A polynomial chaos expansion based molecular dynamics study for probabilistic strength analysis of nano-twinned copper

2016 Mater. Res. Express 3 036501

(http://iopscience.iop.org/2053-1591/3/3/036501)

Materials Research Express

RECEIVED
9 December 2015

REVISED
5 February 2016

ACCEPTED FOR PUBLICATION
12 February 2016

PUBLISHED
7 March 2016

PAPER

A polynomial chaos expansion based molecular dynamics study for probabilistic strength analysis of nano-twinned copper

Avik Mahata[1], **Tanmoy Mukhopadhyay**[2,3] **and Sondipon Adhikari**[2]

[1] Department of Aerospace Engineering, Indian Institute of Science, Bengaluru, India
[2] College of Engineering, Swansea University, Swansea, UK
[3] Author to whom any correspondence should be addressed

E-mail: 212212020@nitt.edu (A Mahata), 800712@swansea.ac.uk (T Mukhopadhyay) and S.Adhikari@swansea.ac.uk

Keywords: molecular dynamics, nano-twinned copper, probabilistic analysis, polynomial chaos expansion, D-optimal design

Abstract

Nano-twinned structures are mechanically stronger, ductile and stable than its non-twinned form. We have investigated the effect of varying twin spacing and twin boundary width (TBW) on the yield strength of the nano-twinned copper in a probabilistic framework. An efficient surrogate modelling approach based on polynomial chaos expansion has been proposed for the analysis. Effectively utilising 15 sets of expensive molecular dynamics simulations, thousands of outputs have been obtained corresponding to different sets of twin spacing and twin width using virtual experiments based on the surrogates. One of the major outcomes of this work is that there exists an optimal combination of twin boundary spacing and twin width until which the strength can be increased and after that critical point the nanowires weaken. This study also reveals that the yield strength of nano-twinned copper is more sensitive to TBW than twin spacing. Such robust inferences have been possible to be drawn only because of applying the surrogate modelling approach, which makes it feasible to obtain results corresponding to 40 000 combinations of different twin boundary spacing and twin width in a computationally efficient framework.

1. Introduction

It is widely believed that the fundamental properties of any material such as strength, ductility, creep, fracture behaviour, durability etc differs widely at nanoscale compare to its bulk specimen [1–7]. With the progress in microscopic equipment and following the requirement in industry to manipulate material properties at nanoscale, it is now a reality to tailor material properties specific to its application [8–10]. One type of nanoscale materials which can be produced by severe plastic deformation that provide high strength by creating barriers for intergranular dislocation is generally known as nanocrystalline material [11, 12]. Here the smaller grain size limits the scope of intragranular dislocation motion. Nano-twinned materials fall in the same category of material where in place of grain boundaries (GBs), few nanometer wide lamelle provides resistance to the crack growth [13–15]. Twins are very common in various metals, alloys, composite with different crystallographic structures [16–19]. Extremely thin twin lamellae structures can possibly be achieved under proper conditions during crystal growth, plastic deformation, phase transformations, or thermal annealing of deformed structures. With the extreme advancement of microscopic instruments in recent years it is also possible to manufacture any kind of desired specimen at nanoscale.

Copper is one of the most studied materials both experimentally and computationally in the context of nano-twinning, where it is found that the strength in tension or compression increases substantially than the non-twinned copper nanowire [19–23]. It has been reported that the decreasing twin spacing and increasing width of twin boundaries results into the increase in strength of nano-twinned copper wires [24, 25, 30]. The experiments and molecular dynamics simulations conducted by Jang *et al* [26] to investigate the influence of diameter, twin-boundary spacing and twin-boundary orientation on the mechanical responses of individual nanopillars, shows a quantitative comparison indicating that the strength of nanotwinned pillars depends on

various internal parameters, including coherent twin boundary spacing (CTBS), twin boundary width (TBW) and twin-boundary orientation. However it also significantly depends on surface morphology (i.e. circular or square cross section) [27, 28]. TBW is basically the width of the specimen. In our model of nanotwinned copper we can find two different boundaries: one is for the twins, which is in the vertical direction and other one is in the horizontal direction, which is the grain boundary. The length between two GBs can also be described by the TBW. In other words we can say that each of the specimens is a nanotwinned grain. For nanowires with square cross-sections, strength increases as twin spacing decreases. In contrast for nanowires with circular cross-sections, strength varies slightly with twin spacing, being marginally lower than that in single crystals. The fact, whether twin boundaries strengthen a metallic nanowire or not, depends on the necessary stress required for dislocation nucleation, which in turn depends on the surface morphology of the nanowires [27]. Deng *et al* studied the evolution of the critical resolved shear stress as a function of number of twins per unit length and size effect on Au-nanowires with diameters of 8.2 and 24.6 nm at 300 K [28, 29]. It was found that addition of CTBS to crystalline nanowires does not always have strengthening effects but also it has weakening effects and no effects, which depends on both twin spacing and sample diameter [28, 30]. Size effect on tilted twin boundaries was studied by Cao *et al* [23] using molecular dynamics and finite element analysis. It shows that the one-dimensional coherent nanostructures could help to develop straight forward understanding on the origin of size effect in strength. Another recent study on molecular dynamics simulation of uniaxial compression in twinned nanopillars elucidates the interplay of intrinsic (twin boundary spacing) and extrinsic (pillar diameter) size-scale effects on the strengthening mechanisms [25]. An intriguing revelation is that there is an optimal aspect ratio for which the yield strength of twinned nanopillars is higher than single crystal nanopillars, in contrast to other aspect ratios for which the yield strength of twinned nanopillars are considerably lower than their single crystalline counterparts. A crucial limitation of all the studies mentioned above is that these investigations are based on limited number of simulations because molecular dynamics simulations are computationally very expensive. Moreover it is a time consuming simulation method where the time scale is inherently extremely low (~pico or femoto second). It is practically impossible to carryout adequate number of molecular dynamics simulations for characterizing the material responses and associated influential factors comprehensively. In case of actual nano-scale experiments, the problem becomes even more aggravated due to the complexity involved and its highly expensive nature.

In this article we propose an alternative surrogate based approach, where adequate number of samples can be analysed in a computationally efficient way to characterize a material in its full inherent complexity by carrying out minimal number of actual molecular dynamics simulations/experiments. Here we have applied polynomial chaos expansion (PCE) for surrogate model formation to analyse nano-twinned copper. D-optimal design algorithm has been utilised to select the optimal points from the entire design space for surrogate model formation. Subsequently probabilistic analysis has been carried out for yield stress of the material. Probabilistic methods are successful in various areas of science and engineering for comprehensive analysis of a system. Such analyses are important because every practical system is inherently stochastic, thus conventional deterministic approach cannot characterize a system comprehensively in the true sense. A careful review on probabilistic analysis in the realm of nano-scale researches shows that the state of application of probability theory in molecular dynamics simulation [31–35] or in the broader aspect of multi-scale modelling is very scarce [35, 36]. One of the obvious reasons being the fact that molecular dynamics simulation is relatively a new area of research and it has received more attention so far in deterministic analysis. Along with molecular dynamics/multiscale modelling stochasticity and uncertainty can also be found in nanowire experiments as well, experimental evidence was illustrated that micropillar deformation is highly stochastic [37], a stochasticity-enhanced gradient plasticity was able to model the stochastic stress–strain response of same-diameter micropillars. Incorporation of stochasticity in the analysis is the most logical extension of research in this field to understand the process from a much wider and more robust perspective. Another crucial reason is the computational expense of molecular dynamics simulations. For carrying out probabilistic analysis using Monte Carlo simulation, results are needed to be obtained for thousands of input parameter points. For that purpose, carrying out molecular dynamics simulation thousands of times is practically impossible due to its computational intensiveness. The application of surrogate modelling approach, as proposed in this article is the only efficient alternative in such situation. Moreover, utilisation of surrogates can capture the material behaviour more precisely in case of deterministic analysis, as responses can be obtained corresponding to any value of input parameters within the design space in a much more computationally efficient manner compared to original molecular dynamics simulation carried out for few selective input points. Application of the proposed technique enables us to predict the strength of nanowires within a continuous domain of varying twin boundary spacing and TBW in a probabilistic framework obtaining a complete map between the design parameters and response to find an optimum design combination for highest strength of nano-twinned copper. This article is organized as, section 1: introduction; section 2: brief description about surrogate modelling approach using PCE; section 3:

simulation methodology for nano-twinned copper using molecular dynamics simulation coupled with PCE; section 4: results and discussions; section 5: conclusion.

2. Polynomial chaos expansion

The PCE is an effective tool for solving stochastic systems. It was first introduced as the homogeneous chaos by Wiener [38]. The basic idea is to project the random variables of problem onto a stochastic space spanned by a set of complete orthogonal polynomials. The orthogonal polynomial chaos basis functions, derived from Gram–Schmidt algorithm [39, 40] is employed in this study for mapping input–output relation. Solution to generalised equation at a random space can be expanded into a PCE as follows:

$$\mathbf{y} = \mathbf{B}\,\psi(\boldsymbol{\xi}), \tag{1}$$

where $\mathbf{y} = [y_1 \;\; y_2 \;\ldots\; y_n]^{\mathrm{T}} \in \Re^{n\times 1}$ denotes the assembled vector of output data, $\psi(\xi) = [\psi_0(\xi) \;\; \psi_1(\xi) \;\ldots\; \psi_p(\xi)]^{\mathrm{T}} \in \Re^{p\times 1}$ denotes the assembled vector of polynomial chaos basis functions and **B** is expressed as

$$\mathbf{B} = \begin{bmatrix} \beta_0^{(1)} & \beta_1^{(1)} & \beta_2^{(1)} & \ldots\ldots\ldots\ldots & \beta_p^{(1)} \\ \beta_0^{(2)} & \beta_1^{(2)} & \beta_2^{(2)} & \ldots\ldots\ldots\ldots & \beta_p^{(2)} \\ \beta_0^{(3)} & \beta_1^{(3)} & \beta_2^{(3)} & \ldots\ldots\ldots\ldots & \beta_p^{(3)} \\ \vdots & \vdots & \vdots & & \vdots \\ \beta_0^{(n)} & \beta_1^{(n)} & \beta_2^{(n)} & \ldots\ldots\ldots\ldots & \beta_p^{(n)} \end{bmatrix}, \tag{2}$$

where $\beta_k^{(i)}$ are the coefficients of polynomial expansion with $k = 1,2,3\ldots.p$ (p is the number of terms retained in the expansion), n is the number of output parameters, ξ is an m-dimensional vector of variables and 'm' is the number of input parameters. Gram–Schmidt algorithm provides the opportunity to derive the polynomial chaos basis functions for any arbitrary probability distribution on '$\boldsymbol{\xi}$'. In this method, the polynomial terms are represented as $\psi_j(\xi_i) = \xi_i^j + O(\xi_i^{j-1})$ where $j = 0,\;\; 1,\ldots,\; h$. This results in $\psi_0(\xi_i) = 1$ and the remaining terms are computed using the following recursive equations:

$$\psi_j(\xi_i) = e_j(\xi_i) - \sum_{k=0}^{j-1} c_{jk}\psi_k(\xi_i), \tag{3}$$

$$\text{where } c_{jk} = \frac{\int_{\text{supp}\xi_i} e_j(\xi_i)\psi_k(\xi_i)p(\xi_i)\mathrm{d}\xi_i}{\int_{\text{supp}\xi_i} \psi_k^2(\xi_i)p(\xi_i)p(\xi_i)\mathrm{d}\xi_i}.$$

The lower and upper bounds of input variables (i.e., $p_i^{(\mathrm{L})}$, $p_i^{(\mathrm{U})}$) can be transformed into the normalised values of −1 and 1, respectively and thus a transformation function $\varphi(\bullet)$ for any intermediate value in the design domain can be obtained as:

$$\xi_i = \varphi(p_i) = 2\left(\frac{p_i - p_i^{(\mathrm{L})}}{p_i^{(\mathrm{U})} - p_i^{(\mathrm{L})}} - \frac{1}{2}\right), \tag{4}$$

$$p_i = \varphi^{-1}(\xi_i) = \frac{(p_i^{(\mathrm{U})} - p_i^{(\mathrm{L})})}{2}\xi_i + \frac{(p_i^{(\mathrm{U})} + p_i^{(\mathrm{L})})}{2}, \tag{5}$$

where ξ_i is the transformed value in domain $[-1, 1]$ corresponding to p_i in the domain $[p_i^{(\mathrm{L})}, p_i^{(\mathrm{U})}]$ for ith input parameter ($\boldsymbol{\xi} = [\xi_1, \xi_2, \ldots, \xi_m]^{\mathrm{T}} \in \Re^{m\times 1}$).

Depending on the type of the input parameter, a set of input variables are selected to obtain the output quantity of interest. In the present study D-optimal design has been utilised to select the design points from the input parameter space [41–43]. D-optimal design is one of the most efficient design of experiment algorithms requiring fewer samples than standard design procedures and thus it needs much lesser number of molecular dynamics simulations/experiments to be carried out for forming the surrogate model. In this design, position of design points is chosen algorithmically according to the number of factors and the desired model to meet the optimality criteria. Optimal designs can be used to create a good design for fitting a linear, quadratic, cubic or higher order models. There can be several types of optimality criteria such as D-optimality, A-optimality and E-optimality. Let X denotes the design matrix as a set of value combinations of coded parameters and X^t is the transpose of X, then D-optimality is achieved if the determinant of $(X^tX)^{-1}$ is minimal. A-optimality is achieved by minimising the trace of $(X^tX)^{-1}$. E-optimality is achieved if the largest eigenvalue of $(X^tX)^{-1}$ is minimal. Among these, D-*optimal* design is the most commonly used owing to better accuracy of approximation than

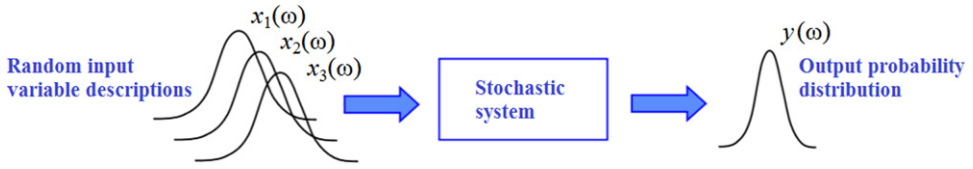

Figure 1. Schematic representation for Monte Carlo simulation based analysis of a stochastic system with three input parameters (x_i, $i = 1, 2, 3$) and one output parameter (y). Here ω represents the stochastic character of a parameter.

others [42]. In D-optimal design, the total sample size (n) comprises of minimum design points (n_d), additional model points (n_a) and lack-of-fit points (n_l) i.e., $n = n_d + n_a + n_l$. The required model points (i.e., minimum design points) is the minimum number of samples to estimate the coefficients for model formation while additional model points are extra samples added by the user to improve precision estimates or coverage of the factor space and lack-of-fit points are the extra points to fill the factor space. The extra information provided by these points can test the fit of the model.

3. Simulation methodology

This section describes the PCE based molecular dynamics simulation approach for analysing strength of nano-twinned copper. We have chosen few efficient and optimal samples (design points) within the design space for molecular dynamics simulation of nano-twinned copper using D-optimal design algorithm. In this study, the twin spacing and twin width has been taken as two input parameters and subsequently their effect on yield strength of the material is investigated. We have chosen a nano-twinned sample of 30 nm length for the present analysis with the design space for CTBS and TBW in the range of 1 nm–10 nm and 5 nm–20 nm respectively, considering practical applications for nano-twinned copper. The ratio of twin width and length of the nano-twinned specimen has been regarded as aspect ratio in this study. Another noteworthy aspect is that the strength varies with the type of cross section selected for simulation, as mentioned in section 1. Thus, the results presented here are pertaining to the strength of nano-twinned materials of rectangular/square cross section which has been considered in this investigation. As discussed in section 1, there are other factors that can influence the strength of nano-twinned copper but in the present research, we have concentrated on twin spacing and TBW only because these are the two most dominant factors to decide strength of nano-twinned specimens. It should be noted that other factors that affect mechanical properties of any nanowires can also be analysed using the proposed method in future.

In this study, probabilistic analysis has been carried out for yield strength of nano-twinned copper using Monte Carlo simulation following a non-intrusive approach. Monte Carlo methods (or Monte Carlo experiment/simulations) are a broad class of computational algorithms that rely on repeated random sampling following a particular probability distribution to obtain numerical results. These methods are often used in physical and mathematical problems and are most useful when it is difficult/impossible to use direct mathematical algorithms. Figure 1 presents a schematic representation for a general stochastic system having three random input parameters and one output response. In general, though Monte Carlo based analyses are capable of obtaining comprehensive results for a physical/mathematical problem, these are computationally very expensive. These methods normally require thousands of simulations/experiments (molecular dynamics simulation in the present study) to be carried out corresponding to random input sets. Thus the entire process becomes exorbitantly cost-intensive, especially for problems where individual simulations/experiments are very costly and time consuming such as molecular dynamics simulations. To overcome this difficulty we have employed the PCE model as a surrogate of the actual molecular dynamics code to carry out Monte Carlo simulation. The entire process of surrogate based analysis of nano-twinned copper is depicted in figure 2. After selecting the design points using D-optimal design algorithm, the next step is to obtain corresponding yield stresses (output) for each of the design points using molecular dynamics simulation. Representative values of CTBS and TBW selected for PCE model formation in the present analysis and corresponding yield strengths are shown in table 1. Once the design matrix is formed, the PCE model is constructed as discussed in section 2. The PCE model can henceforth be used as a surrogate of the actual molecular dynamics simulation, thereby it can be said that the PCE model effectively replaces computationally expensive molecular dynamics simulation process. Thus the PCE model is capable of obtaining yield strength corresponding to any set of values of twin spacing and twin width within the considered design space. Details of the molecular dynamics simulations (corresponding to the 15 design points to construct the PCE model) carried out in this study are discussed next.

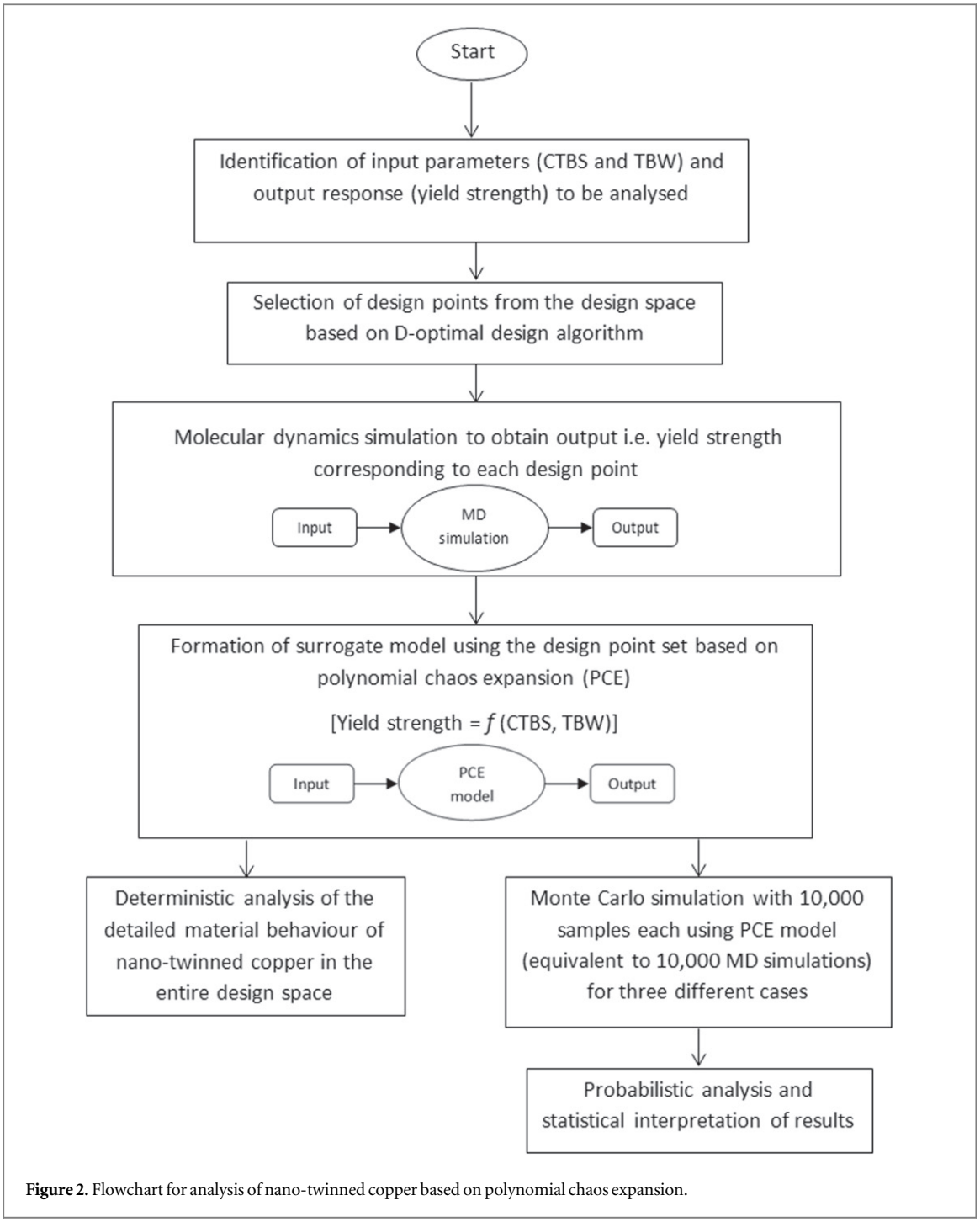

Figure 2. Flowchart for analysis of nano-twinned copper based on polynomial chaos expansion.

Table 1. Typical representation of design points for formation of PCE model (CTBS, TBW, aspect ratio are the three input parameters, while yield strength is the output parameter of interest in the present analysis).

Sample	CTBS (nm)	TBW (nm)	Aspect ratio	Number of atoms	Number of twins	Yield strength (GPa)
1	3.25	19.325	1.56	197 096	9	3.406
2	10	11.75	2.55	160 000	2	3.307
3	7.19	15.8	1.89	163 872	4	2.256
			⋮			
15	5.95	5	6	54 915	5	5.268

In general, a length of 30 nm and a constant thickness of 8 times the dimension of unit cell (for samples of size smaller than four unit cell introduce a separate size scale effect in fixed-end simulations, as our sample is thicker than four unit cell we avoid the size effect) have been considered in this analysis. Simulations have been performed for nano-twinned specimens using the embedded-atom-method potential for copper developed by

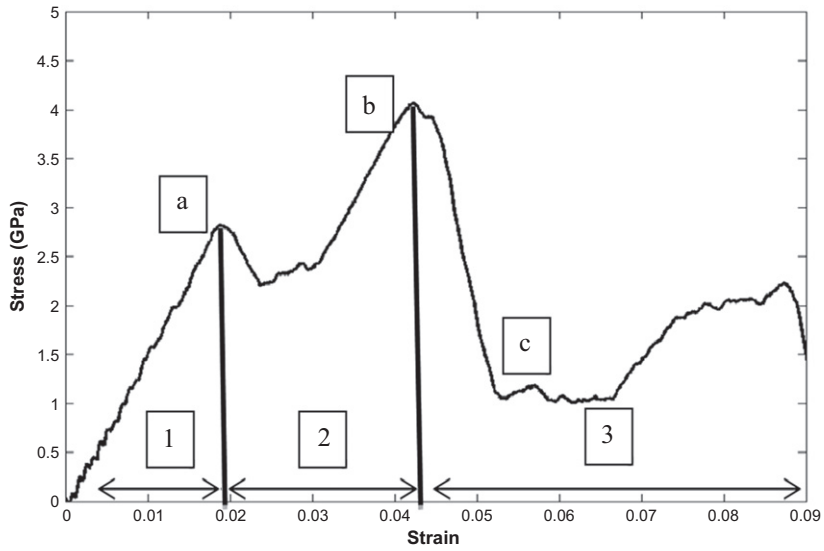

Figure 3. Tensile stress–strain curve of a periodically twinned Cu-nanowire (1-represearnts the elastic part of the curve, 2-represents the strain hardening part and 3-is strain softening where the material finally fails). Snapshots at different stages of deformation are shown from (a) to (c) in figure 4.

Mishin *et al* [44]. The specimens are considered to be aligned along the $[1\bar{1}0]$, $[11\bar{2}]$ and [111] (*x*-, *y*- and *z*-direction for the simulation). The CTBs are aligned normal to the [111] or *z*-direction. The twin boundary creates a mirror image of each other, which is similar to previous experimental and simulation works on this topic [23–29]. All the simulations in the present study have been carried out at 300 K. Periodic boundary conditions have been applied in the $[1\bar{1}0]$ (*x*-direction) direction and non-periodic boundary condition has been applied in rest of the direction. The simulations have been performed in a NVT ensemble (fixed number of atoms, volume and temperature). The free movement of twinning and dislocation can be captured better in non-periodic boundaries as it activates more slip planes favourable to the dislocation and twinning [15]. However our prime focus of the present investigation is on the yield strength of the specimens rather than studying the dislocation/twin movement of the material. Typical dimensions and number of atoms considered for simulation corresponding to the design points are presented in table 1. In the present analysis, the structure has been deformed by applying a constant tensile engineering strain rate of 10^8 s^{-1} along the [111] direction (or in *z*-direction). The top and bottom few layers were kept fixed and were not part of the simulation. The bottom fixed layer kept the nanowire fixed at a plane and the strain rate have been applied by moving the top few layers moving at a constant velocity of 0.03 Å ps^{-1}, which results into the strain rate of 10^8 s^{-1}. The inherent strain rate in any molecular dynamics simulation is very high due to extremely small length and timescale. 10^8 s^{-1} comparatively slow strain rate where the deformation takes place very slowly, in earlier studies of nanotwinned structures of fcc metals this sort of strain rate have been used by Sinha *et al* [21]. At this strain rate we can capture the deformation twinning very accurately and also get a proper value of yield strength of the nanowire, which is the main focus of our work. Atomic positions, velocities and accelerations are updated at each time step using the Velocity-Verlet algorithm [45] with a time-step of 0.001 ps. The high strain rate is inherent to every molecular dynamics simulation, and necessary to obtain a significant amount of deformation within a reasonable simulation time. All the simulation has been performed in LAMMPS parallel code [46] and visualised by OVITO [47] and Atomeye [48].

4. Results and discussions

4.1. Deformation mechanism

In this section we discuss the inherent deformation mechanism of nano-twinned copper in general to put the present research on probabilistic stress analysis in proper context with respect to the earlier descriptions of material behaviour. For all the molecular dynamics simulations that have been performed in this study, the stress–strain plots are found to be similar to each other in nature. A typical stress–strain curve indicating different stages in the deformation process for nano-twinned copper is shown in figures 3 and 4. The initial part of the stress–strain plot is linear and follows Hook's law where the material is under elastic limit. In this stage (up to point (a) of figure 3) the stress required for deforming the material increases linearly. No defect in the nano-

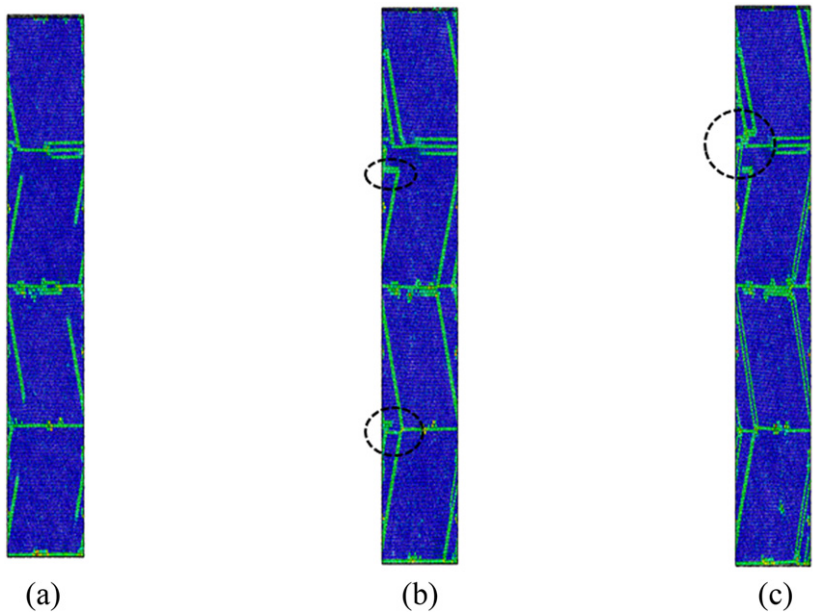

Figure 4. (a) Nucleation of the very first dislocation at initial yield stress. (b) Leading partial dislocations stopped by twin boundaries. (c) Nanowire deformation after the maximum flow stress. Atom colours correspond to the crystal structure. Dashed circles in (b) and (c) show the emission of trailing partial dislocations from the free surface.

twinned copper is formed during this stage. After point (a), the continuous increase of stress initiates defects, resulting the required stress for deformation to drop. As the material behaviour turns into plastic, defects interact with each other and strain hardening occurs due to generation and pile up of dislocations at barriers like GBs during the deformation. After the dislocation pile up and twin formation reaches a critical limit, continuous supply of distortion energy through strain results in breakdown of the pile up barrier and slip on a new plane. The breakdown of dislocation and twin pile up barrier causes flow softening that continues until beginning of the second stage of dislocation pile up. This stage can be observed after point (b) in figure 3. As the number of dislocations and twins increase after this point causing subsequent increase in dislocation and twin density, dislocation propagation becomes more difficult (implying the application of larger load to realise the same deformation as an earlier stage of plastic zone). The initial twinning is created in the form of partial dislocation on edges of the simulation box. Successive progression of the partial dislocations occurs from the edge of the simulation box to the coherent twin boundaries, where it requires more energy to cross the hurdle and thus require more strength to deform. This part of the mechanism can be described as the strain hardening. The figure confirms that the onset of plasticity in copper nanowires is associated with the emission of $\{11\bar{1}\}\langle 112\rangle$ partial dislocations from the free surface. However, there are several other mechanisms of dislocation CTB interaction (Mechanisms I–III as mentioned by Deng and Sansoz [49]) that can be observed in the fcc copper nanowires at the initial yield point, depending on the potential. Similar deformation mechanism, referred Mechanism II by Deng and Sansoz [49] has been observed in the present study.

A typical example of atomic-scale interactions between a leading partial dislocation and a CTB in the copper nanowires at yield point is furnished in figures 3 and 4. Here, the leading partial dislocation (γD) from the parent grain directly transmits through the CTB via the reaction, $\frac{1}{6}(11\bar{1})[\bar{1}\,\bar{1}\,\bar{2}] = \frac{1}{3}(112)[11\bar{1}] + \frac{1}{2}(001)[\bar{1}\,\bar{1}0]$, where $\frac{1}{2}(001)[\bar{1}\,\bar{1}0]$ a full dislocation is transmitted in the twin grain, and $\frac{1}{3}(112)[11\bar{1}]$ represents a sessile stair-rod dislocation remaining in the CTB plane. However, once the twins cross the CTB, strength keeps dropping till failure (refer figure 3, point c).

As mentioned earlier, the key processes observed during the deformation of the nanotwinned models include partial and full dislocation emission from the GBs. Along with that the other major influential factors in the deformation mechanism are CTB migration, interaction of the partial dislocations with fixed and migrating twin boundaries, formation of steps by the CTB (refer figures 5 and 6), and dislocation emission from the steps. In several previous researches [23, 49–56], the deformation behaviours have been investigated on CTBs in nano-crystalline aluminium and copper. Similar to these reports, dislocation nucleation along and across the CTBs, twin boundary migration and change in twin width, pile-up and passing of complete and partial dislocations across the CTBs, grain boundary sliding are found to occur in the present study during the deformation process of nano-twinned copper. One of the dominant mechanisms that have been observed in our simulations is twin boundary migration, similar to the observations by Frøseth *et al* [51] for nano-crystalline aluminium and also by

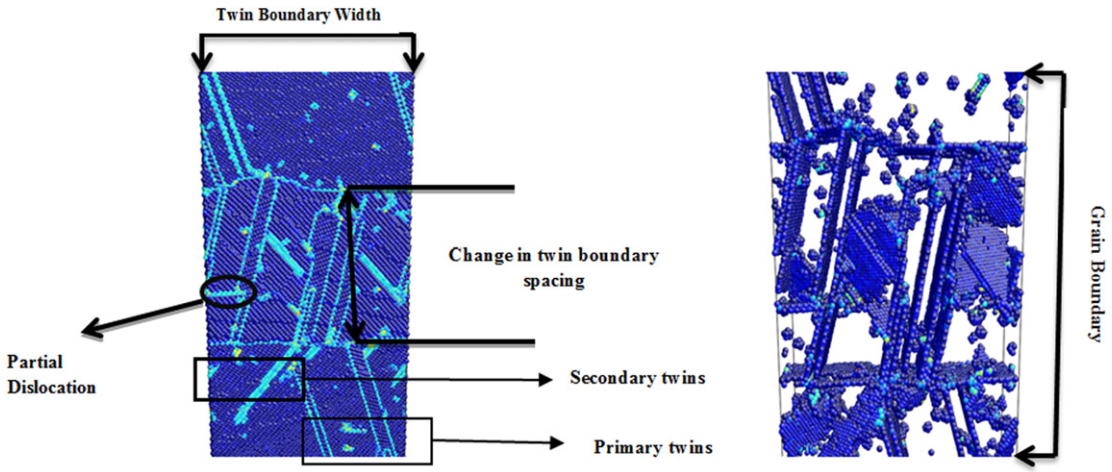

Figure 5. A close look to partial and complete disocation lines. Multiple dislocation lines result into twinning in nano-twinned copper. Change in twin boundary spacing while applying uniaxial tension has also been shown.

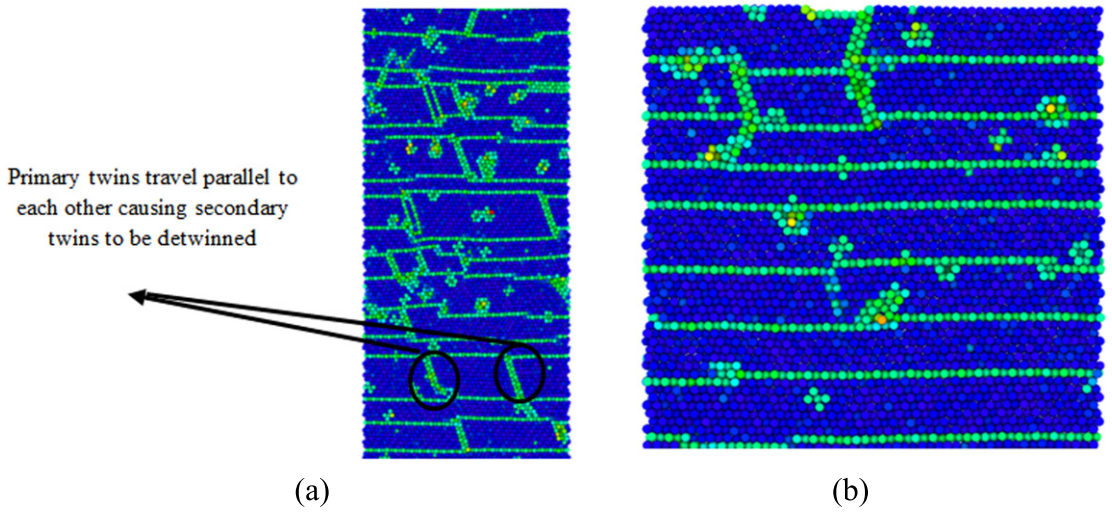

Figure 6. (a) Complete nanowire with multiple TBS of 10 nm. (b) Part of nanowire is shown, where the successive twins resist each other's movement throughout the sample.

Cao and Wei [23] for simulations on Copper. During the deformation process, some of the twin boundaries start to migrate to the neighbouring (1 1 1) planes. This migration process can be attributed to partial dislocations emitted from the GBs and twin boundary junction (figure 5). The migration process starts from the GBs and in few instances, progresses up to the other side of the grain. This shifts the entire CTB into the neighbouring (1 1 1) plane, as shown in figure 5. In this study, we have taken twin spacing (as shown in figure 5) as one of the parameters, wherein we can notice that the twin boundary spacing varies by few Angstrom as the deformation progresses. It may allow some of the partial dislocations to grow or stretch. In the present analysis, actual molecular dynamics simulations on nano-twinned copper have been carried out for only 15 different algorithmically chosen samples. It has been noticed that even though the number of dislocations, staking fault or grain boundary migration differ from sample to sample causing variation in yield strength but the basic mechanism of deformation in all the samples is similar.

4.2. Material response

In this section we will discuss about detailed deterministic analysis of the material behaviour in the entire design space. As mentioned in the previous sections, due to application of PCE based modelling approach we can readily obtain yield strength of a particular sample corresponding to any combination of CTBS and TBW. It is worthy to note here that the constructed PCE model has been checked extensively for its prediction capability

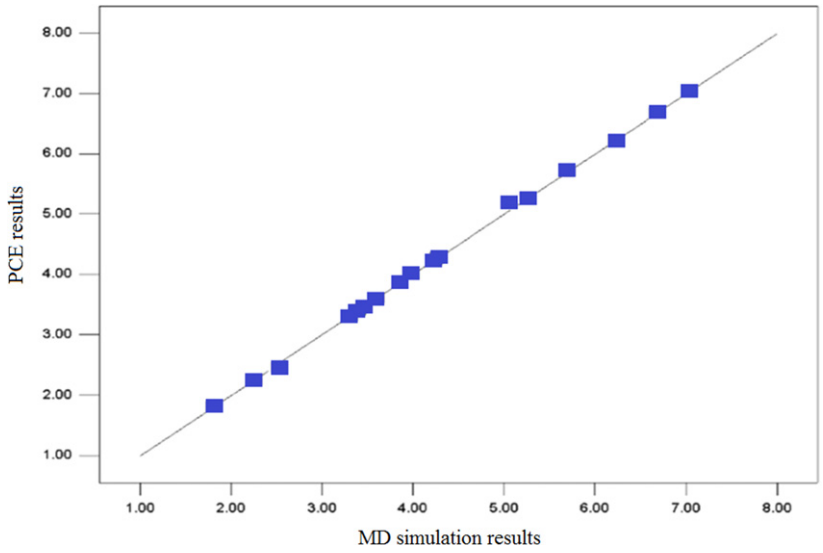

Figure 7. PCE model validation plot showing actual MD simulation results versus PCE results for yield strength (GPa).

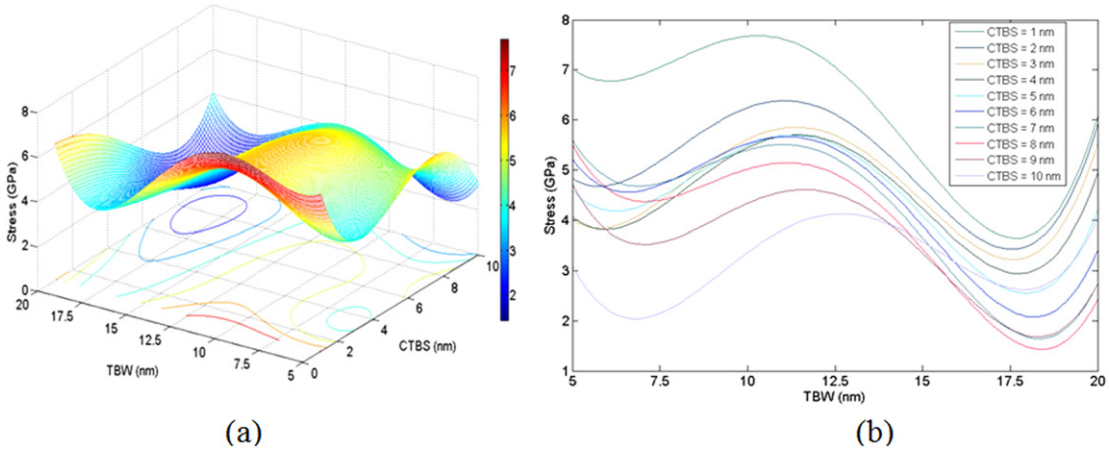

Figure 8. Variation of yield stress with twin boundary width (TBW) and coherent twin boundary spacing (CTBS).

with respect to actual MD simulation results using new sets of input parameters before using it for further analyses. Figure 7 shows the prediction capability of the constructed PCE model with respect to actual molecular dynamics simulation, wherein less deviation of the tested points from the diagonal line indicates high accuracy of the fitted model. Figure 8 shows the variation of yield strength of nano-twinned copper with CTBS and TBW. A detailed 3D response surface for yield stress has been constructed (figure 8(a)) using the predictions of PCE model to get a comprehensive idea of the effect of twin width and twin spacing. For better understanding, variation of yield strength with twin width has been plotted for different values of twin spacing in figure 8(b). To construct figure 8 we have chosen two arrays of data points containing 100 equidistant points each within the design space resulting total 100 $\times$ 100 combinations of CTBS and TBW. Here noteworthy is the fact that it has been practically possible to obtain yield strength for such a huge number of samples only due to application of surrogate modelling approach.

From figure 8 it is evident that yield stress of single crystal nano-pillars exhibits a strong size-dependence, which is quite in agreement with previous investigations [25, 57–59]. Width of the twin boundaries can be regarded as size in the present study. Plasticity is found to initiate with dislocations nucleating from the surface of the specimens. The size-dependence of the yield stress can be attributed to the fact that the critical stress for dislocation nucleation at the surface scales with the pillar width due to the constrained volume offered by the smaller pillars (in width) that makes it harder for the dislocations to nucleate. A general tendency is noticed that yield strength increases with the decrease in twin spacing. The variation of yield strength with CTBS can be explained by the fact that higher number of twin boundary (i.e. the smaller twin boundary spacing) works as an

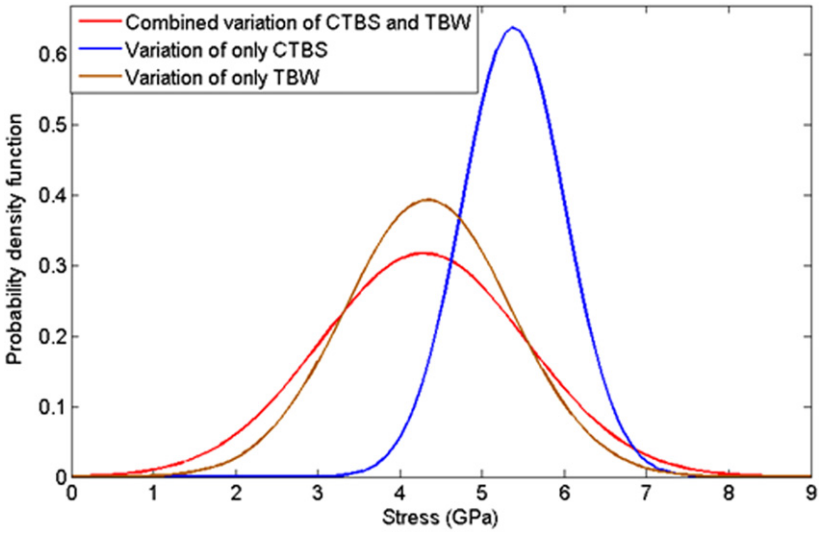

Figure 9. Probability density function plot of yield stress for variation of coherent twin boundary spacing (CTBS) only, twin boundary width (TBW) only and combined variation of CTBS and TBW.

obstacle for the dislocations to travel throughout the specimen, which in turn strengthens the specimen. The relation between yield strength and twin width is found to be not that monotonic in nature as compared to that of yield strength and twin spacing. From figure 8(b), it is clear that there is a strong dependence of CTBS in the relationship between yield strength and TBW. For a particular value of CTBS, we can notice peak strength of nano-twinned copper which grossly appears in the range of 10 nm to 13 nm TBW. The value of TBW, for which a nano-twinned copper exhibits maximum strength, increases with the increase in value of CTBS. Reason behind this correlation between CTBS and TBW is the complex dislocation/twinning mechanism during the deformation process. There exists an adverse effect on the strength of the nano-twinned structures due to interdependence between twins in two successive layers and creation of V shape (figure 6(b)) that resists each other's motion. This investigation reveals that there is a transition in dominating deformation mechanisms occurring at a critical spacing for which strength is maximum. Above that critical spacing, the deformation mechanisms are dominated by two Hall–Petch type strengthening mechanisms. One is partial dislocation emission from GBs travelling across twin boundaries and the other is partial dislocations emitted from twin boundaries travelling across other twin boundaries as shown in figure 6(b). But once it reaches the critical CTB spacing, it actually softens the specimen by partial dislocations emitted from boundaries of primary twins travelling parallel to each other and leading to detwinning of the secondary twins in the same twin boundary (refer figures 5 and 6). Along with that twin boundaries shift itself, leading to a variation in twin spacing (figure 6) making the deformation mechanism even more complex to understand.

Thus from the above discussion it is evident that strength of nano-twinned copper depends on several correlated factors and governed by various complex dislocation mechanisms. In the present analysis we have taken two important factors namely CTBS and TBW and investigated their individual and combined effect on strength of the material using a mathematical and statistical framework. Advantage of the proposed surrogate based approach is that, despite of the internal deformation mechanism of nano-twinned copper being considerably complex; it is possible to construct a fully functional mathematical model using only few molecular dynamics simulation. Such efficient mathematical model can then be used to predict responses corresponding to any combination of the design parameters in later stage of the analysis. Detail response curves have been constructed in this study using such model as shown in figure 8, a careful observation in which leads to the conclusion that there exists an optimum twin width for each twin spacing, for which yield strength of the nano-wire becomes maximum. This finding can be crucial in designing nano-twins for maximum strength.

4.3. Probabilistic analysis

After surrogate based deterministic analysis of the deformation mechanism and thus yield strength of nano-twinned copper along with its relation with two dominating factors (CTBS and TBW), Monte Carlo simulation has been carried out to study the probabilistic behaviour of yield stress. In the present analysis, the input parameter set for probabilistic investigation has been chosen following uniform distribution within the design space. Three different analyses have been carried out (using 10 000 samples in each case) to access their influence on yield stress of the material, namely random variation of CTBS only, TBW only and combined random

Table 2. Statistical analysis results for yield strength of nano-twinned copper.

Parameter	Maximum	Minimum	Mean	Standard deviation
Coherent twin boundary spacing (CTBS)	7.052 719	4.123 635	5.379 037	0.624 221
Twin boundary width (TBW)	5.693 076	2.549 342	4.340 363	1.012 765
Combined variation	7.651 505	1.429 41	4.287 826	1.255 636

variation of both CTBS and TBW. It is interesting to notice from the probability density function plots (figure 9) that in spite of the input parameters being chosen from uniform distribution, the response yield strength follows a Gaussian distribution in all three cases. This observation obeys the central limit theorem of probability theory that states that: given certain conditions, the arithmetic mean of a sufficiently large number of iterates of independent random variables, each with a well-defined expected value and well-defined variance, will be approximately normally distributed, regardless of the underlying distribution. Gaussian distribution of the yield strength implies that probability of occurrence increases as we approach towards the mean value of yield strength. The individual and combined effects of the stochastic input parameters, as quantified in this study, provide clear understanding about the statistical character and their relative influence on the probability distribution of yield strength. Having the probabilistic character of the response quantity of interest (yield strength) quantified, optimisation for CTBS and TBW can be carried out considering the inherent uncertainty of the system with a particular degree of confidence. The trade-off between the degree of confidence and the level of optimisation should be decided based on the required reliability of the system. Table 2 shows the response bounds, mean and standard deviation for yield strength corresponding to the three cases. From the table, normalised coefficient of variation (with respect to combined variation) for CTBS and TBW can be calculated as 0.332 and 0.668 respectively, whereby it can be concluded that TBW is more sensitive than CTBS to yield strength of the material. Same inference can be attributed from figure 9, where more dispersion is noticed in case of TBW compared to CTBS. The dispersion is more than TBW and CTBS in case of the combined variation, as expected.

5. Conclusion

This article presents a detailed deterministic investigation and probabilistic analysis on the strength of nano-twinned copper using PCE. In the proposed surrogate based approach, computationally expensive molecular dynamics simulations are effectively replaced by efficient mathematical models. Effect of two dominating factors on yield strength of the material, namely TBW and twin spacing is analysed and thereby relative sensitivity of these two factors is investigated in this study. Subsequently material responses obtained using the present approach has been explained using deformation mechanism of the nano-twinned copper and they are found to be in good agreement, affirming the accuracy and validity of the proposed methodology.

Previous researches on this subject are based on limited number of samples because the molecular dynamics simulations are normally very expensive and time consuming. In this work, the entire design space has been comprehensively explored by performing thousands of virtual experiments showing the variation of yield strength with different possible combinations of TBW and twin spacing. This will allow researchers to have a complete idea about the optimal combination of these two factors to maximise yield strength of the material. It can be noted here that, as mathematical model for yield strength is possible to be constructed in the PCE based approach, future investigations in this field can follow actual implementation of global optimisation algorithms efficiently to obtain optimum combination of design parameters on a strong mathematical foundation. One of the major findings of this research is that twin width is a more sensitive parameter than twin boundary spacing for yield strength of the material. Such quantitative sensitivity analysis results cater a better understanding on the degree of control necessary for the design parameters.

In this study, a mapping between the two input parameters (TBW and twin spacing) and the output response (yield strength) has been established using surrogate modelling approach enabling us to ascertain the response bound due to random variation of the input parameters. This analysis gives a robust idea about range of variation of yield strength along with an intrinsic knowledge about its probabilistic distribution, which in turn can be very useful in the design process of such material. By application of the proposed approach, it has been feasible to carry out 10 000 virtual simulations for the deterministic analysis and 30 000 virtual simulations for probabilistic analysis (equivalent to total 40 000 molecular dynamics simulations), effectively using only 15 actual molecular dynamics simulations. Thus the integration of PCE with molecular dynamics simulation can significantly reduce the computational cost and time required to comprehensively analyse the response quantity of interest. The number of input factors to explore their effect on the responses in nano-scale can be increased in

future investigations. This will however increase the required number of actual molecular dynamics simulations marginally. The proposed algorithm for efficient probabilistic analysis of yield strength for nano-twinned copper is general in nature and therefore, it can be extended for analysing other materials in nano-scale.

References

[1] Edelstein A S and Cammaratra R C 1998 *Nanomaterials: Synthesis, Properties and Applications* (Boca Raton, FL: CRC Press)
[2] Han J, Globus A, Jaffe R and Deardorff G 1997 *Nanotechnology* **8** 95
[3] Ye Y Y, Biswas R, Morris J R, Bastawros A and Chandra A 2003 *Nanotechnology* **14** 390
[4] Ma X-L and Yang W 2003 *Nanotechnology* **14** 1208
[5] Koh S J A and Lee H P 2006 *Nanotechnology* **17** 3451
[6] Zhang S B 2015 *Comput. Mater. Sci.* **98** 320–7
[7] Ju S-P, Jenn-Sen L and Lee W-J 2004 *Nanotechnology* **15** 1221
[8] Hla S-W 2005 *J. Vac. Sci. Technol.* B **23** 1351–60
[9] Colton R J 2004 *J. Vac. Sci. Technol.* B **22** 1609–35
[10] Valiev R 2002 *Nature* **419** 887–9
[11] Gleiter H 1989 Nanocrystalline materials *Prog. Mater. Sci.* **33** 223–315
[12] Valiev R 2004 *Nat. Mater.* **3** 511–6
[13] Lu K, Lei L , and Suresh S 2009 *Science* **324** 349–52
[14] Li X, Wei Y, Lu L, Lu K and Gao H 2010 *Nature* **464** 877–80
[15] Wu Z X, Zhang Y W and Srolovitz D J 2009 *Acta Mater.* **57** 4508–18
[16] Luque A, Maryam G and William A C 2014 *Acta Mater.* **81** 442–56
[17] Zheng Y G, Zhao Y T, Ye H F and Zhang H W 2014 *Nanotechnology* **25** 315701
[18] Zhang J J, Hartmaier A, Wei Y J, Yan Y D and Sun T 2013 *Modelling Simul. Mater. Sci. Eng.* **21** 065001
[19] Zhang J, Wei Y, Sun T, Hartmaier A, Yan Y and Li X 2012 *Phys. Rev.* B **85** 054109
[20] Lu L, Chen X, Huang X and Lu K 2009 *Science* **323** 607–10
[21] Sinha T and Kulkarni Y 2011 *J. Appl. Phys.* **109** 114315
[22] Bezares J, Jiao S, Liu Y, Bufford D, Lu L, Zhang X, Kulkarni Y and Asaro R J 2012 *Acta Mater.* **60** 4623–35
[23] Cao A J and Wei Y G 2007 *J. Appl. Phys.* **102** 083511
[24] Li L and Nasr M G 2009 *Phys. Rev.* B **79** 075444
[25] Hammami F and Kulkarni Y 2014 Size effects in twinned nanopillars *J. Appl. Phys.* **116** 033512
[26] Jang D, Li X, Gao H and Greer J R 2012 Deformation mechanisms in nanotwinned metal nanopillars *Nat. Nanotechnol.* **7** 594–601
[27] Zhang Y and Huang H 2009 Do twin boundaries always strengthen metal nanowires *Nanoscale Res. Lett.* **4** 34–8
[28] Deng C and Sansoz F 2010 *Phys. Rev.* B **81** 155430
[29] Deng C and Sansoz F 2009 *Appl. Phys. Lett.* **95** 091914
[30] Wang W, Zhong Y, Lu K, Lu L, McDowell D L and Zhu T 2012 *Acta Mater.* **60** 3302–9
[31] Rizzi F, Najm H N, Debusschere B J, Sargsyan K, Salloum M, Adalsteinsson H and Knio O M 2012 *Multiscale Model. Simul.* **10** 1428
[32] Rizzi F, Jones R E, Debusschere B J and Knio O M 2013 *J. Chem. Phys.* **138** 194105
[33] Race C P 2014 *Molecular Simulation* **41** 1069–73
[34] French W R, Pervaje A K, Santos A P, Iacovella C R and Cummings P T 2013 *J. Chem. Theory Comput.* **9** 5558–66
[35] Papadrakakis M and Stefanou G 2013 Multiscale modeling and uncertainty quantification of materials and structures *Proc. IUTAM Symp.* (*Santorini, Greece*)
[36] Tengen T B, Wejrzanowski T, Iwankiewicz R and Kurzydlowski K J 2010 *Mater. Sci. Eng.* A **527** 3764–8
[37] Konstantinidis A A, Katerina E A and Jeff T M D H 2014 *Mater. Sci. Eng.* A **597** 89–94
[38] Wiener N 1938 The homogeneous chaos *Am. J. Math.* **60** 897–936
[39] Witteveen J A S and Bijl H 2006 Modeling arbitrary uncertainties using Gram–Schmidt polynomial chaos *44th AIAA Aerospace Sciences Meeting and Exhibit, AIAA paper, AIAA 2006-896* (*Reno, Nevada, 9–12 January 2006*)
[40] Dey S, Mukhopadhyay T, Khodaparast H H and Adhikari S 2015 *Appl. Math. Modelling* (doi:10.1016/j.apm.2015.11.038)
[41] Mukhopadhyay T, Dey T K, Dey S and Chakrabarti A 2015 *Struct. Eng. Int.* **25** 173–83
[42] Mukhopadhyay T, Dey T K, Chowdhury R and Chakrabarti A 2015 *Arab. J. Sci. Eng.* **40** 1027–44
[43] Dey S, Mukhopadhyay T, Khodaparast H H and Adhikari S 2015 *Acta Mech.* **226** 2537–53
[44] Mishin Y, Mehl M J, Papaconstantopoulos D A, Voter A F and Kress J D 2001 *Phys. Rev.* B **63** 224106
[45] Verlet L 1967 *Phys. Rev.* **159** 98
[46] Plimpton S 1995 *J. Comput. Phys.* **117** 1–19
[47] Stukowski A 2010 *Modelling Simul. Mater. Sci. Eng.* **18** 015012
[48] Li J 2003 *Modelling Simul. Mater. Sci. Eng.* **11** 173
[49] Deng C and Sansoz F 2009 *Acta Mater.* **57** 6090–101
[50] Cao A J, Wei Y G and Scott X M 2007 *Appl. Phys. Lett.* **90** 151909
[51] Frøseth A, Van Swygenhoven H and Derlet P M 2004 *Acta Mater.* **52** 2259–68
[52] Jin Z-H, Gumbsch P, Ma E, Albe K, Lu K, Hahn H and Gleiter H 2006 *Scr. Mater.* **54** 1163–8
[53] Stukowski A, Albe K and Farkas D 2010 *Phys. Rev.* B **82** 224103
[54] Yamakov V, Wolf D, Phillpot S R and Gleiter H 2003 *Acta Mater.* **51** 4135–47
[55] Jin Z-H, Gumbsch P, Albe K, Ma E, Lu K, Gleiter H and Hahn H 2008 *Acta Mater.* **56** 1126–35
[56] Uchic M D, Dimiduk D M, Florando J N and Nix W D 2004 *Science* **305** 986–9
[57] Zuo L, Ngan A H W and Zheng G P 2005 *Phys. Rev. Lett.* **94** 095501
[58] Deshpande V S, Needleman A and Van der Giessen E 2005 *J. Mech. Phys. Solids* **53** 2661–91
[59] Yuan F and Wu X 2013 *J. Appl. Phys.* **113** 203516