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Statistics for estimating the population average of a Lifshitz–Slyozov–Wagner (LSW) distribution

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Abstract A generic version of the Lifshitz–Slyozov–Wagner (LSW) distribution is introduced. Using this generic form, a maximum likelihood (ML) estimator for the population average has been developed. The statistical properties of the estimates obtained by the ML method, as well as the conventional sample average method, have been assessed by Monte Carlo simulations for seven sample sizes ranging from 10 to 1000. Results showed that (i) both estimators yield practically unbiased results, (ii) the standard deviation of estimates obtained by the ML method is significantly less than that of the sample averages, (iii) the distribution of estimates is neither normal, lognormal nor 2-parameter Weibull. Percentage points of the distribution of estimates for both methods have been developed. The use of these points for calculating confidence limits for the population average of the LSW distribution is demonstrated by examples in this article.

Introduction and background

It is of considerable interest to model the evolution of the size distribution of a dispersed second-phase particles during heat treatment. An important part of modeling efforts should concentrate on diffusion-controlled coarsening of the second-phase particles, which has drawn considerable attention. Consequently, an extensive body of theoretical as well as experimental results has been produced, as outlined previously in extensive reviews by Ratke and Voorhees [1], Doherty [2], Wang and Glicksman [3], and Baldan [4].

One of the most important contributions to the field of coarsening was made jointly by Lifshitz and Slyozov [5] and by Wagner [6], who showed theoretically that:

1. The average size of particles (μ) increases with time (t) following;
$$\mu^3 - \mu_0^3 = kt \tag{1}$$
where μ_0 is the initial average diameter and k is a temperature dependent constant.
2. Eventually, the distribution of particle size (x) reaches steady state and follows the Lifshitz–Slyozov–Wagner (LSW) distribution, the density function (f) of which is written as:
$$f\left(\frac{x}{\mu}\right) = \frac{4}{9}\left(\frac{x}{\mu}\right)^2 \left(\frac{3}{3+\frac{x}{\mu}}\right)^{7/3} \left(\frac{1.5}{1.5-\frac{x}{\mu}}\right)^{11/3} \exp\left(\frac{\frac{x}{\mu}}{\frac{x}{\mu}-1.5}\right); \quad 0 < \frac{x}{\mu} < 1.5$$
$$f\left(\frac{x}{\mu}\right) = 0; \quad \frac{x}{\mu} \geq 1.5 \tag{2}$$

Note in Eq. 2 that the probability density function is for the normalized particle size, i.e., x/μ, and μ is the only parameter to be estimated. In addition, Eq. 2 includes a cutoff at $x/\mu = 1.5$, i.e., no particle is expected to have a normalized size larger than 1.5. The probability density (f)

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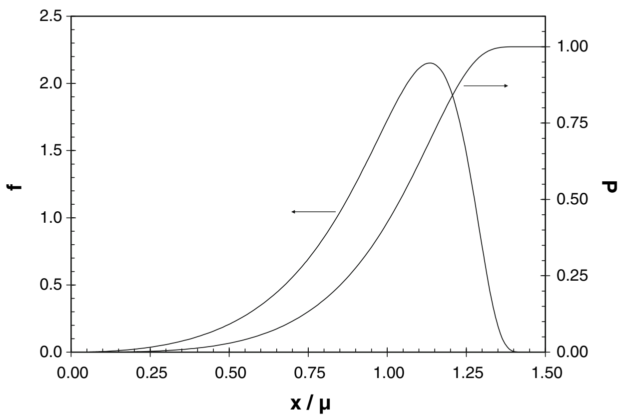

Fig. 1 The probability density (*f*) and cumulative probability (*P*) functions of the LSW distribution plotted versus x/μ

and cumulative probability (*P*) functions of the LSW distribution are presented in Fig. 1.

There have been many attempts to apply the LSW distribution by comparison with experimental size distributions when the evolution of particle size during coarsening was found to follow Eq. 1. To apply the LSW distribution, the researchers had to (i) take sample average as the estimate for μ, (ii) superpose the LSW probability density function (Eq. 2) on the histograms for normalized particle size as a measure of goodness-of-fit. Results in most studies showed that the actual size distribution was somewhat wider and did not have the sharp cutoff of the LSW distribution [2, 7].

$$L(\mu;x_1,\ldots,x_n)=\prod_{i=1}^{n}\left(\frac{4}{9\mu}\right)\left(\frac{x_i}{\mu}\right)^2\left(\frac{3}{3+\left(\frac{x_i}{\mu}\right)}\right)^{7/3}\left(\frac{1.5}{1.5-\left(\frac{x_i}{\mu}\right)}\right)^{11/3}\exp\left(\frac{\frac{x_i}{\mu}}{\frac{x_i}{\mu}-1.5}\right) \tag{5}$$

Consequently, modifications were made to the LSW coarsening theory which yielded particle size distributions different from the LSW distribution. Most notable are the size distributions proposed by Ardell [8], Brailsford and Wynbladt [9], Enomoto et al. [10], Davies et al. [11], Tsumuraya and Miyata [12], and Marqusee and Rose [13]. This article is not intended to address the coarsening theory but the statistics of the LSW distribution. It is the authors' observation that in all the studies where LSW distribution was used, population average was estimated from the sample average and more statistically sound techniques, such as the maximum likelihood (ML) method, have not been developed for the LSW distribution. An ML estimator is first developed for the LSW distribution. Then by using Monte Carlo simulation results, the two methods of estimating μ, namely taking sample average and using the ML estimator, are compared for the LSW distribution. ML estimators for improved particle size distribution as well as formal goodness-of-fit techniques will be addresses in future studies.

ML estimator of μ

For the ML estimator to be generated, the probability density function, *f*, should be a function of *x* only and not the normalized form given in Eq. 2. The generic (or general) form of the LSW probability density function can be written as:

$$f(x;\mu)=\frac{4}{9\mu}\left(\frac{x}{\mu}\right)^2\left(\frac{3}{3+\frac{x}{\mu}}\right)^{7/3}\left(\frac{1.5}{1.5-\frac{x}{\mu}}\right)^{11/3}\exp\left(\frac{\frac{x}{\mu}}{\frac{x}{\mu}-1.5}\right);\quad 0<\frac{x}{\mu}<1.5$$
$$f(x;\mu)=0;\quad \frac{x}{\mu}>1.5 \tag{3}$$

Equation 2 can now be referred to as the standard LSW distribution with $\mu=1$. The derivation of Eq. 2 is provided in the Appendix. The likelihood function, *L*, is written as

$$L(\mu;x_1,\ldots,x_n)=\prod_{i=1}^{n}f(x_i;\mu) \tag{4}$$

where *n* is the sample size. Hence for the LSW distribution, the likelihood function is

The value of μ which maximizes $L(\mu;x_1,\ldots,x_n)$ is the ML estimate of μ. To find this value, we first compute $M=\log(L)$;

$$M=n\left[\log\left(\frac{4}{9}\right)+\frac{7}{3}\log(3)+\frac{11}{3}\log(1.5)-\log(\mu)\right]+2\sum_{i=1}^{n}\log\left(\frac{x_i}{\mu}\right)-\frac{7}{3}\sum_{i=1}^{n}\log\left(3+\frac{x_i}{\mu}\right)-\frac{11}{3}\sum_{i=1}^{n}\log\left(1.5-\frac{x_i}{\mu}\right)+\sum_{i=1}^{n}\frac{x_i}{x_i-1.5\mu} \tag{6}$$

We then differentiate *M* with respect to μ and solve $dM/d\mu=0$:

$$-3n+\frac{7}{3}\sum_{i=1}^{n}\frac{x_i}{3\mu+x_i}-\frac{11}{3}\sum_{i=1}^{n}\frac{x_i}{1.5\mu-x_i}+1.5\sum_{i=1}^{n}\frac{x_i\mu}{(x_i-1.5\mu)^2}=0 \tag{7}$$

The solution of this equation for μ is the ML estimator for the LSW distribution, and will be denoted by $\tilde{\mu}$. We will compare $\tilde{\mu}$ with sample average, $\bar{x}$, to determine the statistical characteristics of the ML and sample average estimators for various sample sizes.

Research methodology

Monte Carlo simulations were run using the Mathematica software to generate n data points from an LSW distribution with $\mu = 1$. Seven sample sizes (n), namely 10, 25, 50, 100, 250, 500, and 1000, were investigated. For each sample, the sample average was calculated. In addition, an ML estimate of μ was obtained iteratively by using the Newton–Raphson method. For each sample size, the experiment was repeated 20,000 times ($=n_r$).

Results and discussion

The average (m) and standard deviation (s) of the estimates obtained by the two methods are presented in Table 1 for all sample sizes investigated in this study. In Table 1, the 95% confidence limits on the average of the estimates are also provided for the two methods. The confidence limits for the average of the estimators were calculated by

$$m - z\frac{s}{\sqrt{n_r}} \leq \delta \leq m + z\frac{s}{\sqrt{n_r}} \quad (8)$$

where δ is the population average of the estimates of μ and z is 1.95996 for 95% confidence. Note that unity is within the confidence limits for the sample average method for all sample sizes. Hence, the sample average method tends to yield unbiased estimates of μ. For the ML method, however, unity is not within the confidence limits for any sample size. Therefore, the ML estimates have a negative bias, although the bias is very small being less than 1.5 and 0.5% in the worst cases, $n = 10$ and 25, respectively. For practical purposes, the bias of the ML method can be ignored for $n \geq 25$.

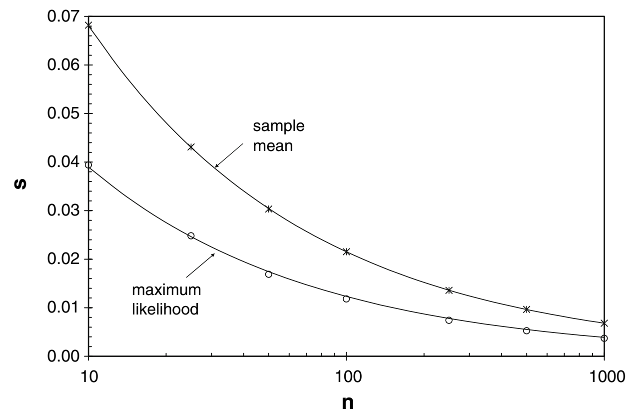

Fig. 2 The change in the standard deviation of the estimates obtained by the two methods plotted as a function of the sample size

The change in standard deviation with sample size for the two methods is presented in Fig. 2. The standard deviation of the estimates obtained by the ML method is approximately one half of that of sample averages. The curves in Fig. 2 were drawn by the equation:

$$s = \frac{A}{\sqrt{n}} \quad (9)$$

where A is 0.2155 and 0.1232 for sample average and ML methods, respectively. Both curves have an R^2 in excess of 0.999. Because the standard deviation of estimates is much lower for the ML method and both methods yield practically unbiased estimates, it is recommended that the ML estimator, i.e., Eq. 7, developed in this study be used in lieu of taking the sample average.

The distribution of $\tilde{\mu}/\mu$ and $\bar{x}/\mu$, estimated by the two methods, was analyzed for normality for all sample sizes. Fits to the normal distribution were assessed by the Anderson–Darling goodness-of-fit test [14–16]:

Table 1 The average and standard deviation of estimates obtained by the two methods, as well as the 95% confidence intervals on the mean for all sample sizes investigated in this study

	n						
	10	25	50	100	250	500	1000
Sample average							
m	0.9999	0.9997	1.0002	1.0000	0.9999	1.0000	1.0000
s	0.0682	0.0431	0.0303	0.0215	0.0136	0.0097	0.0068
$\bar{x}_{0.025}$	0.9990	0.9991	0.9997	0.9997	0.9997	0.9999	0.9999
$\bar{x}_{0.975}$	1.0009	1.0002	1.0006	1.0003	1.0001	1.0001	1.0001
ML							
m	0.9863	0.9943	0.9973	0.9985	0.9994	0.9997	0.9999
s	0.0394	0.0248	0.0169	0.0118	0.0074	0.0053	0.0037
$\tilde{\mu}_{0.025}$	0.9869	0.9940	0.9971	0.9984	0.9993	0.9996	0.9998
$\tilde{\mu}_{0.975}$	0.9857	0.9947	0.9975	0.9987	0.9995	0.9998	0.9999

$$A^2 = -n - \frac{1}{n}\sum_{i=1}^{n}\left[(2i-1)\ln(P_i) + (2n+1-2i)\ln(1-P_i)\right] \quad (10)$$

where i is the rank (in ascending order) and P is the cumulative probability for each data point. For both methods and all sample sizes, the hypothesis that the distribution of estimates is normal was rejected. The two-parameter Weibull and lognormal distributions yielded similar results. Consequently, percentage points of the distributions for the estimates of μ obtained by the sample average and ML methods were developed, as presented in Tables 2 and 3, respectively. These tables can be used to determine the confidence limits for population average. For instance, assume that μ was estimated to be 26.5 by the sample average method from a sample of 250 particles. Let us suppose that 95% confidence limits are needed. From Table 2, $X_{0.025,250}$ and $X_{0.975,250}$ are 0.9728 and 1.0262, respectively. Therefore,

$$0.9728 \leq \frac{26.5}{\mu} \leq 1.0262 \quad (11)$$

$$\frac{26.5}{1.0262} \leq \mu \leq \frac{26.5}{0.9728} \quad (11a)$$

Hence, μ lies between 25.82 and 27.24 with 95% confidence interval.

An effort was made to develop an empirical equation to interpolate the percentage points to all sample sizes between 10 and 1000. Best results were obtained using

$$X = \frac{\beta_0 + \beta_1 n}{n^{\xi}} \quad (12)$$

where β_0, β_1, and ζ are constants, the values of which are presented in Tables 4 and 5 for sample average and ML

Table 4 Constants for Eq. 12 for various percentage points for sample average method

	β_0	β_1	ζ
0.005	−1.0203	0.8804	0.9835
0.01	−0.9858	0.8977	0.9862
0.025	−0.8112	0.9142	0.9885
0.05	−0.6665	0.9277	0.9904
0.1	−0.4868	0.9424	0.9924
0.25	−0.2266	0.9694	0.9960
0.5	0.0289	1.0003	1.0000
0.75	0.2640	1.0317	1.0040
0.9	0.4541	1.0596	1.0074
0.95	0.5485	1.0761	1.0094
0.975	0.6087	1.0930	1.0114
0.99	0.6796	1.1127	1.0138
0.995	0.7234	1.1244	1.0151

Table 2 Percentage points for the distribution of $\bar{x}/\mu$ (sample average method)

n	0.005	0.01	0.025	0.05	0.1	0.25	0.5	0.75	0.9	0.95	0.975	0.99	0.995
10	0.8100	0.8262	0.8563	0.8811	0.9103	0.9559	1.0031	1.0480	1.0856	1.1059	1.1228	1.1423	1.1541
25	0.8812	0.8935	0.9123	0.9270	0.9434	0.9712	1.0010	1.0297	1.0544	1.0679	1.0807	1.0953	1.1041
50	0.9179	0.9274	0.9387	0.9487	0.9606	0.9803	1.0009	1.0210	1.0385	1.0488	1.0576	1.0676	1.0734
100	0.9401	0.9475	0.9567	0.9643	0.9725	0.9857	1.0005	1.0150	1.0273	1.0340	1.0404	1.0478	1.0528
250	0.9633	0.9669	0.9728	0.9774	0.9826	0.9910	1.0001	1.0091	1.0171	1.0217	1.0262	1.0308	1.0337
500	0.9749	0.9773	0.9807	0.9839	0.9876	0.9935	1.0001	1.0065	1.0124	1.0157	1.0188	1.0220	1.0240
1000	0.9824	0.9840	0.9866	0.9887	0.9911	0.9954	1.0001	1.0045	1.0087	1.0112	1.0133	1.0156	1.0172

Table 3 Percentage points for the distribution of $\tilde{\mu}/\mu$ (ML method)

n	0.005	0.01	0.025	0.05	0.1	0.25	0.5	0.75	0.9	0.95	0.975	0.99	0.995
10	0.8465	0.8677	0.8941	0.9128	0.9319	0.9615	0.9908	1.0163	1.0355	1.0461	1.0547	1.0642	1.0699
25	0.9221	0.9312	0.9419	0.9517	0.9617	0.9787	0.9962	1.0116	1.0243	1.0319	1.0382	1.0447	1.0493
50	0.9503	0.9551	0.9627	0.9684	0.9753	0.9864	0.9980	1.0091	1.0182	1.0236	1.0285	1.0336	1.0377
100	0.9661	0.9699	0.9748	0.9787	0.9834	0.9908	0.9988	1.0067	1.0133	1.0172	1.0206	1.0246	1.0275
250	0.9796	0.9816	0.9845	0.9870	0.9899	0.9945	0.9995	1.0044	1.0087	1.0113	1.0134	1.0163	1.0181
500	0.9858	0.9874	0.9892	0.9910	0.9929	0.9962	0.9998	1.0033	1.0064	1.0082	1.0097	1.0116	1.0128
1000	0.9903	0.9912	0.9925	0.9937	0.9952	0.9974	0.9999	1.0024	1.0046	1.0059	1.0070	1.0083	1.0092

Table 5 Constants for Eq. 12 for various percentage points for ML

	β_0	β_1	ζ
0.005	−1.1672	0.9513	0.9939
0.01	−0.9731	0.9529	0.9940
0.025	−0.7446	0.9565	0.9944
0.05	−0.5984	0.9619	0.9951
0.1	−0.4654	0.9696	0.9960
0.25	−0.2684	0.9834	0.9979
0.5	−0.0879	0.9994	0.9999
0.75	0.0557	1.0153	1.0019
0.9	0.1482	1.0299	1.0037
0.95	0.1912	1.0393	1.0049
0.975	0.2170	1.0484	1.0060
0.99	0.2517	1.0571	1.0071
0.995	0.2546	1.0652	1.0081

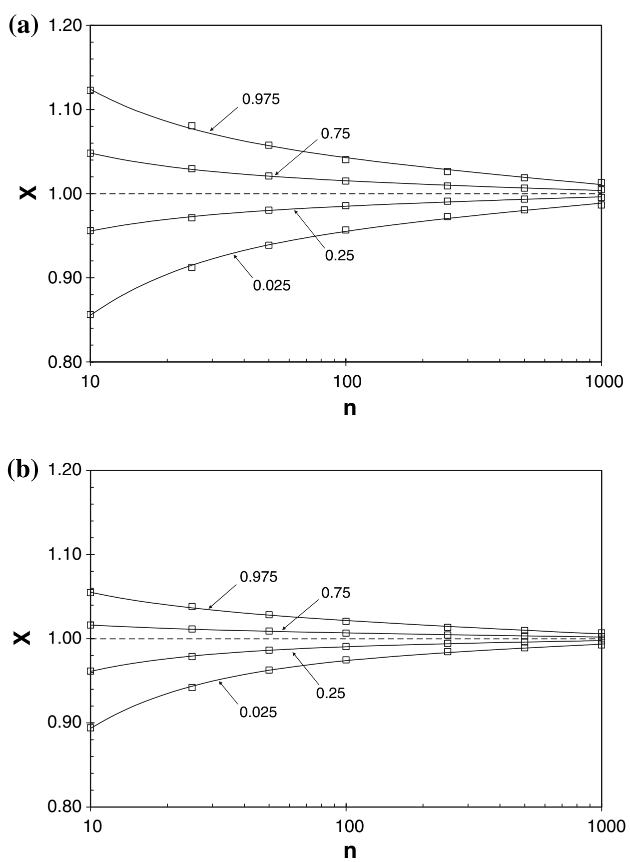

Fig. 3 The change in the four critical points of distribution of **a** $\bar{x}/\mu$ (sample average method) and **b** $\tilde{\mu}/\mu$ (ML method), on semi-log scale

methods, respectively. For all fits, R^2 was in excess 0.997. The change in four percentage points listed in Tables 4 and 5 with sample size and the predictions of Eq. 12 are presented in Fig. 3, which shows an excellent agreement. In Fig. 3, the solid line represents the predicted values of the percentile points and the markings represent the actual values from Tables 4 and 5.

To illustrate the use of Tables 4 and 5, let us assume that μ was estimated to be 85.5 from a sample of 155 precipitates by using the ML method. If 90% confidence limits are desired, then by using the values of β_0, β_1, and ζ given for 0.05 and 0.95 in Table 5, the limits are calculated as

$$\frac{85.5(155)^{1.0049}}{0.1912 + 1.0393(155)} \leq \mu \leq \frac{85.5(155)^{0.9951}}{-0.5984 + 0.9619(155)} \tag{13}$$

Hence, the 90% confidence limits for μ are 84.23 and 87.07.

As stated previously, most studies in the literature reported particle size distributions that are somewhat wider than the LSW distribution. Most of these distributions undoubtedly do not follow the LSW distribution. In some studies, however, the reported histograms are not only negatively skewed, but also only marginally wider than the LSW distribution (e.g., Figs. 7 and 13 by Mahalingam et al. [17], Fig. 8 by Cho and Ardell [18], Fig. 12 by Novotny and Ardell [19], Fig. 12 by Baldan [20]). The data for those distributions should be reanalyzed and μ should be estimated by the ML method. Moreover, fits to the LSW distribution should be reanalyzed by using the confidence intervals for μ introduced in this study. The statistical tools introduced in this study should be used whenever researchers need to fit the LSW distribution to their data.

The statistical procedures applied to the LSW distribution in this article should be applied to the other particle size distributions, suggested as improvements over the LSW distribution in the literature. Moreover, formal goodness-of-fit hypothesis tests should replace the subjective assessment by superposing distributions over particle size histograms. The authors will report their findings in these two aspects in the future.

Conclusions

- The probability density function that has been used so far in the literature for steady state during particle coarsening is the standard LSW distribution. A generic (or general) probability density function was developed in this study.
- By using the generic probability density function, an ML estimator for μ was introduced in this study.
- Monte Carlo simulations for sample sizes ranging from 10 to 1000 showed that the ML estimator is practically unbiased for $n \geq 25$, as the conventional method of taking the sample average as the estimate of μ. Moreover, the standard deviation of ML estimates is approximately half of that obtained by sample averages. Therefore, it is

recommended that ML estimator be used in lieu of taking sample average to estimate the population average of the LSW distribution.

- The distribution of the estimated μ was found neither to follow the normal, lognormal nor 2-parameter Weibull distributions by using the Anderson–Darling goodness-of-fit test.
- Percentage points for the distribution of estimated μ for both methods and for all sample sizes investigated in this study were provided. Moreover, an empirical equation was developed to interpolate to any sample size between 10 and 1000 for various percentage points.
- Fits to the LSW distribution should be reanalyzed by using the confidence intervals for μ introduced in this study. The statistical tools introduced in this study should be used whenever researchers need to fit the LSW distribution to their data.
- The statistical methods presented in this article should be applied to the other particle size distributions, suggested as improvements over the LSW distribution in the literature. Moreover, formal goodness-of-fit hypothesis tests should replace the subjective assessment by superposing distributions over particle size histograms.

Appendix

The probability density function for the LSW distribution is:

$$f\left(\frac{x}{\mu}\right)=\frac{4}{9}\left(\frac{x}{\mu}\right)^2\left(\frac{3}{3+\frac{x}{\mu}}\right)^{7/3}\left(\frac{1.5}{1.5-\frac{x}{\mu}}\right)^{11/3}\exp\left(\frac{\frac{x}{\mu}}{\frac{x}{\mu}-1.5}\right);\quad 0<\frac{x}{\mu}<1.5 \tag{2}$$

For simplicity, put $y=\frac{x}{\mu}$ $(0<y<1.5)$. We have

$$\int_0^{1.5}\frac{4}{9}y^2\left(\frac{3}{3+y}\right)^{7/3}\left(\frac{1.5}{1.5-y}\right)^{11/3}\exp\left(\frac{y}{y-1.5}\right)\mathrm{d}y=1 \tag{14}$$

Multiply and divide the integral by μ to get

$$1=\frac{1}{\mu}\left(\mu\int_0^{1.5}\frac{4}{9}y^2\left(\frac{3}{3+y}\right)^{7/3}\left(\frac{1.5}{1.5-y}\right)^{11/3}\exp\left(\frac{y}{y-1.5}\right)\mathrm{d}y\right) \tag{15}$$

Since for any function h and positive μ, we have

$$\mu\int_0^{1.5}h(y)\mathrm{d}y=\int_0^{1.5\mu}h\left(\frac{y}{\mu}\right)\mathrm{d}y \tag{16}$$

Equation 14 can now be written as

$$\begin{aligned}1&=\frac{1}{\mu}\int_0^{1.5\mu}\frac{4}{9}\left(\frac{y}{\mu}\right)^2\left(\frac{3}{3+\frac{y}{\mu}}\right)^{\frac{7}{3}}\left(\frac{1.5}{1.5-\frac{y}{\mu}}\right)^{\frac{11}{3}}\exp\left(\frac{\frac{y}{\mu}}{\frac{y}{\mu}-1.5}\right)\mathrm{d}y\\&=\int_0^{1.5\mu}\underbrace{\frac{4}{9\mu}\left(\frac{y}{\mu}\right)^2\left(\frac{3}{3+\frac{y}{\mu}}\right)^{\frac{7}{3}}\left(\frac{1.5}{1.5-\frac{y}{\mu}}\right)^{\frac{11}{3}}\exp\left(\frac{\frac{y}{\mu}}{\frac{y}{\mu}-1.5}\right)}_{g(y)}\mathrm{d}y\end{aligned} \tag{17}$$

Replacing the dummy variable y above by x, we obtain

$$g(x)=\frac{4}{9\mu}\left(\frac{x}{\mu}\right)^2\left(\frac{3}{3+\frac{x}{\mu}}\right)^{\frac{7}{3}}\left(\frac{1.5}{1.5-\frac{x}{\mu}}\right)^{\frac{11}{3}}\exp\left(\frac{\frac{x}{\mu}}{\frac{x}{\mu}-1.5}\right) \tag{18}$$

and

$$\int_0^{1.5\mu}g(x)\mathrm{d}x=1 \tag{19}$$

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