Development of High Throughput Assays for Establishing Process-Structure-Property Linkages in Multiphase Polycrystalline Metals: Application to Dual-Phase Steels

Ali Khosravani^a, Ahmet Cecen^b, and Surya R. Kalidindi^{a*}

^aWoodruff School of Mechanical Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA.

^bSchool of Computational Science & Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA

*Corresponding author

Abstract

Process-structure-property (PSP) linkages are central to the development and deployment of advanced materials in emerging technologies. Conventional approaches for establishing these are generally highly customized, qualitative, and demand major investments of time and effort. In this paper, we formulate novel, data-driven, high throughput, assays for exploring PSP linkages in structural metals. These assays are built on recent advances in low-dimensional quantification of material structure using spatial correlations and principal component analyses, as well as in the mechanical characterization using spherical nanoindentation. These novel protocols are demonstrated on a dual phase (DP) steel that exhibits rich multiphase polycrystalline microstructures.

Keywords: Spherical indentation; Process-Structure-Property Linkages; Dual phase steel; 2-point spatial correlations.

1 Introduction

A great deal of attention in materials innovation is now focused on the accelerated deployment of advanced materials in commercial products [1-6]. An important strategy for realizing this ambitious goal is the development, validation, and adoption of high throughput assays for rapid exploration of the extremely large materials and process design spaces involved. Indeed, significant effort in recent years [7-21] has been aimed at extracting high value (most useful for a selected application) process-structure-property (PSP) linkages [22-34] using a variety of approaches (including multiscale experiments, multiscale models, and data analytics).

Our focus in this paper will be on the protocols needed for the accelerated, experimental, exploration of PSP linkages in multiphase structural metals and alloys (e.g., dual-phase or DP steels). The processing histories employed on these materials typically involve a sequence of thermo-mechanical treatments, whereas the main properties (or performance characteristics) of interest are largely related to the plastic response of the material. Since one typically employs standard tensile tests [35] for evaluating the plastic properties of the material, it becomes necessary to make relatively large quantities of material samples with statistically homogeneous microstructures throughout the gage section (of the sample used in the tension test) for each combination of material chemistry and process path studied. This requirement drives up the cost and effort substantially, as is evident from some of the recent innovative efforts in this direction [12]. In other words, if it were possible to measure reliably the plastic response of the material in very small volumes, it would become possible to reduce dramatically the cost and effort involved in rapid screening of large design spaces in material chemistry and process histories.

Indentation has been employed extensively in prior literature [36-52] for evaluating the mechanical response of materials in small volumes. However, the use of this technique has been

restricted largely to estimation of modulus and hardness. Only recently, it has been shown that it is possible to extract meaningful indentation stress-strain (ISS) curves directly from spherical indentation through a rigorous set of data analyses protocols. These new protocols have thus far been demonstrated mainly at the very small length scales (indentation zones much smaller than a single grain) [53-60]. There is, however, substantial promise for their application at the meso-length scales (where the indentation zone covers several grains [61]).

Beyond the measurement of mechanical response in small volumes, the next major impediment in extracting PSP linkages in advanced structural metals comes from the lack of a rigorous, yet practical, framework for the statistical quantification of their rich microstructures. Most of the currently employed approaches [38, 45, 47, 48, 51, 62-67] are based on highly simplified measures such as elemental compositions, phase volume fractions, average grain sizes of constituent phases, and the orientation distribution function (ODF). Clearly, the number of distinct microstructures one can reconstruct while meeting specific targets on these measures is extremely large, suggesting that this set of measures is likely to be inadequate for capturing all of the salient features of the microstructure. More formally, one might recognize most of the measures listed above (except the average grain size) as 1-point statistics of the microstructure in that they do not capture the morphological attributes of the microstructure. Indeed, one can employ the more advanced 2-point statistics (formally called the 2-point spatial correlations) [30, 31, 68-75] to arrive at a more rigorous quantification of the material microstructure. However, most of the prior examples of the application of the framework of 2-point statistics have been limited to multiphase materials, with only a few focused on the application to polycrystalline materials [76-78]. The main difficulty of applying the 2-point statistics measures to polycrystalline samples arises from the very large number of local states present (for example,

each distinct grain orientation encountered in the sample needs to be treated as a distinct local state), and the fact that the corresponding number of the distinct 2-point statistics is exceedingly large. In addition to being multiphase and polycrystalline, the microstructures of advanced alloys such as DP steels exhibit grain-scale heterogeneity in the spatial distribution of the dislocation densities (see Figure 1). Not surprisingly, a rigorous quantification of these complex microstructures is not readily accomplished with the currently available toolsets. Clearly, one needs to make suitable simplifications in order to establish practically useful PSP linkages for advanced structural metal alloys.

The main goal of this work is to explore the viability of high throughput experimental assays for establishing PSP linkages in multiphase polycrystalline metals, while utilizing small sample volumes and leveraging some of the recent advances described earlier (i.e., spherical microindentation stress-strain protocols and the framework of 2-point statistics). We have selected DP steels for this study because of their importance to several advanced technologies, owing to their excellent combination of continuous yielding behavior, high tensile strength, high work-hardening rate, and good ductility [38, 41, 45, 47, 51, 62, 63, 66, 79-87]. These properties are generally achieved in DP steels through a special heat treatment process called intercritical annealing [36, 52, 64, 65, 67, 80, 84, 88] during which the material is heated up to the austenite/ferrite region, held for a certain amount of time, and quenched to room temperature. Intercritical annealing results in formation of hard particles of martensite (α') in a soft matrix of ferrite (α) grains. This step is usually followed by additional cold work and heat treatment called bake hardening (BH [50, 66, 79, 82, 85-96]; cf. Figure 1) to achieve the desired combination of mechanical properties. The main mechanisms involved in this overall thermomechanical process are as follows: (i) Introduction of mobile dislocations in ferrite grains at the vicinity of α/α'

interfaces ([47, 62, 67, 79, 82, 83, 90, 97-99]; see Figure 1b), generally attributed to the volumetric plastic strains induced during the austenite to martensite transformation that occurs during the quenching from high temperature. This hypothesis is supported by transmission electron microscopy (TEM) observations [66, 85, 97, 98, 100] of higher densities of mobile dislocations in the ferrite matrix close to α/α' interfaces and the continuous yielding behavior exhibited by the DP steels. (ii) Formation of tangled networks of dislocations during the imposed plastic deformation, resulting in higher work hardening rates [79, 81, 85, 91, 97, 98] (see Figure 1c). (iii) Pining of dislocations, relieving residual stresses, and tempering martensite during the bake hardening process through diffusion of carbon atoms [66, 82, 83, 85, 86, 90-93, 95-97, 101] (see Figure 1d). As one might expect, the final properties exhibited by DP steel are critically dependent on the extent of the different mechanisms described above, which in turn depend critically on the process parameters employed in the production of the DP steel.

It is therefore clear that the process space (this is the set of all combinations of process parameters that could be utilized) to be explored in optimizing the set of final properties of interest in DP steels is extremely large. Consequently, high throughput assays mentioned earlier, if successfully employed, could produce dramatic acceleration in the deployment of these alloys in emerging technologies. In this work, we have formulated a foundational framework for the development of high throughput assays for establishing PSP linkages in advanced structural metals and alloys, and demonstrated its successful application to a selected process space in DP steels.

2 Sample Prototyping

The first step in the assays described in this paper is the high throughput prototyping of a sample library. Since our intent is to use indentation techniques for measuring plastic responses,

we need to produce only small quantities of the material (no significant stipulations on sample shape either) for each selected material chemistry and/or process history. In this study, we decided to select a single chemical composition, and allow for variations in the thermo-mechanical processing history.

A four millimeter strip of low carbon steel with a chemical composition (in wt.%) of 0.15C, 1.4Mn, 0.04P, 0.04S was secured and used in this study. The strip was cut into coupons of 10 mm $\times$ 20 mm $\times$ 4 mm. All of the coupons were heat treated at 450 °C for 2 hours in order to produce an annealed microstructure, which then served as the starting material for the study described here. Table 1 (cf. Figure 1) summarizes the different processing histories included in this study, which were selected following the studies published in prior literature [38, 45, 47, 50, 51, 63-65, 67, 82, 83, 85, 86, 90, 102-106]. In the intercritical annealing process, the samples are heated to the specified temperature in a salt bath furnace (taking them into the austenite/ferrite region), held for 4 minutes, and quenched in an oil bath. Subsequent to the intercritical annealing, plane strain deformation was applied by rolling, and was followed by bake hardening at 170 °C for 20 minutes and quenching in water to the room temperature. As described in Table 1, three different intercritical annealing temperatures (750, 780, and 810 °C) and three amounts of total thickness reduction (0, 5, and 10%) were selected for this study. This resulted in nine different processing conditions, with the sample labelling (see Table 1) capturing the processing history information. As an example, sample 750-05-170 indicates an intercritical annealing temperature of 750 °C and 5% thickness reduction, followed by bake hardening process at 170 °C for 20 minutes.

It is emphasized here that the time and effort expended in sample prototyping for the present study are minimal because the assays we intend to use do not pose stringent requirements on

either the size or the shape of material volumes to be produced in each different processing history. In the present study, all of the sample prototyping was completed by one person in about 100 hours.

3 Spherical Microindentation Stress-Strain Protocols

As mentioned earlier, most of the prior work on indentation stress-strain curves has been focused on evaluating the plastic response at length scales that were significantly smaller than the grain size [53, 56, 58-60, 107]. Only recently, these protocols have been extended to microindentation, where the length scales correspond to several grains (about 10-1000) in the indentation zone [108]. For microindentations conducted in the present study, we utilized a customized Zwick-Roell Z2.5 hardness tester with a 6.35 mm radius tungsten-carbide spherical tip. A constant crosshead speed of 0.1 mm/min was utilized in all the tests reported here.

A significant challenge in extracting meaningful indentation stress-strain curves from the indentation experiments comes from the need to estimate reliably the evolving contact radius throughout the entire loading history [57, 109]. In modern nanoindenters (where the indentation zone sizes are typically much smaller than the grain size), this is accomplished through the CSM (continuous stiffness measurement) capability [109]. However, this capability does not yet exist for microindentation experiments. Consequently, the only way to estimate the contact radius reliably is to employ a multitude of unloading segments, and analyze them using Hertz theory [57]. Therefore, custom protocols were designed and implemented to allow a multitude of loading-unloading (corresponding to about 30-50% of the peak force) cycles (see Figure 2(a)). Since each unloading segment produces one data point on the indentation stress-strain curve (Figure 2(b)), the number and frequency of the unloading segments has to be suitably adjusted to produce a meaningful and practically useful indentation stress-strain curve.

The protocols for extracting indentation stress-strain curves from the raw load-displacement data from spherical indentation are based largely on Hertz theory (Eqs. 1-4) expressed as

$$P = \frac{4}{3} E_{eff} \sqrt{R_{eff} h_e^3} \quad (1)$$

$$\frac{1}{E_{eff}} = \frac{1-\nu_i^2}{E_i} + \frac{1-\nu_s^2}{E_s} \quad (2)$$

$$\frac{1}{R_{eff}} = \frac{1}{R_i} + \frac{1}{R_s} \quad (3)$$

where P and h_e are the indentation load and the elastic indenter displacement, and R_{eff} and E_{eff} are the effective radius and effective modulus, respectively. Constants (E_i, ν_i) and (E_s, ν_s) are Young's modulus and Poisson ratio for the indenter and the sample, respectively. For the initial elastic loading of a flat sample surface, the effective radius, R_{eff} , is equal to the indenter radius, R_i . Therefore, it would be possible to extract E_{eff} from the measured load-displacement using standard regression techniques; E_s can be recovered using Eq. (2). The main hurdle in this protocol comes from having to make “zero-point” corrections to both measured load and measured displacement signals. However, with larger the indenter tips, the measurements are less sensitive to the surface quality of both sample and indenter. Consequently, for the microindentation testing here, there was no need for a zero-point load correction.

After the material in the indentation zone experiences plastic deformation, Eq. (1) needs to be modified to introduce the permanent displacement or residual height, h_r :

$$h = \left[\frac{3}{4} \frac{1}{E_{eff}} \frac{1}{\sqrt{R_{eff}}} \right]^{\frac{2}{3}} P^{\frac{2}{3}} + h_r \quad (4)$$

Note that Eq. (4) can only be applied to unloading segments (these are assumed to be elastic). Applying standard regression techniques on the unloading segments to fit Eq. (4) produces reliable estimates of h_r and R_{eff} . In this analysis protocols, it is assumed that E_{eff} obtained

from the initial elastic loading segment remains unchanged. This is a reasonable approximation for most materials, given the very small strains introduced in the indentation studies presented in this work. An unloading segment between 95% and 50% of the peak load (in each unloading) was employed in this work to estimate the evolving values of h_r and R_{eff} . After the initiation of plastic deformation in the sample, R_{eff} is no longer equal to R_i .

Indentation stress, σ_{ind} , and indentation strain, ϵ_{ind} , corresponding to the start of each unloading segment are computed as

$$\sigma_{ind} = \frac{P_{max}}{\pi a^2} \quad (5)$$

$$\epsilon_{ind} = \frac{4}{3\pi} \frac{h_{s,max}}{a} \quad (6)$$

$$a = \sqrt{R_{eff}(h_{s,max} - h_r)} \quad (7)$$

where P_{max} and $h_{s,max}$ are load and displacement at the peak of each unload, and a is the contact radius. $h_{s,max}$ is used to compensate the elastic displacement of the indenter, h_i , and is computed as

$$h_s = h - h_i \quad (8)$$

$$h_i = \frac{3(1-\nu_i^2)P}{4E_i a} \quad (9)$$

Young's modulus, E_i , and a Poisson ratio, ν_i , of 640 GPa and 0.21 [110] were used for the 6.35 mm radius tungsten-carbide spherical indenter. Eq. (9) essentially estimates the indenter displacement by using Hertz's theory for indentation on a rigid flat surface.

An example indentation stress-strain curve produced in this work is plotted in Figure 2(b), which corresponds to sample 750-10-170. In this test, there were 20 unloading segments. The sample elastic modulus, E_s , is estimated from the initial elastic loading segment (shown

highlighted in red in Figure 2(a)). The sample Young's modulus estimated from this indentation measurement is 207.8 GPa. The indentation yield strength defined based on 0.2% offset on the indentation stress-strain curve using an indentation modulus of $\frac{E_s}{1-\nu_s^2}$, was estimated to be 1191.2 MPa for this sample. Indentation results from the multiple tests (at least 10 tests on each sample) conducted in this work are summarized in Table 2. The average Young's modulus for all indentation measurements on all samples was 201.4 GPa with a standard deviation of 14.2 GPa, which compares well with reported Young's modulus in literature for steel from conventional tensile testing [111]. It is important to recognize that the indentation zone in the experiments reported here is sufficiently large to include multiple microstructural features, including ferrite and martensite regions. This is illustrated in Figure 3 for sample 750-00-000, where the estimated indentation zone sizes are delineated both at yield and at the end of the indentation test. The solid line circle in Figure 3(a) shows the estimated indentation zone size at the end of test, and the dashed line circle corresponds to the zone size at the indentation yield point. The image on right, Figure 3(b), is an SEM image taken on the same sample, whose size was made to correspond to the contact area at indentation yield. As one can see, the indentation area is large enough to justify the measurements reported here as measurements of the bulk mechanical response of the sample, especially keeping in mind that multiple indentations are conducted on each sample and an average is reported for all indentations conducted on the sample (see Table 2). An estimate of the number of grains in the indentation zone can be obtained assuming a cylindrical primary indentation zone of radius a and height $2.4a$ [109]. The contact radii a at the yield point are listed in Table 2 for all tests. Considering the average grain size (30 μm) for the material tested and an average contact radius at yield in the range of 142-182 μm (see Table 2), the number of grains in the primary zone is estimated to be in the range of 190-400 grains.

It is clear from Table 2 that the strength of the DP steel is largely controlled by the amount of both martensite and the imposed cold work. Increasing the amount of martensite (by means of quenching from higher intercritical annealing temperatures) results in strengthening of the DP steel. For example, the indentation yield strength, σ_Y^{ind} , increases from 899.5 ± 62.9 MPa to 1168.6 ± 186.8 MPa, when the quenching temperature is increased from 750 °C to 810 °C. The same trends can be seen even after 5% and 10% cold work and bake hardening were applied. These trends are consistent with the observed results in literature [45, 51, 66, 82, 87, 92, 93, 95, 104]. Although, it is possible to establish suitable conversion factors to estimate uniaxial yield values from the indentation yield values [108], this was not pursued in the present work as the focus in this work is on the establishment of high throughput assays.

It is emphasized here that the sample preparation for the microindentation tests reported here is identical to the sample preparation used for microstructure characterization (described next). In other words, the same sample preparation is used for both aspects of the study, leading to substantial economy in both time and effort expended.

4 Microstructure Characterization

The next step in the assays employed in this work focused on microstructure characterization. In prior literature, the most common approaches employed in microstructure characterization of DP steels included optical microscopy [36, 38, 45, 51, 64-66, 83, 88, 104-106], scanning electron microscopy [36, 41, 42, 47, 51, 63, 88, 104, 106], and electron back-scattered diffraction (EBSD) [47, 48, 112-116]. While each of these techniques has demonstrated certain advantages over the others for highlighting specific aspects of the microstructure, our goal here is to identify and employ the protocols that give us the best chance at establishing reliable PSP linkages.

Furthermore, we also desire to minimize the overall effort required so that we can maximize the potential for high throughput.

After a careful consideration of the relative merits of the different protocols, it was decided to employ the EBSD technique for the present study. In addition to mapping lattice orientations in ferrite regions and indirectly identifying martensite regions (based on very poor diffraction patterns), the EBSD technique provides critically needed information on the extent of deformation pinning (in the ferrite grains next to the martensite grains; cf. Figure 1) through an analysis of the lattice misorientations. Consequently, although the use of the EBSD technique represented a time and effort intensive option for the present study, it was deemed essential as the other microscopy techniques are incapable of capturing any relevant information on the extent of pinned dislocations in the samples. It is emphasized that some measure (even if it is an indirect measure) of the pinned dislocation density is essential to correlate to the plastic properties of the sample.

EBSD scans were carried out in a field emission gun scanning electron microscope at 20 kV. Samples were prepared (for both EBSD and indentation studies) using standard metallography protocols. This involved grinding with silicon carbide papers down to grade 4000, and polishing sequentially with diamond suspensions of 3 μm , 1 μm , and OP-A alumina suspension. The final step in sample preparation was electro-polishing using A3 Struers electrolyte at 5 °C.

Several scans (around 9 or 10) were collected on ND plane on all samples with 1 μm resolution. It was also decided to keep the scan size consistent with the primary indentation zone in the mechanical characterization described earlier. The EBSD scan size was selected to be 400×400 μm^2 . This is important because one of the goals of this study is to find linkages between indentation yield strength, σ_Y^{ind} , and the microstructure. Just as we conducted multiple

indentation measurements on each sample, we have also conducted multiple EBSD scans on each sample to allow for the quantification of variance.

The collected raw data was post-analyzed using TSL OIM[®] software. Example EBSD scans obtained for different samples in our sample library are shown in Figure 4. In these figures, multicolored regions represent ferrite grains whose lattice orientations have been mapped out with a 1 μm spatial resolution and martensite regions are colored black. One can see that the martensite mostly forms at grain boundary regions and triple points. The amounts of martensite appear to increase with quenching from a higher intercritical temperature (from top row to the bottom row in Figure 4). Furthermore, a close inspection of the EBSD scans in Figure 4 reveals that samples subjected to higher deformation levels show significantly higher levels (and extent) of in-grain misorientations (i.e., compare left side maps with right side ones in Figure 4). The main implication here is that the misorientation fields in the samples do capture some information on the extent of pinned dislocations and their spatial distributions in the microstructure. This information will be utilized later in establishing the PSP linkages of interest in this study.

Figure 5 shows $\varphi_2 = 45^\circ$ section of the orientation distribution functions (ODFs) for all samples produced in this study. All textures depict α fiber, β fiber, and cube components. Applying deformation by means of cold work moves the strongest texture component from the $\{001\}\{110\}$ α fiber component to the $\{111\}\{110\}$ β fiber component. It is seen that the differences in texture between the samples are not very significant.

As it was mentioned earlier, EBSD technique was chosen for this study because it provides information about phase as well as the extent of pinned dislocations (as indicated by local orientation gradients in the ferrite). In the case of DP steel, martensite can be identified based on

low values of CI (confidence index) and IQ (image quality) [47, 48, 112, 113, 115, 116]. The poor quality of diffraction patterns in martensite is generally attributed to the high density of defects in these regions. More importantly, as the DP steel is deformed, dislocations accumulate at the martensite-ferrite boundaries producing significant local gradients in the lattice orientations (see the right column in Figure 4). In other words, a suitable measure of the local orientation gradients can serve as an indicator of the local (microscale) hardening in the sample.

Currently used measures of in-grain misorientations can be broadly grouped as either grain-based or kernel-based approaches. Grain based approaches such as GOS (grain orientation spread) [117-119], GAM (grain average misorientation) [117, 119] estimate an average value of misorientation based on all measurements within one grain. In contrast, in kernel based approaches such as KAM (kernel average misorientation) [99, 120-122], quantities such as local average misorientation and local misorientation spread are computed at each grid point in the measurement scan. Clearly, the KAM is a better indicator of the local hardening in the microstructure.

KAM is defined as the average misorientation between the orientation at the center of the kernel and all its neighbors (Figure 6(a)). In this approach, misorientations that exceed some tolerance value (maximum misorientation, 5°) are excluded from the averaging calculation, to avoid erroneous consideration of neighbors across a grain boundary. The distributions of KAM values recovered from the EBSD scans for the different samples in our sample library are shown in Figure 6(b).

For the quantification of each microstructure, we need to make a judicious selection of the local states to be included in the analyses. Local states denote attributes needed to assign the local properties associated with each voxel of the microstructure. For the samples studied, the

local state would be ideally defined by a combination of the phase identifier (i.e., ferrite or martensite), the lattice orientation, the KAM value, among several other potential attributes. It should, however, be recognized that adding more local state attributes will exponentially increase the effort expended in arriving at PSP linkages of interest. Therefore, it behooves us to make smart choices in the selection of local states.

In the present study, we have decided to include only the phase identifier and the KAM as the local state attributes. Since the texture in the sample did not change appreciably between the different processing histories employed in the study, the lattice orientation was not included as a local state variable in each voxel. This is also partly because the inclusion of the lattice orientation as a state variable increases dramatically the effort involved in microstructure quantification [76, 78], which is not justified when there do not exist significant differences in crystallographic and/or morphological textures in the ensemble of samples studied. Finally, it was also decided to use a discretized representation of the KAM value in the microstructure quantification pursued in this work. Based on the histograms shown in Figure 6(b), it was decided that a KAM value of 0.8° or more would be labelled as “deformed ferrite”, while KAM values lower than 0.8° would be labelled as “undeformed ferrite”. Note that 95% of the KAM values in all samples quenched at 750, 780, and 810 °C (without any cold work) exhibited values below 0.8° (see Figure 6(b)), justifying the selection of this value.

As a consequence of the simplifications introduced above, each voxel in the microstructure is assigned to one of three discrete local states: martensite, deformed ferrite, or undeformed ferrite. Although one might raise objections to such gross simplifications, it is important to recognize and understand that a much more detailed quantification of the microstructure may actually be not warranted in the present study. Indeed, we will demonstrate that this simple selection of local

states is adequate in the present study to establish reliable PSP linkages of interest. Of course, as one expands the processing windows to be included in the PSP linkages being formulated, one would need to add additional attributes in the description of the local states.

Figure 6(c)-(d) show selected areas of the measured IPF maps for 750-00-000 and 750-10-170 samples. Figure 6(e) and 6(f) show the corresponding discretized microstructures employed in the quantification of the material structure in this study. As noted earlier, these microstructures have only three discrete states, which are colored black, white, and blue in Figures 6(e) and 6(f). Note the increase in the amount of blue voxels, especially near the martensite interfaces, in the 750-10-170 microstructure compared to the 750-00-000 microstructure.

In this study, we will employ the framework of 2-point statistics to quantify the microstructures generated in this work. These are denoted as $f_r^{hh'}$ and are computed as [123]

$$f_r^{hh'} = \frac{1}{S_r} \sum_{s=1}^S m_s^h m_{s+r}^{h'} \quad (10)$$

where S is the total number of bins in the microstructure, m_s^h is discretized representation of the microstructure denoting the volume fraction of local state h in voxel s , and r indexes discretized vectors placed into the microstructure for evaluating the 2-point statistics. In essence, the 2-point spatial correlations capture the probability of finding discrete local states h and h' at the tail and head of a discrete vector indexed by r . Since, the microstructures studied here have three local states, a total of nine 2-point spatial correlations can be defined. Three of the computed 2-point correlations for sample 780-10-170 are shown in Figure 7, along with the original EBSD scan and the discretized microstructure. The autocorrelation of the martensite (colored black), the autocorrelation of the deformed ferrite (colored blue), and the cross-correlation of the martensite and the deformed ferrite are shown in Figure 7(c), 7(d), and 7(e), respectively. These spatial statistics maps contain a tremendous amount of information about the morphology and

distribution of the local states in the microstructure. For example, the center peaks in the autocorrelations provide the volume fraction information. Consequently, it can be seen that the volume fractions of martensite and deformed ferrite are 0.45 and 0.276, respectively, for the microstructure shown in Figure 7. The contours in the center of the autocorrelation plots provide quantitative information on the shape of local state field in the microstructure. One can also extract a large number of conventional microstructure measures from these plots [30, 31], and even reconstruct the original microstructure from its spatial correlations [124-126].

The quantification of microstructures using the framework of 2-point statistics produces a very large vector of descriptors for the microstructure (every voxel in each of the correlation maps would be an entry in this vector). Principal component analyses (PCA) has been shown to be remarkably effective in arriving at objective (data-driven) low dimensional representations [34, 68, 74, 127-131] of the microstructure. Mathematically, this can be expressed as

$$f_r^{(k)} = \sum_{i=1}^{P-1} \alpha_i^{(k)} \varphi_{ir} + \tilde{f}_r \quad (11)$$

where $f_r^{(k)}$ denotes a contracted representation (i.e., as a single vector) of all the spatial correlations of a microstructure indexed by k , $\tilde{f}_r$ denotes the ensemble average of all microstructures included in the analyses, φ_{ir} denote the directions of the new linearly transformed reference frame (identified by PCA) and α_i are the coordinates (also called PC scores) in the new reference frame. A salient feature of PCA is that it transforms the data into a new orthogonal frame where the axes are ordered by the maximum variance seen in the data.

For the present case study, PCA was performed on a total of 81 EBSD scans (nine scans for each of nine process histories). The analyses indicated that only two PC scores (see Figure 8) are adequate to capture 99.4% of the variance between the different microstructures in this ensemble (i.e., all 81 scans included in the study). In other words, only two numbers (i.e., the first two PC

scores denoted as PC1 and PC2) are adequate to distinguish each of the microstructures produced in this study. This is indeed a remarkable dimensionality reduction, and to the best of our knowledge, unmatched by any other existing framework for microstructure quantification.

For microstructures with three local states, only two of the nine 2-point spatial correlations would be independent [72], if the boundaries of the microstructure were treated as periodic. However, through repeated trials of the PCA described above, we observed that using the two autocorrelations and the cross-correlation shown in Figure 7 provided a good separation between the microstructures of each of the nine groups of samples.

Figure 9 shows all of the microstructures included in the present study in the low dimensional PC space, where each point represents one microstructure. The interclass (nine different processing histories) and the intraclass (nine microstructures from each sample) variances are clearly discernable in this plot. Furthermore, one should note that the variance in PC1 is higher than the variance in PC2 for the ensemble of microstructures studied. It is observed that the intraclass variance for the deformed samples is higher than the intraclass variance for the undeformed samples. Most importantly, one can discern trends in the microstructure evolution by following the centroids of each class of microstructures. For example, one can capture quantitatively the effect of quenching temperature by following the dashed lines. Similarly, one can follow the effect of cold work by following the solid lines in this figure. These trend lines can be called processing paths or processing streamlines, as they capture quantitatively the salient aspects of the influence of process parameters on the evolved microstructures. It is indeed remarkable that a simple data-driven approach involving 2-point spatial correlations and PCA can capture so much detail with just two PCs.

5 Extraction of PSP Linkages

Building on the low dimensional structure representation obtained in the previous section (i.e., PC1 and PC2), we are now in a position to establish PSP linkages of interest. For the present case study, the process parameters are taken as the intercritical annealing temperature (T) from which the samples are quenched, and the imposed thickness reduction during the rolling process (D). The desired process-structure linkages are then established using least squares regression and cross-validation techniques [68], which resulted in a second-order polynomial fit expressed as

$$PC1 = -5612.478 + 14.089 T - 5.138 D + (15.308 (D \times T) - 8.862 T^2 - 332.461) \times 10^{-3} \quad (12)$$

$$PC2 = -2734.355 + 6.665 T - 1.682 D + (1.823 (D \times T) - 4.035 T^2 - 131.892) \times 10^{-3} \quad (13)$$

The accuracy of these models is displayed as parity plots in Figure 10, where each point corresponds to one process history (i.e., the average of all 9 scans from each sample).

PC1 and PC2 denote the strengths of certain spatial patterns in the microstructure. These patterns are visualized in Figure 11. The first column in this figure shows the ensemble average, while the second and third columns depict the patterns captured by PC1 and PC2, respectively. In the PC space, each microstructure is approximated as a weighted deviation from the ensemble average (see Eq. (11)) using the φ_{ir} (plotted in second and third columns of Figure 11) as an orthonormal basis. As an example, as PC1 is increased, it is seen (from the second column in Figure 11) that the volume fractions of martensite and deformed ferrite will both increase and the martensite regions will become more elongated in the horizontal direction. Likewise, when PC2 is increased, there is an increase in the volume fraction of martensite accompanied with a decrease in the volume fraction of the deformed ferrite, while making the martensite regions more equiaxed. Of course, there is a lot more information embedded in the basis maps shown in

Figure 11, which actually contributes to the remarkable efficacy of PCA in arriving at the low dimensional representation employed in this study. There currently do not yet exist any systematic approaches for carefully studying these basis maps.

The approach described above can also be applied to establish a structure-property linkage. For the present case study, the recovered structure-property linkage is expressed as

$$\sigma_Y^{ind} = 1156.4 + 9.2 (PC1) + 5.5(PC2) \quad (14)$$

where σ_Y^{ind} denotes the yield strength of the material measured in the indentation protocols described earlier. The accuracy of this linkage is depicted in the parity plot shown in Figure 12.

The reader is reminded that the linkages developed here are data-driven. One of the implications is that the PSP linkages built here will change when new data is added. This is to be expected, especially since we used only nine different processing histories in this study. When a substantial amount of data becomes available, we could expect the PSP linkages to become stable (i.e., there would be no need to change the PSP linkages with the addition of new data points). This indeed is the most attractive feature of the protocols described here. They allow a coordinated effort by research groups and/or investigators in ways that leverage all available data and minimize redundant effort in establishing such linkages. Furthermore, with the use of high throughput strategies, such as the indentation methods described in this paper, it is conceivable to aggregate PSP linkages of high value to the scientific community at dramatically accelerated pace compared to the conventional approaches used currently.

6 Discussion

The case study described above points mainly to the tremendous potential for developing and implementing high throughput assays in the exploration of PSP linkages in structural alloys. It is fully acknowledged that the term “high throughput” is being used here to signify the acceleration

when compared to conventional approaches. We now review the main elements of the envisioned high throughput assays, and identify specific opportunities for future development.

One of the key elements of the high throughput assays described in the paper is in the use of instrumented indentation techniques for local measurements of mechanical response and their expression as normalized stress-strain curves. Indeed, the expression of the local mechanical response in the form of stress-strain curves is critically important for ensuring reliability and reproducibility of the measurements. Furthermore, the ability to conduct a large number of these tests at different length scales (by simply changing the indenter tip radius) even in a single sample, is clearly of major significance to multiscale modeling efforts. At the present time, this is the most practical approach for producing the large amount of experimental observations needed to calibrate the numerous parameters in multiscale modeling efforts [132-134]. However, in order to realize its full potential, additional work is needed in automation of this technique across all length scales of interest. As mentioned earlier, the experimental protocols are already fairly automated for the indentations conducted at the very small length scales. However, the experimental protocols for the indentations conducted at the larger length scales (indentation zone sizes covering multiple grains) are yet to be automated. The indentation stress-strain analyses protocols used in this work have been coded and are being prepared for broad dissemination as an open source software. While this software speeds up the analyses significantly, it still needs further development and automation. One other significant benefit of the indentation protocols is that they allow for parallelization. Indeed, several of the commercial vendors for instrumented indenters are currently exploring concepts that allow for simultaneous indentation measurements on a grid (using multiple indenters simultaneously). Success in one or more of the concepts mentioned above will contribute significantly to the further development of the high throughput assays presented in this work.

A second key element of the high throughput assays described here is in the use of the emerging data science techniques for formulating the reduced-order PSP linkages. The efficacy is derived from the ability to template the workflows in ways that they can be applied to a broad range of material systems over a multitude of length/structure scales [68, 127, 135-138]. However, the use of a common template does not imply lack of flexibility. On the contrary, the modular approach comprising a specific sequence of steps involving a (i) digital representation of the microstructure, (ii) evaluation of microstructure statistics, (iii) low dimensional representation of the microstructure, and (iv) extraction of a reduced-order model allows for the development of highly efficient (high performance) computer codes leading to a systematic evaluation and objective identification of the best protocols. This is because the modern data science approaches allow one to establish a large number of possible workflow pipelines (utilizing the best available codes), push a large amount of data through these pipelines in parallelized efforts, and disseminate the findings in open source and open access repositories. As examples, in the case study presented in this work, one could evaluate a series of other choices for the digital representation of the microstructure (e.g., use other measures of local lattice misorientation or even the full information of the lattice orientation in the local state descriptors employed, use information gathered by other microscopy techniques), microstructure statistics (e.g., use chord-length distributions [139] instead of 2-point statistics), low dimensional representations (e.g., factor analysis [140], projection pursuit [141], independent component analysis [142]), and machine learning approaches (e.g., M5 model trees [143], support vector machines [144]). A further complicating factor in arriving at the most robust and reliable PSP linkages comes from the fact that the models produced in this approach are, to a significant extent, dependent on how much data is available (this is why these have been referred as data-driven throughout this paper). In other words, one should expect the best protocols and the

models to change as more data is added to the analyses (one can hope that the models stabilize after a critical amount of data has been employed – but in the interim one can still use the best available models at any time). Because of the very large number of choices one faces as well the significant cross-disciplinary expertise required for such a systematic evaluation, it is necessary to transition this pursuit from individuals, or even small groups of individuals, to a community-level curation effort. The data science workflows of the type presented in this work can lead this important transformation, and really accelerate the collection and curation of the PSP linkages for a broad range of materials systems of general interest to the entire materials science community [32, 33, 145, 146].

7 Conclusions

In this study, we have formulated and demonstrated novel high throughput assays for extracting data-driven, high value, PSP linkages in complex multiphase structural alloys such as DP steels. These new protocols utilized spherical indentation stress-strain measurements, 2-point spatial correlations, principle component analysis (PCA), and regression techniques. Following are the major findings from this study:

1. Spherical indentation is a reliable high throughput tool for probing the changes in the yield behavior of dual phase steel processed at different intercritical annealing temperatures and different amounts of cold work applied before bake hardening. While indentation elastic modulus showed only small variations as expected, indentation yield strength varied significantly with the processing history. This is attributed to the many changes induced in the microstructure of the sample with the processing history. These have included not only changes in the volume fractions of the different constituents, but also their spatial distributions. Additionally, the spatial

distribution of the dislocation content at the martensite/ferrite interface also played an important role in controlling the final plastic yield values in the sample.

2. The application of PCA on the 2-point correlations provided a rigorous framework for objective, low-dimensional, quantification of the microstructures in DP steels. In this work, this protocol was applied only on the phase and misorientation information. The protocol found that only two PCs are needed to distinguish the microstructures studied and establish reliable PSP linkages. However, the framework is highly extensible for including more details of the microstructure (additional local states as well as higher-order spatial correlations) as more data becomes available.

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Figure 1. Schematic of a dual phase steel processing path and the expected strengthening mechanisms in each step of the thermo-mechanical process.

Figure 2. Example microindentation load-displacement curve, and indentation stress-strain curve for a test on sample 750-10-170. The red data corresponds to the initial elastic segment used to determine the effective modulus.

Figure 3. a) An optical micrograph showing the size of indentation after unloading, dashed line circle shows the estimated size of the contact area at yield. b) A representative BSE image with the same size as the area indicated by the dashed line.

Figure 4. A sampling of microstructures produced in the present study.

Figure 5. Ferrite textures in the $\varphi_2 = 45$ degree section of the ODF for the different samples produced in this study.

Figure 6. (a) Schematic of KAM definition for each pixel in the EBSD scan. (b) KAM distributions for the samples studied. (c) and (d) IPF maps from selected areas in samples with and without cold work. (e) and (f) Corresponding KAM maps identifying regions with KAM values larger than 0.8 in blue.

Figure 7. (a) An IPF map from the sample 780-10-170. (b) A map showing the three local states selected for the study - martensite as black pixels, deformed ferrite as blue pixels, and undeformed ferrite as white pixels. (c) Autocorrelation of martensite, (d) autocorrelation of deformed ferrite, and cross-correlation of martensite and deformed ferrite.

Figure 8. The individual and cumulative variances from the PCA for the first 3 principal components shows that PC1 and PC2 carry 99.4% of the total variance in the dataset.

Figure 9. Low-dimensional representation of the entire ensemble of 81 micrographs obtained from PCA, colored by the different processing histories. Solid lines capture the influence of cold work, while the dashed lines show the effect of the intercritical annealing temperature.

Figure 10. The accuracy of process-structure linkages established in this work using data science approaches. The process variables included the intercritical annealing temperature (T) in $^{\circ}\text{C}$ and the amount of cold work (percentage of thickness reduction) from rolling process. Each point is an ensemble average of 2-point statistics of all scans from one processing history.

Figure 11. The ensemble average and the first two PC basis (ϕ_{1r} and ϕ_{2r} ; see Eq. (11)) computed in the PCA on the entire ensemble of microstructures studied.

Figure 12. The accuracy of the structure–property linkage established in this work using data science approaches. The property captured is the indentation yield strength. Each point is an ensemble average of all measurements from one processing history.

Table 1. Sample library and the labelling scheme employed in this study. The label includes information on intercritical annealing temperature, amount of cold work. and bake hardening temperature.

Sample code	Intercritical annealing temperature (°C)	Cold work -Thickness reduction (%)	Bake hardening at 170 °C for 20 min
750-00-000	750	0	✕
750-05-170	750	5	✓
750-10-170	750	10	✓
780-00-000	780	0	✕
780-05-170	780	5	✓
780-10-170	780	10	✓
810-00-000	810	0	✕
810-05-170	810	5	✓
810-10-170	810	10	✓

Table 2. Comparison of the indentation yield strength, the indentation elastic modulus, and the contact area at yield point between samples subjected to different thermo-mechanical treatments. At least ten measurements were made on each sample.

Sample code	Average indentation yield strength (MPa)	Average indentation elastic modulus (GPa)	Radius of the contact area at yield point (μm)
750-00-000	899.5 ± 62.9	174.5 ± 25.3	142.3 ± 7.0
750-05-170	950.4 ± 29.7	193.4 ± 23.3	166.7 ± 13.2
750-10-170	1100.9 ± 130.3	200.9 ± 22.3	176.2 ± 11.5
780-00-000	1097.7 ± 41.6	216.8 ± 23.3	165.7 ± 14.8
780-05-170	1299.4 ± 68.7	209.6 ± 15.4	178.1 ± 17.7
780-10-170	1336.3 ± 69.5	201.9 ± 16.7	181.3 ± 14.2
810-00-000	1168.6 ± 186.8	188.5 ± 14.7	153.3 ± 10.9
810-05-170	1340.4 ± 130.9	219.2 ± 14.5	169.3 ± 10.2
810-10-170	1506.5 ± 132.3	207.6 ± 9.7	180.5 ± 15.1

Spatial Correlations of Microstructure

Microindentation Stress-Strain Measurements

Low-dimensional Representation

Process-structure-property (PSP) linkages