{b} = \begin{bmatrix} \ln\gamma_{1/3} - A_{1/3} - \dfrac{C_{1/3}}{\eta} \\ \ln\gamma_{4/3} - A_{4/3} - \dfrac{C_{4/3}}{\eta} \\ \ln\gamma_{5/3} - A_{5/3} - \dfrac{C_{5/3}}{\eta} \\ \ln\gamma_2 - A_2 - \dfrac{C_2}{\eta} \end{bmatrix}$$

Reference [2] also describes initialization and solution of the forward problem by the quadrature method of moments and the reconstruction of the predicted product distributions.

The results with the more complex 4-parameter daughter distribution were improved. Again, the data used in solving the inverse problem (mean size time trajectory and product (similarity solution) moment trajectories for the 1/3, 4/3, 5/3 and 2nd scaled moments) were well represented by the model parameters. The reconstructed distributions were much closer to those measured. Five data sets were given the full treatment of inverse problem solution followed by working the forward problem and reconstructing distributions from the model-predicted moments.

There were five types of error evaluated to describe how well or poorly the technique worked. The first of these was the root-mean-square error in representing the cumulative mass data points at self-similarity by the two-term modified gamma distribution. These ranged from 0.21% to 3.02%. The next was the error in representing the daughter moments obtained from the regression by a set of daughter distribution parameters. In only one case were all four parameters obtainable exactly with no error. In all other cases, only a subset of the daughter moments could be represented exactly with the remainder chosen to minimize the root-mean-square error. This error was generally less than 10%, but in one case, was as large as 70%. The next type of error was a comparison of the moments predicted by the forward problem solution compared to the data. This root-mean-square error ranged from 5-20% as did the moments calculated from the reconstructed two-term modified gamma distributions as compared to the data. Finally, the root-mean-square error for direct comparison of the reconstructed distributions to the data points ranged from 0.90 to 3.63%.

These different types of error were not necessarily correlated. There was one case that had the smallest error of every type. However, the case with large error in representing the daughter moments was mid-range in the error between the forward solution moments and those obtained from the data. Then again, the reconstruction had one of the two highest errors in comparison to the individual data points.

The most disappointing aspect of this is the inability to retrieve daughter moments in all cases that can be well represented by a set of daughter distribution parameters. It was this observation that motivated the work described in the rest of this paper.

Singular Value Decomposition Analysis

One possible source of error in this technique is the possibility that the **A** matrix being inverted is ill conditioned. This can be evaluated by obtaining the eigenvalues of the square matrix formed from $\mathbf{A^T A}$, and computing its condition number as the square root of the ratio of largest to smallest non-zero eigenvalues. Large values of the condition number indicate ill-conditioning and suggest that the dimension is too large for reliable solution by inversion. One can eliminate the smallest eigenvalue and calculate the condition number for a matrix reduced in dimension by one row and column. This process can be repeated until the condition number is sufficiently small to reliably solve the system. In this case, the dimension of such a reduced matrix indicates the maximum number of daughter moments that can be reliably obtained.

Since the coefficients are functions of η, Figure 1 shows the eigenvalues of $\mathbf{A^T A}$ as a function of η. Figure 2 gives the condition number of the full 4x4, a reduced 3x3 and a further reduced 2x2 matrix. The values at η for the five data sets are shown as points in the graphs.
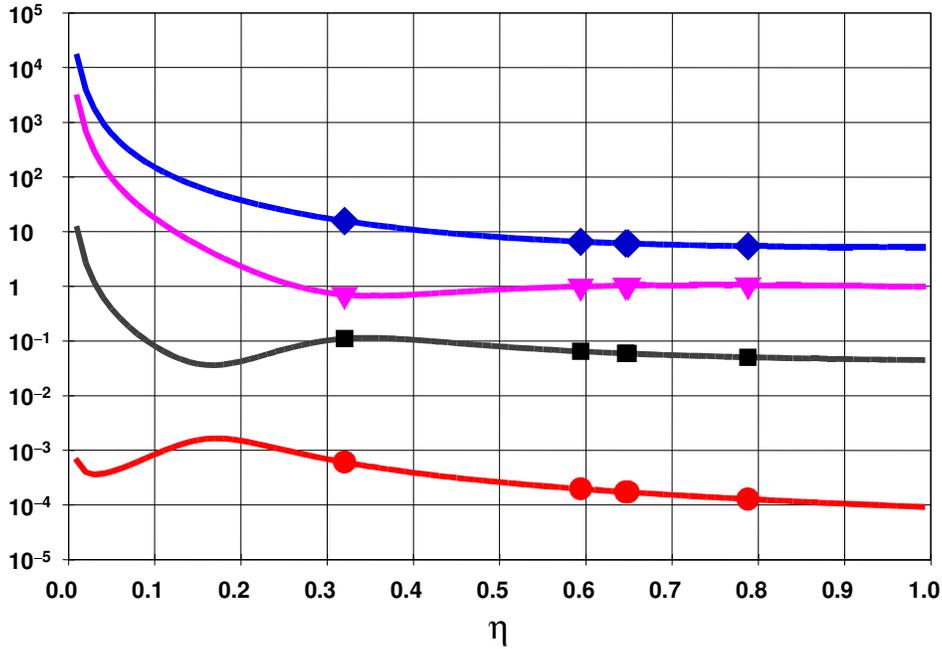
Figure 1 – Eigenvalues of $\mathbf{A^T A}$
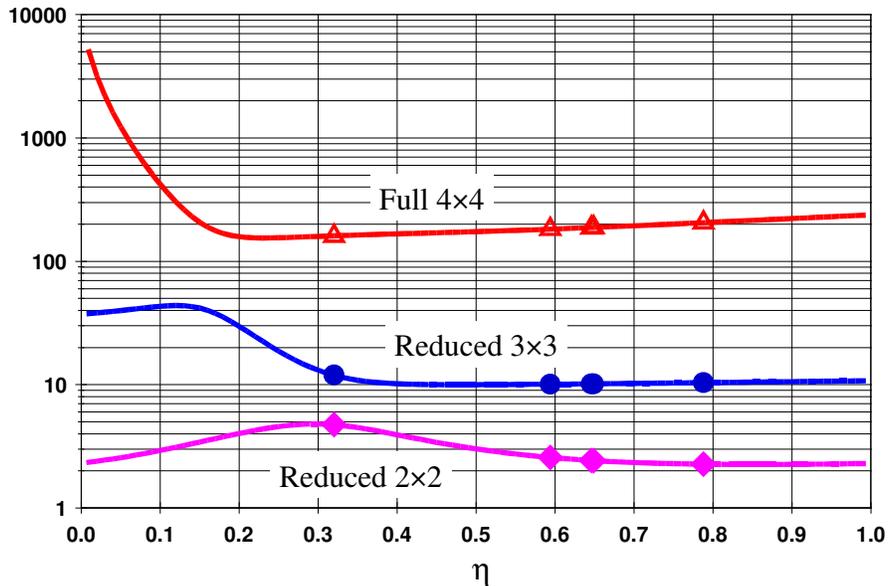
Figure 2 – Condition Number of $\mathbf{A}$

What these results show is that the full 4x4 matrix is ill-conditioned, but that the reduced 3x3 matrix is suitably conditioned to allow solution. This means that successfully extracting 4 daughter distribution moments by the matrix inversion is an iffy proposition and gives some clue as to why it was generally not possible to fully represent all 4 daughter moments with daughter distribution parameters.

Propagation of Regression Error

Another source of error is propagation of errors in the regression itself. In general, correlation coefficients were so close to unity that one would not expect large errors to be present. But the relations that were fit cover many decades of values for the similarity solution moments so even small error bands relative to the slope of the correlation line can translate into large relative errors in the moment values themselves. To test this notion, several cases were evaluated in which the true values of both the similarity solution and the daughter distribution moments were known. The table below shows the results:

System Specification	Case 1	Case 2	Case 3
η	.3333	.3333	1
q_1	.01	.005	1
w	.5	.2	1
r_2	3	1000	1
p_2	4	15000	2
True Values			
$\ln(p-1)$	3.941582	8.058264	0.000000
$\ln(b_{1/3}-1)$	0.487362	3.857011	−0.693147
$\ln(b_{2/3}-1)$	−0.728424	1.023389	−1.609438
$\ln(b_{4/3}-1)$	−1.316673	−0.965268	−1.945910
$\ln(\gamma_{1/3}-1)$	−2.206716	−3.244017	−0.113192
$\ln(\gamma_{4/3}-1)$	1.698808	3.993498	0.174490
$\ln(\gamma_{5/3}-1)$	3.651734	8.674401	0.408511
$\ln(\gamma_2-1)$	5.811161	13.742609	0.693147
Values via Matrix Inversion			
$\ln(p-1)$	3.862251	8.113482	0.293293
$\ln(b_{1/3}-1)$	0.390977	3.891068	−0.456786
$\ln(b_{2/3}-1)$	−0.822750	1.080657	−1.373776
$\ln(b_{4/3}-1)$	−1.413516	−0.925507	−1.720174
$\varepsilon_{rms}(\%)$ vs. true values	9.6	4.9	28.4
Values via Newton-Raphson			
q_1	.006867	.006918	1.000
w	.691273	.225308	1.000
r_2	1.387983	866.766	1.297
p_2	4.781224	14324.5	2.341
$\varepsilon_{rms}(\%)$ in representing fit	0.0	0.0	1.8

This table shows that even when all the daughter moments obtained from the matrix inversion can be represented exactly by daughter distribution parameters, the error propagation from the regression through the matrix inversion can be significant, and even as little as 1.8% error in representing the matrix inversion solution can correspond to 30% error against the true daughter moments. This argues that another approach is necessary, one that avoids regression entirely if possible.

Interpolation of the Analytical Solution

Prior to abandoning regression entirely, a search for relations that might incur reduced regression error was begun. The analytical expression for the similarity solution was the point of departure. In the course of manipulating this expression, it was realized that one could develop a revised expression for η a reciprocal integer as follows:

$$\phi_{j\eta} \equiv \gamma_{j\eta}\gamma_{\eta}^{(1-j\eta)/\eta} = \begin{cases} \displaystyle\prod_{m=j}^{1/\eta-1} \rho_{m\eta}^{-1} & ; \quad 1 \le j \le \dfrac{1}{\eta} \\[12pt] 1 & ; \quad j = \dfrac{1}{\eta} \qquad \text{for } \eta = \dfrac{1}{n} \\[12pt] \displaystyle\prod_{m=1/\eta}^{j-1} \rho_{m\eta} & ; \quad \dfrac{1}{\eta} < j \end{cases}$$

with:

$$\rho_k = (p-1)\left(\frac{k-1}{1-b_k}\right)$$

Everything on the LHS can be calculated directly from measurements of size and distribution trajectories. Everything on the RHS is a function of the daughter distribution parameters. If the value of η obtained from the data is a reciprocal integer, then the relation is exact as stated. The issue becomes what to do when η lies between reciprocal integers. Noting that:

$$\phi_\eta = \gamma_\eta^{1/\eta}$$

one can rearrange the above to give:

$$\gamma_{j\eta} = \phi_{j\eta}\phi_\eta^{j\eta-1}$$

If one treats these expressions as functions of η, then one can imagine finding their values by interpolating the known solutions at the reciprocal integer points. An alternative attack on the inverse problem would then be to search a space of daughter distribution parameters until one minimized the error in representing a set of $\gamma_{j\eta}$ values. One exchanges the interpolation error for the regression error in the previous approach and hopes that it turns out to be smaller.

Figure 3 shows a graph of $\ln \phi_{j\eta}$ versus $1/\eta$ showing that in this space, the relations are nearly linear. This suggests that polynomial interpolation should do a reasonably good job of representing the similarity solution in these coordinates.

The following program has been developed to characterize the error associated with this approach. One needs some kind of standard for assessing how close the interpolation comes to the "true" value. The forward solution of a moment model offers one possibility. So, first it must be validated against the known analytical solutions. In the model, the restriction of the 1st term of the daughter distribution to be Peterson type has been relaxed so that both terms are of the general power-law product type. Thus, there are 5 daughter distribution parameters. In validating the forward solution, there are several questions to be asked:

- Is the error sensitive to the moments that are the "subjects" of the quadrature method? The subject moments are those upon which the model equations are based. These should include the targets to be used for obtaining daughter parameters, i.e., orders that are multiples of η, but will probably include additional moments. How should these be chosen?
- Is the error sensitive to method? One could use an alternative method also discussed in [5] known as MOMIC or in this case, MOM*EC* (Method of Moments with *Extrapolative* Closure) rather than QMOM (Quadrature Method of Moments).

A set of cases for various reciprocal integer values of η and various sets of daughter parameters must be calculated. Provided one can arrive at a rationale for choosing the closure method and the selection of "subject" moments such that the forward solution predicts the known values to within small error, the next step is to validate the interpolation.

To validate the interpolation, the best rendering of the forward solution would be used to calculate for a set of η that lie between the reciprocal integers so that comparison to the interpolation can be made. As a final test, raw data sets should be developed from known solutions but with random error superimposed to test the ability of the method to represent the data to within the tolerance of the error in the data itself.

Execution of this program is now underway.
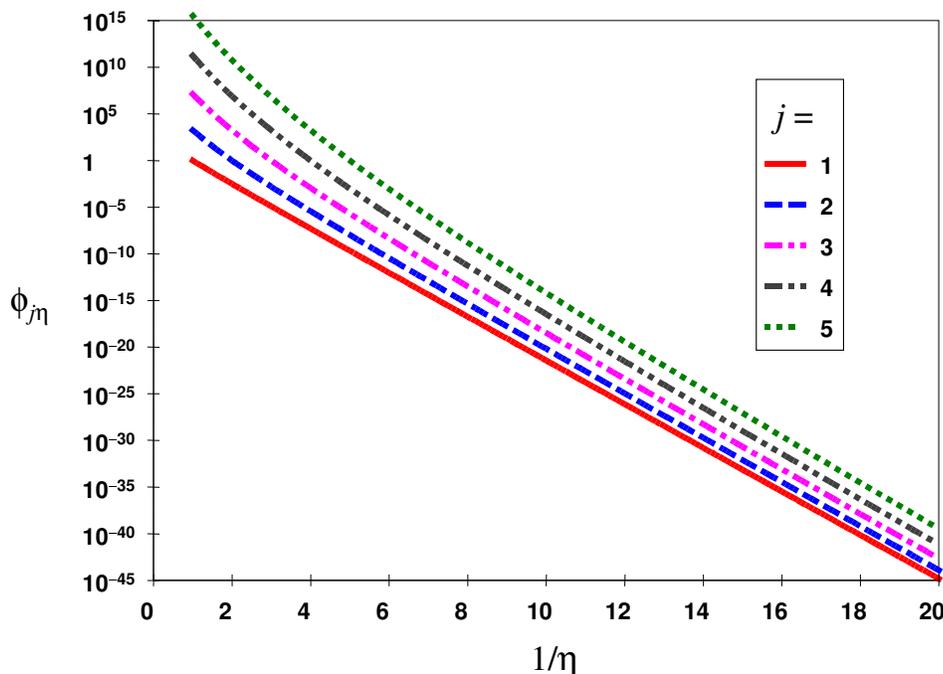
Figure 3 – Similarity Solution in Nearly Linear Space

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