

**Chao Wang<sup>1</sup>**

Pacific Northwest National Laboratory,  
Richland, WA 99354  
e-mail: chao.wang@pnnl.gov

**Zhijie Xu**

Pacific Northwest National Laboratory,  
Richland, WA 99354  
e-mail: zhijie.xu@pnnl.gov

**Deborah Fagan**

Pacific Northwest National Laboratory,  
Richland, WA 99354  
e-mail: deborah.fagan@pnnl.gov

**David P. Field**

Voiland College of Engineering and Architecture,  
Washington State University,  
Pullman, WA 99164  
e-mail: dfield@wsu.edu

**Curt Lavender**

Pacific Northwest National Laboratory,  
Richland, WA 99354  
e-mail: curt.lavender@pnnl.gov

**Vineet V. Joshi**

Pacific Northwest National Laboratory,  
Richland, WA 99354  
e-mail: Vineet.joshi@pnnl.gov

# Quantifying and Qualifying Alloys Based on Level of Homogenization: A U-10Mo Alloy Case Study

*Homogenization heat treatment is performed to attain uniformity in microstructure which is helpful to achieve the desired workability and microstructure in final products and, eventually, to gain predictive and consistent performance. Fabrication of low-enriched uranium alloys with 10 wt% molybdenum (U-10Mo) fuel plates involves multiple thermomechanical processing steps. It is well known that the molybdenum homogeneity in the final formed product affects the performance in the nuclear reactor. To ensure uniform homogenization, a statistical method is proposed to quantify and characterize the molybdenum concentration variation in U-10Mo fuel plates by analyzing the molybdenum concentration measurement data from scanning electron microscopy energy dispersive spectroscopy line-scan. Statistical tolerance intervals (TI) are employed to determine the qualification of the U-10Mo fuel plate. We formulate an argument for the minimum number of independent samples to define fuel plate qualification if no molybdenum measurement data are available in advance and demonstrate that the given TI requirements can be equivalently reduced to a sample variance criterion in this application. The outcome of the statistical analysis can be used to optimize casting design and eventually increase productivity and reduce fabrication costs. The statistical strategy developed in this paper can be implemented for other applications especially in the field of material manufacturing to assess qualification requirements and monitor and improve the process design. [DOI: 10.1115/1.4044891]*

**Keywords:** tolerance interval, U-10Mo, qualification specification, statistical analysis, casting and homogenization, quality control, materials processing

## 1 Introduction

The US National Nuclear Security Administration drives the need to develop and design low-enriched uranium (LEU) fuels to progressively reduce and eventually replace high-enriched uranium (HEU) fuels for US high-power civil and research reactors [1]. Uranium alloys with 10 wt% molybdenum (U-10Mo) are identified as the most promising candidate for LEU fuel due to its high intrinsic density and good irradiation behavior [2–4]. The manufacturing of the U-Mo alloy requires multiple complex thermomechanical processes [5,6] including casting, homogenization, hot/cold rolling, annealing, and hot isostatic pressing. Every process is highly challenging and requires a tremendous amount of planning and preparation. Fuel plate quality after casting and homogenization is fundamental to ensure high quality of the final U-Mo product because it can greatly affect the subsequent microstructure evolution and eventually influence the fuel performance in a reactor. To determine the qualification of a given fuel plate, a statistical analysis of the chemical composition measured using an energy dispersive spectroscopy (EDS) technique is a reliable, efficient, and cost-effective. Statistical methods have been widely used to characterize the quality of processing products in many applications [7–10]. For this study, tolerance interval (TI) calculations will be used to determine the quality of as-cast plates, identify

potential quality problems, and eventually enhance the quality and efficiency of the entire fabrication process, thereby reducing fabrication costs.

Previous works [6,11] reveal that the distribution of molybdenum in fuel plates has a significant impact on the quality and performance of U-Mo fuel. In principle, the overall average molybdenum concentration should be 10% throughout the entire plate. However, the variation in molybdenum concentrations after casting is significant due to molybdenum segregation and a homogenization treatment is necessary to produce a more uniform molybdenum distribution. Therefore, statistical analysis is needed to quantify the variation in molybdenum concentrations that determines the qualification of the fuel plate.

Two common measures of product quality compare the confidence interval (CI) for a parameter and/or a TI to specified limits [12,13]. A CI specifies the statistical interval that might contain the true value of population parameter, e.g., mean or variance, with a given probability. The width of the confidence interval will approach zero with the increasing sample size. A TI bounds a specified proportion of a sampled population at a given CI. The width of TI is also dependent on the number of samples and variance.

Extensive work [14] has been performed for quality control in a broad range of applications, where the TI is used as a quantitative metric for assessing the product quality. However, no such attempt has been published on characterizing the molybdenum concentration variation in U-Mo fuel plates for fuel qualification. A recent statement of qualification specification for the U-Mo fuel plate has been proposed [15] and quoted herein as follows: “A sufficient number of samples shall be randomly taken from a sufficient number of fuel plates randomly selected from a fuel plate lot to allow statistical determination of 95% confidence interval that 95% of the U-Mo has  $10.0 \pm 1.0$  wt% molybdenum microscopically throughout the U-Mo.” This statement prompts one to use TI to

<sup>1</sup>Corresponding author.

Contributed by the Materials Division of ASME for publication in the JOURNAL OF ENGINEERING MATERIALS AND TECHNOLOGY. Manuscript received February 6, 2019; final manuscript received September 5, 2019; published online September 18, 2019. Assoc. Editor: Peter W. Chung.

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quantify the molybdenum concentration variation and ensure product quality. This specification can be equivalently interpreted as follows: 95% of the total U-Mo population within a plate must fall between 9% and 11% molybdenum wt% with a confidence interval level of 95%. However, there is no guideline on the “sufficient number of samples” in the statement. As in other sampling scenarios, there is a tradeoff between the number of high-quality measurements that can be obtained and the total time cost for data collection. Specifically, a single molybdenum wt% measurement from EDS line-scans takes about 30 s to 1 min, and a large number of measurements are required to meet statistical specifications, given inherent molybdenum variability within a plate. In addition to determining the fuel plate qualification, this study also provides insights into an inverse problem, i.e., how to determine the optimal number of measurements required to qualify the given plate.

Fuel plate qualification can be specified in terms of the sample variance rather than the calculation of tolerance limits, which is a simplified metric that may be applied given reasonable assumptions. One of these assumptions is that the collected samples must be independent of one another, which ensures that the selected data properly represent the characteristics of the entire fuel plate. The product is considered qualified only if the simplified metrics are satisfied and the assumptions can be verified. The final results of the statistical analysis can be used to optimize the casting design, refine casting specifications, and eventually enhance the casting productivity. The statistical strategy developed in this paper can be implemented in other applications, especially in the field of material manufacturing, to define reasonable qualification requirements and provide insights to improve the process design.

The rest of the paper is organized as follows: Section 2 describes the statistical methodology including a comprehensive introduction of TI approaches and key mathematical formulas. Section 3 presents the TI implementation on the specified U-Mo qualification specification, and two examples are analyzed as demonstration. Section 4 introduces our recommendation on the minimum number of molybdenum wt% measurements and the sample variance criterion to determine the fuel plate qualification. The conclusions are summarized in Sec. 5.

## 2 Methodology

The general definition of a TI can be described as follows. Let  $\mathbf{Y} = (Y_1, Y_2, \dots, Y_N)$  be  $N$ -independent random samples drawn from a continuous cumulative distribution function,  $F$ . In addition, let an interval  $[L(\mathbf{Y}), U(\mathbf{Y})]$  based on the sample vector  $\mathbf{Y}$  be a two-sided TI for  $F$  such that for any given  $0 < \gamma < 1$  and  $0 < P < 1$ , the following probability expression is satisfied:

$$\Pr(F(U(\mathbf{Y})) - F(L(\mathbf{Y})) \geq P) = \gamma \quad (1)$$

where  $\gamma$  and  $P$  define a general two-sided TI that has a confidence level of  $\gamma \times 100\%$  containing at least  $P \times 100\%$  of the population. If  $L(\mathbf{Y}) = -\infty$  and  $U(\mathbf{Y}) < +\infty$ , then the previous two-sided interval becomes  $[-\infty, U(\mathbf{Y})]$  and can be defined more properly as a one-sided TI  $(\gamma, P)$  with an upper tolerance bound. Similarly, if  $L(\mathbf{Y}) > -\infty$  and  $U(\mathbf{Y}) = +\infty$ , the interval  $[L(\mathbf{Y}), +\infty]$  is a one-sided TI  $(\gamma, P)$  with a lower tolerance bound only [16].

Now, suppose that the sample vector  $\mathbf{Y} = (Y_1, Y_2, \dots, Y_N)$  is independently drawn from a standard normal distribution  $N(\mu, \sigma^2)$  with the population mean  $\mu$  and the population variance  $\sigma^2$ . We can use  $\bar{Y}$  and  $s$  to denote sample mean and sample variance, respectively. The mathematical expressions for  $\bar{Y}$  and  $s$  can be written as

$$\bar{Y} = \sum_{i=1}^N Y_i \quad (2)$$

$$s = \sqrt{\frac{\sum_{i=1}^N (Y_i - \bar{Y})^2}{N - 1}} \quad (3)$$

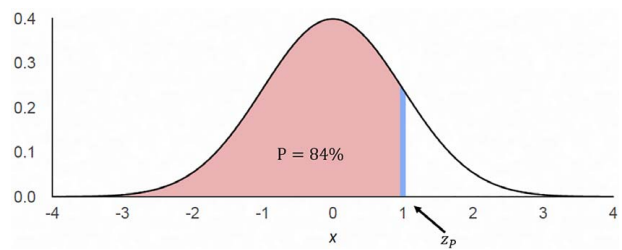


Fig. 1 The critical value  $z_p$  for a standard normal distribution

Table 1 Critical values  $z_p$  for some commonly used probability  $P$

| $P$   | 0.01   | 0.025  | 0.05   | 0.1    | 0.9   | 0.95  | 0.975 | 0.99  |
|-------|--------|--------|--------|--------|-------|-------|-------|-------|
| $z_p$ | -2.326 | -1.960 | -1.645 | -1.282 | 1.282 | 1.645 | 1.960 | 2.326 |

The formulas to describe the one-sided TI can be expressed as

$$L(\mathbf{Y}) = \bar{Y} - k_1(N, \gamma, P)s \quad (4)$$

or

$$U(\mathbf{Y}) = \bar{Y} + k_1(N, \gamma, P)s \quad (5)$$

where  $k_1$  is the factor to ensure that at least a proportion  $P \times 100\%$  of the entire population with a confidence interval level of  $\gamma \times 100\%$  is satisfied based on the sample of size  $N$ . The value of  $k_1$  can be calculated based on the inverse cumulative distribution function for a non-central  $t$  distribution [17]:

$$k_1(N, \gamma, P) = \frac{t_{N-1, \gamma, \delta}}{\sqrt{N}} \quad (6)$$

where  $t_{N-1, \gamma, \delta}$  is the critical value ( $\gamma \times 100$  percentile), i.e.,  $F(t_{N-1, \gamma, \delta}) = \gamma$ , for a non-central  $t$  distribution with  $N - 1$  degrees-of-freedom and the non-centrality parameter  $\delta = z_p \sqrt{N}$ . Here,  $z_p$  is the critical value of the standard normal distribution associated with a cumulative probability of  $P \times 100\%$ . For example, Fig. 1 shows that the accumulated probability for  $-\infty \leq x \leq 1$  is 84%, which is equivalent to  $\Pr[x \leq z_p = 1] = 84\%$ . The value of  $z_p$  can be found in Ref. [18] for any given  $P$ . Some commonly used  $P$  and the associated  $z_p$  values are listed in Table 1.

If  $N$  is greater than 50 [19,20],  $k_1$  can be approximated using the following equation:

$$k_1(N, \gamma, P) = \frac{z_p + \sqrt{z_p^2 - ab}}{a} \quad (7a)$$

$$a = 1 - \frac{z_\gamma^2}{2(N - 1)} \quad (7b)$$

$$b = z_p^2 - \frac{z_\gamma^2}{N} \quad (7c)$$

Next, the upper and lower bounds for a two-sided TI are defined as

$$L(\mathbf{Y}) = \bar{Y} - k_2(N, \gamma, P)s \quad (8a)$$

and

$$U(\mathbf{Y}) = \bar{Y} + k_2(N, \gamma, P)s \quad (8b)$$

<sup>2</sup><https://homepage.divms.uiowa.edu/~mbognar/>

The exact solution for the two-sided  $k_2$  factor can be obtained by solving the following implicit nonlinear integral equation [21,22]:

$$\sqrt{\frac{2N}{\pi}} \int_0^{+\infty} \Pr\left(\chi_{N-1}^2 > \frac{(N-1)\chi_{1,P,\delta}^2}{k_2(N, \gamma, P)^2}\right) e^{-(1/2)Nz^2} dz = \gamma \quad (9)$$

where  $\chi_{N-1}^2$  is the chi-squared probability distribution function with  $N-1$  degrees-of-freedom;  $\chi_{1,P,\delta}^2$  is the  $P \times 100\%$  percentile of the chi-squared distribution with one degree-of-freedom and non-centrality parameter  $\delta = z^2$ , where  $z$  is a dummy variable used for integration. Equation (9) is quite complicated to solve directly. Moreover, the following approximation can be used to simplify the calculation of the two-sided  $k_2$  factor [23]:

$$k_2(N, \gamma, P) = \left( \frac{(N-1) \left(1 + \frac{1}{N}\right) z_{(1+P)/2}^2}{\chi_{1-\gamma, N-1, 0}^2} \right)^{1/2} \quad (10)$$

where  $\chi_{1-\gamma, N-1, 0}^2$  is the critical value ( $1-\gamma$  percentile) of the central chi-squared distribution with  $N-1$  degrees-of-freedom and non-centrality parameter  $\delta = 0$ , i.e.,  $F(\chi_{1-\gamma, N-1, 0}^2) = 1-\gamma$ . Figure 2 shows an example chi-squared distribution with  $N-1 = 3$ . The probability of having a value greater than  $\chi_{1-0.57, 3, 0}^2 = 2$  is 57%, which is equivalent to  $\Pr[x \geq \chi_{1-\gamma, N-1, 0}^2 = 2] = 57\%$ . The values of  $\chi_{1-\gamma, N-1, 0}^2$  for any given  $\gamma$  and  $N$  can be found in Ref. [18]. Some commonly used critical values of chi-square distribution are listed in Table 2. To evaluate the quality of the  $k_2$  approximation from Eq. (10), a comparison is conducted by solving the true solutions calculated from Eq. (9) using the R package developed in Ref. [22]. The relative error between approximation and true solution is defined as  $1 - (k_2(\text{approximation}) / (k_2(\text{true solution})))$ . Figure 3 plots the number of sample  $N$  versus the relative error for  $P = \gamma = 0.95$ . The approximations first slightly underpredict and then overpredict the true value of  $k_2$  with the increasing  $N$ , but the relative error is always within the range of  $\pm 0.36\%$  and gradually approaches zero with increasing  $N$ . These results indicate that the  $k_2$  values obtained from Eq. (10) can be considered as good approximations to represent the true solutions for  $N$  approaching 100.

### 3 Statistical Analysis for Qualification of U-10Mo

Based on the specification statement for U-10Mo, we can set  $P = \gamma = 0.95$ . In addition, this requirement also defines the lower and upper bounds for 95/95 tolerance interval as 9% and 11%, respectively. Note that an average concentration  $\bar{Y} = 10\%$  is assumed for all samples and is assumed known without error so Eqs. (4), (5), and (8) become a function of  $k_2$  and  $s$ . The number of samples,  $N$ , namely the number of measurements of molybdenum concentration required when performing EDS scan experiments for each fuel plate is not explicitly specified and will be determined in this study. Molybdenum concentration will drop significantly where the scan intersects a carbide or secondary phase spot. To avoid the bias due to carbides, only molybdenum weight percentage between 7% and 13% is considered as effective

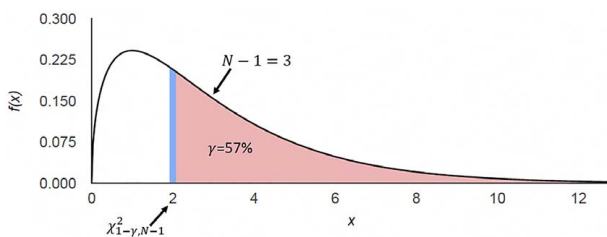


Fig. 2 The critical value for a central chi-squared distribution with  $N-1 = 3$  and  $\gamma = 57\%$

Table 2 Commonly used critical values ( $\chi_{1-\gamma, N-1, 0}^2$ ) of the chi-square distribution with different  $1-\gamma$  and  $N-1$

| $N-1$ | $1-\gamma$ |        |        |        |        |
|-------|------------|--------|--------|--------|--------|
|       | 0.1        | 0.05   | 0.025  | 0.01   | 0.001  |
| 1     | 0.016      | 0.004  | 0.001  | 0      | 0      |
| 2     | 0.211      | 0.103  | 0.051  | 0.02   | 0.002  |
| 3     | 0.584      | 0.352  | 0.216  | 0.115  | 0.024  |
| 4     | 1.064      | 0.711  | 0.484  | 0.297  | 0.091  |
| 5     | 1.61       | 1.145  | 0.831  | 0.554  | 0.21   |
| 6     | 2.204      | 1.635  | 1.237  | 0.872  | 0.381  |
| 7     | 2.833      | 2.167  | 1.69   | 1.239  | 0.598  |
| 8     | 3.49       | 2.733  | 2.18   | 1.646  | 0.857  |
| 9     | 4.168      | 3.325  | 2.7    | 2.088  | 1.152  |
| 10    | 4.865      | 3.94   | 3.247  | 2.558  | 1.479  |
| 15    | 8.547      | 7.261  | 6.262  | 5.229  | 3.483  |
| 20    | 12.443     | 10.851 | 9.591  | 8.26   | 5.921  |
| 25    | 16.473     | 14.611 | 13.12  | 11.524 | 8.649  |
| 30    | 20.599     | 18.493 | 16.791 | 14.953 | 11.588 |
| 35    | 24.797     | 22.465 | 20.569 | 18.509 | 14.688 |
| 40    | 29.051     | 26.509 | 24.433 | 22.164 | 17.916 |
| 45    | 33.35      | 30.612 | 28.366 | 25.901 | 21.251 |
| 50    | 37.689     | 34.764 | 32.357 | 29.707 | 24.674 |
| 55    | 42.06      | 38.958 | 36.398 | 33.57  | 28.173 |
| 60    | 46.459     | 43.188 | 40.482 | 37.485 | 31.738 |
| 65    | 50.883     | 47.45  | 44.603 | 41.444 | 35.362 |
| 70    | 55.329     | 51.739 | 48.758 | 45.442 | 39.036 |
| 75    | 59.795     | 56.054 | 52.942 | 49.475 | 42.757 |
| 80    | 64.278     | 60.391 | 57.153 | 53.54  | 46.52  |
| 85    | 68.777     | 64.749 | 61.389 | 57.634 | 50.32  |
| 90    | 73.291     | 69.126 | 65.647 | 61.754 | 54.155 |
| 95    | 77.818     | 73.52  | 69.925 | 65.898 | 58.022 |
| 100   | 82.358     | 77.929 | 74.222 | 70.065 | 61.918 |

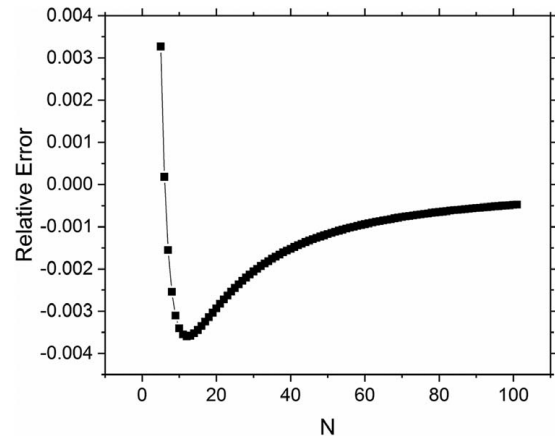


Fig. 3 Relation between relative error and  $N$  for  $k_2$

measurements and used in the plate qualification analysis. The effect of the distribution of carbides and/or secondary phase spots on the fuel plate quality is currently not considered in this study.

For any independently drawn molybdenum concentration vector of length  $N$  which is close to the neighborhood of 100, Eq. (10) can be applied to calculate the  $k_2$  factor. With  $P = 0.95$ , we can find  $z_{(1+P)/2} = z_{0.975} = 1.96$ . The values of  $\chi_{1-\gamma, N-1, 0}^2 = \chi_{0.05, N-1, 0}^2$  and  $k_2(N, \gamma, P) = k_2(N, 0.95, 0.95)$  become a function of  $N$  only and are calculated once  $N$  is provided. Equation (8) can be used to determine lower and upper bounds ( $L$  and  $U$ ) for the observed vector of molybdenum concentrations. The fuel plate is determined to be qualified in terms of the molybdenum distribution if both the upper and lower bounds satisfy the specification requirement, i.e.,  $L \geq 9\%$  and  $U \leq 11\%$ . Otherwise, the plate will be rejected.

Two examples with EDS line-scan measurements of molybdenum concentration were used to demonstrate the procedure of

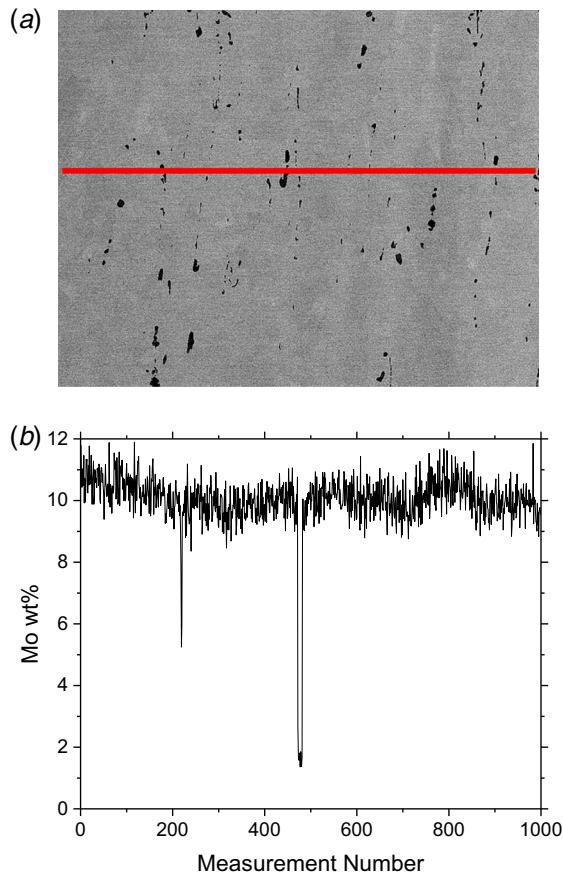


Fig. 4 (a) SEM image and (b) EDS line scan for Plate 1

qualification determination. Figure 4 shows the secondary electron image from the scanning electron microscope (SEM), together with its EDS line-scan in red for the first sample fuel plate (Plate 1). The domain size for Fig. 4 is  $726 \times 567 \mu\text{m}$ . Figure 5 shows the observed probability density function from a total of 999 molybdenum concentration measurements taken along the line. A bimodal distribution with two peaks is evident, the smaller of which represents the carbide regions. To exclude the anomalies from the carbides, only 946 observations are considered as effective measurements with molybdenum concentrations between 7 wt% and 13 wt%. Given the exclusion of carbides, molybdenum wt% is expected to

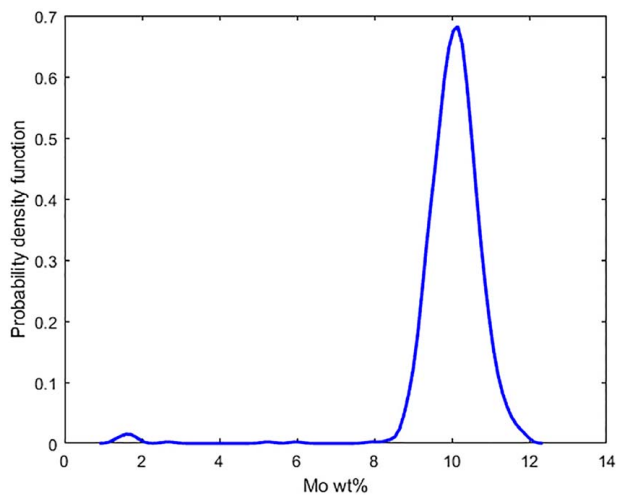


Fig. 5 Probability density function from 999 molybdenum wt% measurements

be normally distributed and consistent with the assumptions described in Sec. 2. Next, the dependence of the molybdenum concentration measurements for Plate 1 is explored using the autocorrelation function (ACF), which is a commonly used method for detecting non-randomness in data. Given equally spaced measurements, the autocorrelation function  $r$  for lag  $j$  is defined as

$$r_j = \frac{\sum_{i=1}^{N-j} (Y_i - \bar{Y})(Y_{i+j} - \bar{Y})}{\sum_{i=1}^N (Y_i - \bar{Y})^2} \quad (11)$$

A unit lag distance ( $j = 1$ ) in our study is  $1 \mu\text{m}$  which represents the space between the adjacent molybdenum concentration measurements. By observing the line-scan data listed in Fig. 4(b), it is evident that the measurements vary in a fashion that is not purely random, and it is a strong indication of data correlation. This trend becomes even more obvious if carbides are excluded. The findings from Fig. 6, which plots the ACF results for Plate 1, are consistent with the observations from the measured data. In particular, the critical lag distance indicating no data intercorrelation increases from  $\sim 15 \mu\text{m}$  to  $\sim 100 \mu\text{m}$  after neglecting carbides. Critical lag distance is an important parameter for determining the

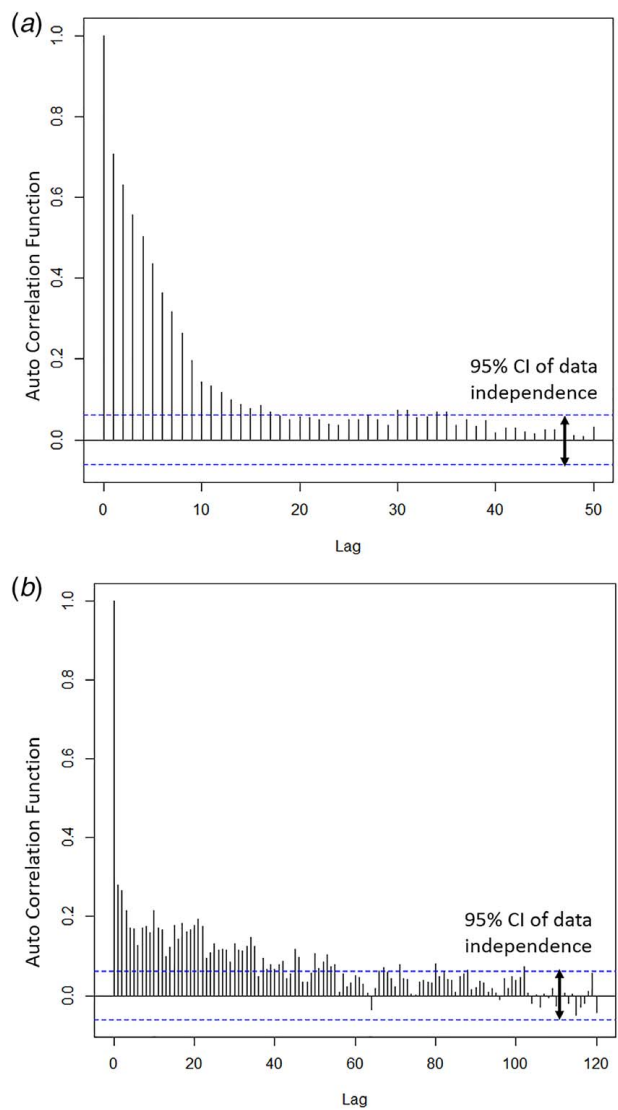


Fig. 6 ACF results for space (a) with and (b) without carbide particles

**Table 3 Impact of the number of samples on plate qualification for Plate 1**

| $N_s$               | 5     | 10    | 20    | 50    | 100   | 200   | 946   |
|---------------------|-------|-------|-------|-------|-------|-------|-------|
| Repetition          | 1000  | 1000  | 1000  | 1000  | 1000  | 1000  | 1     |
| $k_2$               | 5.09  | 3.38  | 2.75  | 2.37  | 2.23  | 2.14  | 2.03  |
| Mean ( $s$ )        | 0.53  | 0.55  | 0.56  | 0.57  | 0.57  | 0.57  | 0.57  |
| Mean ( $k_2s$ )     | 2.58  | 1.77  | 1.45  | 1.30  | 1.21  | 1.18  | 1.12  |
| Lower bound ( $L$ ) | 7.42  | 8.23  | 8.55  | 8.70  | 8.79  | 8.82  | 8.88  |
| Upper bound ( $U$ ) | 12.58 | 11.77 | 11.45 | 11.30 | 11.21 | 11.18 | 11.12 |

repeating data pattern. If multiple data are collected within the critical lag distance for data analysis, then the behavior and the characteristic of the entire data set cannot be properly represented. In other words, the measurement must be taken at the interval of critical lag distance to ensure data independence and to avoid data similarity. The critical lag distance is approximated at where the ACF values start to vary between the dashed lines which specifies a 95% CI of data independence.

To simulate the distribution of  $k_2$  as a function of the number of samples, we randomly sampled  $N_s$  samples from the observed data vector, repeating the experiment 1000 times for each value of  $N_s$  in Table 3. The last column in this table shows the results for  $N_s = N = 946$ . For any given  $N_s$ , the  $k_2$  for 95/95 TI can be calculated from Eq. (10) and appear in the third row of Table 3. The mean value of sample standard deviation,  $s$ , listed in the fourth row slightly increases with  $N_s$  and then remains constant for  $N_s$  greater than 20.

The relationship between the sample and population variance is expressed as [24]

$$s^2 = \left(1 - \frac{1}{N}\right)\sigma^2 \quad (12)$$

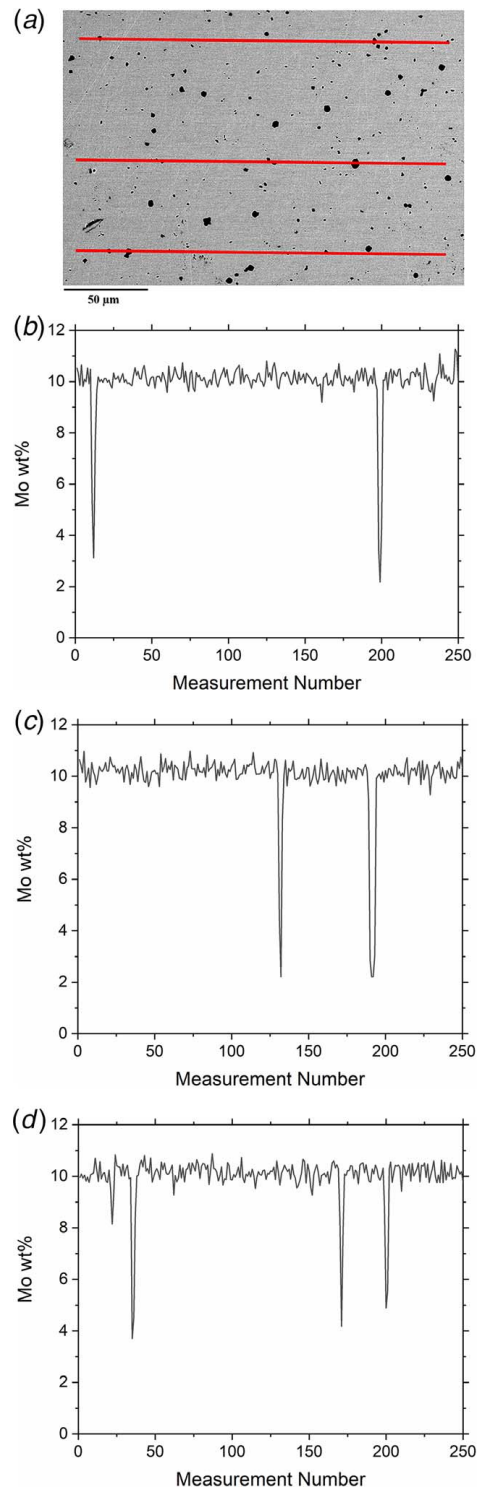
Equation (12) shows that sample variance will gradually increase and approach the population variance (constant) with increasing  $N$ —our findings are consistent. The next row of Table 3 shows the mean value of  $k_2s$  (multiplication of  $k_2$  and  $s$ ), which decreases with increasing  $N_s$ . The last two rows list the corresponding lower and upper bounds for each sample size  $N_s$ .  $L < 9\%$  and  $U > 11\%$  are observed for all cases, which indicates that this plate will not meet specified qualification and the molybdenum spatial distribution is not sufficiently homogeneous.

Next, Fig. 7 shows a second example plate (Plate 2) with its SEM and three EDS lines (250 measurements on each solid line). The statistics for varying  $N_s$  on plate qualification for Plate 2 is listed in Table 4. The minimum value of  $N_s$  to make  $L > 9\%$  and  $L < 11\%$  and reach the qualification requirement is 10 for Plate 2. The corresponding column is highlighted in bold. Therefore, the molybdenum distribution in Plate 2 is determined to be sufficiently homogeneous and hence qualified for the subsequent fabrication processes.

A spatial independence study using ACF is also conducted for Plate 2. The critical lag distance for both with and without particles scenarios is similar and close to  $5 \mu\text{m}$  (Fig. 8). The ACF results are in line with the observation from Fig. 7, which shows no clear pattern of data distribution except white noise.

#### 4 Recommendation on the Minimum Number of Molybdenum wt% Measurements to Determine Fuel Plate Qualification

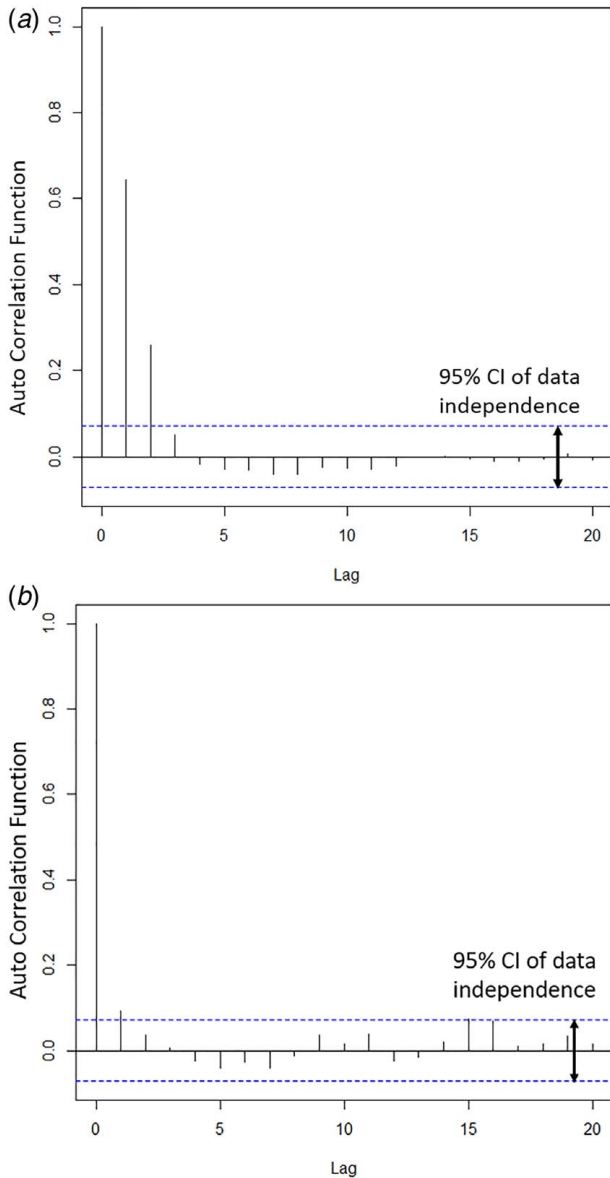
It was shown in the second example that ten measurements are already sufficient for Plate 2 to be qualified. However, this number can be different for other areas within a plate (or just a different plate) with different degrees of homogenization. Next, we provide a general recommendation for the optimal number of measurements to determine fuel plate qualification if no available measurement data exist in advance. As mentioned earlier, Eq. (10)



**Fig. 7 (a) SEM image (b) with top, (c) middle, and (d) bottom EDS line scans for Plate 2**

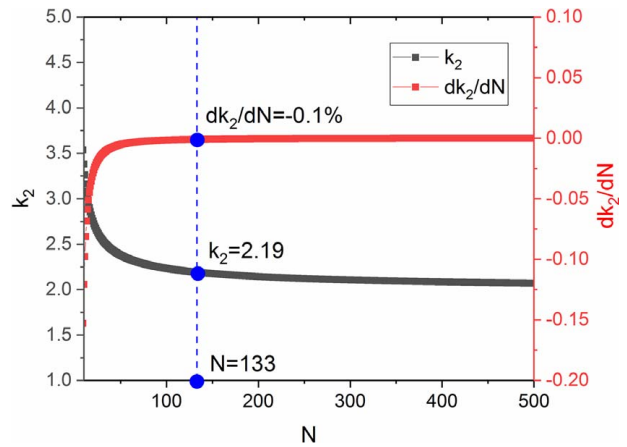
**Table 4 Impact of the sample size on plate qualification for Plate 2**

| $N$                 | 5     | 10           | 20    | 50    | 100   | 200   |
|---------------------|-------|--------------|-------|-------|-------|-------|
| Repetition          | 1000  | <b>1000</b>  | 1000  | 1000  | 1000  | 1000  |
| $k_2$               | 5.09  | <b>3.38</b>  | 2.75  | 2.37  | 2.23  | 2.14  |
| Mean ( $s$ )        | 0.26  | <b>0.28</b>  | 0.28  | 0.29  | 0.29  | 0.29  |
| Mean ( $k_2s$ )     | 1.36  | <b>0.96</b>  | 0.78  | 0.69  | 0.64  | 0.62  |
| Lower bound ( $L$ ) | 8.64  | <b>9.34</b>  | 9.22  | 9.31  | 9.36  | 9.38  |
| Upper bound ( $L$ ) | 11.36 | <b>10.96</b> | 10.78 | 10.69 | 10.64 | 10.62 |



**Fig. 8 ACF results for Plate 2 (a) with and (b) without particles**

indicates that  $k_2(N, \gamma, P)$  is only a function of sample size  $N$  with fixed  $P = \gamma = 0.95$ . A plot of  $k_2$  versus  $N$  in Fig. 9 shows that  $k_2$  gradually decreases (lower curve) but its slope slowly increases (top curve) with the increasing  $N$ . The value of  $k_2$  eventually approaches an asymptote of  $\sim 2$ . A gradient of  $k_2$  greater than  $-0.1\%$  can be considered as a mathematical indication of stability. The corresponding value  $N$  at  $dk_2/dN = -0.1\%$  is 133, and the associated measurement time ( $\sim 2$  h) is experimentally affordable. Therefore, it is reasonable to select  $N = 133$  as the suggested



**Fig. 9 Relationship between  $k_2$  and its gradient with  $N$**

number of measurements to determine the fuel plate qualification, resulting in  $k_2 = 2.19$ .

Based on these findings, a suggested procedure of data collection is described as follows to make sure that the selected data are not autocorrelated and are independent:

- (1) Collect the initial 133 EDS line-scan data with a normal  $1 \mu\text{m}$  spacing between each measurement.
- (2) Conduct the ACF analysis and then determine the critical lag distance.
- (3) Determine the number of valid collections among the original 133 data and then make additional collections based on the critical lag distance obtained from step 2.
- (4) Discard the measurements if the molybdenum weight percentage is outside the range of 7–13% to avoid the influence of carbides.

For any given fuel plate that meets the above-mentioned criterion, it can be considered as a qualified piece and be supplied to the subsequent processes only if all 133 collected data satisfy  $s \leq 0.46$ , which is equivalent to  $k_2s \leq 1$ . Otherwise, the plate is disqualified.

## 5 Conclusion

Characterizing the quality of U-Mo in casting is important and also challenging. A statistical approach based on the concept of TI is implemented to determine fuel plate qualification. This study also recommends a minimum number of required molybdenum wt% measurements to represent the distribution behavior of molybdenum for the entire fuel plate when other data are not available to determine the number of samples. Then, the given qualification specification can be simplified using only sample variance under reasonable assumptions. In particular, for the fuel plates discussed here, at least 133 data points across the line-scans are sufficient. Data points wherein the line-scan intersects a carbide or a secondary phase particle discarded. The collection of data must be independent without any correlation to assure that the collected measurements are a valid representation of the entire fuel plate. For sample standard deviation values equal to or less than 0.46 and means equal to 10 wt% molybdenum the given plate will ideally be qualified, whereas for standard deviation values higher than 0.46 the material is not qualified. By monitoring the fuel plate quality using the proposed method, the efficiency of the plate qualification is greatly enhanced, and fabrication costs are reduced. The developed methodology can be extended to other fields and serve as a metric for quality control.

## Acknowledgment

The authors would like to thank Shelley Carlson, Mark Rhodes, and Alan Schemer-Kohn for helping with the metallography

samples and capturing the SEM images. Pacific Northwest National Laboratory is operated by Battelle Memorial Institute for the United States Department of Energy under Contract DE-AC06-76RLO1830.

## Funding Data

- U.S. Department of Energy (Funder ID: 10.13039/100000015).
- National Nuclear Security Administration's Office of Material Management and Minimization (Funder ID: 10.13039/100006168).

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