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A simple isotropic hardening rule accounting for grain size effects in metal plasticity: modeling from coarse-grained to nanocrystalline materials

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Abstract. This work presents a novel phenomenological constitutive model intended to predict quasi-static stress-strain response in metal plasticity, from coarse-grained to nanocrystalline materials. To this end, a modified Voce hardening law is proposed, in which the initial yield stress and the saturation hardening parameters depend on the initial grain size. The constitutive model is adjusted considering experimental data available in literature for pure aluminum, copper and titanium. Comparisons with experimental data show the constitutive model capabilities, which in general provide good predictions for the three materials within wide strain and grain size ranges. The proposed model proved to be a simple and powerful constitutive tool for engineering and material design purposes.

Keywords: Grain size effects, Coarse-grained materials, Nanocrystalline materials, Hardening rule, Hall-Petch effect

INTRODUCTION

In general, metals with smaller grains present improved mechanical properties as higher strength and higher hardness (Hall, 1951; Petch, 1953), and also better surface finishing. The last one have gained more and more significance due to requirements or limitations associated with the manufacturing of micro-parts (Wang et al., 2015). Such desired properties have unleashed a growing demand by ultrafine-grained $(100\,\text{nm} < d < 1\,\mu\text{m})$, d being the grain size, or even nanocrystalline materials $(d < 100\,\text{nm})$ to be used in diverse industrial applications where improved mechanical properties are required, or in the manufacturing of micro-parts which are used, for instance, in medical devices and micro-electromechanical devices. Other properties, as ductility, toughness, and strain-rate sensitivity are strongly affected by the grain size (d), which has been recognized as an important parameter in the selection/design of materials (Kumar, Swygenhoven & Suresh, 2003; Meyers, Mishra & Benson, 2006; Estrin & Vinogradov, 2013).

Several synthesis techniques to obtain ultrafine and nanocrystalline grains in metals are reported in the literature. These techniques include inert gas condensation, electrodeposition, crystallization from amorphous material, severe plastic deformation (SPD), among others (Meyers, Mishra & Benson, 2006). In order to bear the extreme conditions imposed by processing techniques, like in SPD, and to describe the behavior of ultrafine and nanocrystalline materials under the most diverse mechanical conditions, several experimental investigations have been conducted considering *quasi*-static conditions (Witkin, Han & Lavernia, 2005; Meyers, Mishra & Benson, 2006), under different strain-rate and temperature levels (Khan, Farrokh & Takacs, 2008a,b).

Aiming at predicting grain size effects on mechanical behavior of polycrystalline metals either under manufacturing process or service operation conditions, a variety of constitutive models have been proposed. Most of the models have as foundation the pioneer *Hall-Petch* model (Hall, 1951; Petch, 1953) and its generalization presented by Armstrong et al. (1962). These models are based on the concept of dislocation piling-up at grain boundaries, which act as barriers to plastic flow. However, both models seem as phenomenological, since empirical equations to describe the yield strength in terms of grain sizes are used. Further extensions of the *Hall-Petch* model have been proposed, trying to incorporate more information from micro- or even nano-mechanics in constitutive formulations, as done in works of Gryaznov & Trusov (1993) and Pande & Cooper (2009), in which fundamental mechanisms of the plastic deformation process have been considered.

In some constitutive approaches the different mechanisms responsible for the plastic flow of polycrystalline metals at lower scales are considered in a separated manner, such as in the core-mantle models (Meyers & Ashworth, 1982; Kim, Estrin & Bush, 2000; Jiang & Weng, 2004). In these formulations, each grain is viewed as a composite constituted by cores (interior of grains) and mantles (grain boundaries). Each constituent has specific plastic deformation mechanisms: grain interior deforms by dislocation slip and lattice diffusion (creep), and grain boundary by grain boundary sliding induced by mass diffusion. The polycrystalline aggregate response is obtained from a particular homogenization technique (rule of mixtures, self-consistent method, etc.). This approach allows the identification of the governing deformation mechanisms for each grain size range, and therefore allows to predict inverse *Hall-Petch* effect observed in nanocrystalline materials. Furthermore, size effects have been also modeled by non-local theories of plasticity formulated at macroscopic scales (Fleck et al., 1994) or at single crystal scales (Evers, Brekelmans & Geers, 2004; Cordero, Forest & Busso, 2012). These formulations are based on incompatibility measure (Burgers vector measure) in the plastic deformation field, leading to the concept of geometrically necessary dislocations (Cottrell, 1964; Ashby, 1970), which naturally introduces a length scale into the constitutive formulation.

In this context there are several proposals that can be applied to the constitutive modeling of metals considering size effects. In general terms, the majority of the micromechanical models have demonstrated aptitude to predict experimental results. However, this type of constitutive formulation requires more advanced knowledge of fundamental plasticity mechanisms and often involves a higher number of material parameters (when compared with phenomenological models). In many cases these are difficult to identify by simple calibration procedures, requiring costly and very time consuming experiments. In the searching for efficient macroscopic models, the *Khan-Huang-Liang* model (Khan & Huang, 1992; Khan & Liang, 1999; Khan, Suh & Kazmi, 2004) was extended by Farrokh & Khan (2009) (where the *Khan-Liang-Farrokh* model was proposed) to predict the behaviour of metals, such as copper and aluminum, with ultrafine and nanocrystalline grains at different strain-rates and temperatures. The *Khan-Liang-Farrokh* model was recently employed in reference (Liu et al., 2015) to simulate compression testing of pure titanium. In general, the model has been demonstrated to be an efficient constitutive tool for modelling ultrafine-grained and nanocrystalline metals. A simple and robust approach intended to account for size effects in nano-grained and nano-twinned metallic materials was proposed in works (Zhang, Romanov & Aifantis, 2011, 2015). Using gradient plasticity and physically-based arguments, these authors modified a *Voce*-type (Voce, 1948; Kocks, 1976) hardening rule in order to incorporate size-effects in the constitutive formulation. Size effects were incorporated by considering a dependence of *Voce* hardening parameters, such as the yield stress and hardening saturation, on a length scale parameter (grain size or twin width). However, the formulation preserved a *Voce*-type hardening law in its original format, thus predicting stress-strain curves that are not able of modeling *quasi*-linear hardening observed in the latter stages of straining, for FCC metals (Tome et al., 1984; Rollett et al., 1989).

The aim of this work is to propose a simple but efficient phenomenological constitutive model specifically devised for predicting the mechanical behavior of metals with different average grain sizes, varying form coarse ($d = O(10\mu\text{m})$) to nanocrystalline ($d = O(10\text{nm})$) grains. The model is formulated considering stress-strain response under *quasi*-static, adiabatic, and isothermal (constant room temperature) conditions. The present constitutive extends a previous modified *Voce* hardening law (dos Santos et al., 2016, 2018), which is phenomenologically-based on dislocation generation-annihilation and misorientation concepts. The latter proved successful to predict *quasi*-constant hardening rate at large strains. In the extended hardening model, the initial yield stress σ_y and the non-linear hardening saturation A_∞ parameters are given in terms of the average initial grain size d, similarly to what was done in (Zhang, Romanov & Aifantis, 2011, 2015). However, differently from these works, such a dependence is deduced from the micromechanics-based model proposed by Gryaznov & Trusov (1993). Due to its simplicity, the constitutive parameters involved in the modeling can be identified in subsequent steps, and thus a conventional non-linear least-square procedure can be employed. The material model calibration is performed having as reference experimental data presented in literature for pure aluminum, cooper (Farrokh & Khan, 2009) and pure titanium (Liu et al., 2015; Ahn et al., 2015).

CONSTITUTIVE MODEL

Assuming rigid-plasticity, the material yield strength is prescribed in terms of the accumulated plastic strain ε by

$$\sigma = \sigma_y + A_\infty \left[1 + c\varepsilon - \exp\left(-\delta\varepsilon\right)\right], \tag{1}$$

in which σ_y is the initial yield stress, A_∞ is the non-linear hardening saturation, c is a proportionality parameter relating the hardening rate (*quasi*-linear) observed at advanced stages of straining, to the hardening saturation, i.e., $\frac{\partial\sigma}{\partial\varepsilon} \approx A_\infty c$ for large deformations. Parameter δ is the rate at which the stress reaches saturation at initial states of deformation.

Constitutive equation (1) was initially proposed for the phenomenological modeling of FCC metals by dos Santos et al. (2016). However, the development resulting in equation (1) has not considered grain size effects. In this article, the original model shall be modified in order to explicitly account for the effect of the initial grain size on the yield strength under monotonic *quasi*-static deformation processes. Furthermore, Eq. (1) is in itself a modified form of the *Voce* equation (Voce, 1948; Kocks, 1976), which has been widely employed to model inelastic stress-strain response of metals in engineering problems. The modified hardening law in Eq. (1) has actually been conceived as an advanced hardening rule similarly to those presented in works (Tome et al., 1984; Simo & Armero, 1992). In addition to their capacity to describe the phenomenological behavior of a wide range of metals and having functional forms linked to dislocation density evolution equations (Kocks, 1976; Kocks & Mecking, 2003), the *Voce*-type hardening rules allow for the clear graphical identification of their material parameters.

It is widely admitted that the initial yield stress σ_y depends on the grain size d. To account for such a dependence, one may quote the well known *Hall-Petch* (Hall, 1951; Petch, 1953) equation, which can not however by employed over a wide range of grain sizes (Kumar, Swygenhoven & Suresh, 2003; Meyers, Mishra & Benson, 2006). The starting point of the proposed approach is the model originally proposed by Gryaznov & Trusov (1993), which reads

$$\sigma_y = \sigma_{y_0} + k_0 \ln d + k_1 d^{-\frac{1}{2}} + k_2 d + \sum_{n=3}^{\infty} k_n d^{-\frac{n}{2}}, \tag{2}$$

where the parameters k_n, $n = \{0, 1, 2...\infty\}$, depends on the structure and density of dislocations as well as on the specific plastic deformation mechanism. In this work, the proposal of Gryaznov & Trusov (1993) is employed, but here the power series in Eq. (2) is truncated to $n = 2$ terms and a modification in the logarithmic term is introduced:

$$\sigma_y = \sigma_{y_0} + k_0 \ln\left(\frac{d_0}{d}\right) + k_1 d^{-\frac{1}{2}} + k_2 d, \tag{3}$$

for $d \leq d_0$, where d_0 is a measure of a coarse grain, $d_0 = O(10\mu\text{m})$, and σ_{y_0} is a parameter. As it will be demonstrated in the present analysis, Eq. (3) allows to account for grain size effects, within a wide range of d ($O(10\,\text{nm}) < d < O(10\mu\text{m})$), without specifying a critical grain size, as done e.g. by Khan et al. (2006) and Pande & Cooper (2009).

In general, it is well known that coarse-grained FCC metals present pronounced work hardening, while those with ultrafine or nanocrystalline grains tend to present a perfect plastic behavior (i.e. negligible hardening) (Meyers, Mishra & Benson, 2006; Khan, Farrokh & Takacs, 2008a,b). In this respect, analogously to Eq. (3), a dependence on the grain size will be assigned to parameter A_∞:

$$fA_\infty = A_{\infty_0} - \left[k_0^A \ln\left(\frac{d_0}{d}\right) + k_1^A d^{-\frac{1}{2}} + k_2^A d\right], \tag{4}$$

for $d \leq d_0$, where k_n^A, $n = \{0, 1, 2\}$, and and A_{∞_0} are material constants. Other parameters in Eq. (1), c and δ, are assumed to be constants in this work. In summary it is expected from Eqs. (3) and (4) that the initial yield stress increases and that the saturation A_∞ decreases or increases, depending on the material, with grain refinement.

CONSTITUTIVE MODEL CALIBRATION

The main objective of this section is to describe an identification procedure of the proposed model from available data. The material parameters involved in Eqs. (1), (3) and (4) are actually evaluated using the experimental data provided by Farrokh & Khan (2009) for aluminum and copper[1], and by Liu et al. (2015) for titanium. The data from *quasi*-static laboratory tests performed by Ahn, Huh & Yoon (2015) on samples of coarse-grained titanium are also used. Once the identification procedure is achieved, the ability of the proposed model to represent the stress-strain curve of metals considering a wide range of grain sizes, from coarse to nanocrystalline grains is therefore assessed. The coarse grain sizes for the aluminum, copper, and titanium are $d_0 = 50\mu$m, $d_0 = 50\mu$m, and $d_0 = 20\mu$m, respectively.

Firstly, the material parameters of Eq. (1) are identified considering the coarse-grain stress-strain curves of each material, thus providing the values of parameters $\sigma_y|_{d=d_0}$, $A_\infty|_{d=d_0}$, c, and δ for the aluminum, copper and titanium given in Tab. 1.

The ultrafine and nanocrystalline grains have the following sizes: 693 nm, 166 nm, 82 nm and 75 nm for aluminum, 720 nm, 350 nm, 118 nm, 51 nm, 32 nm, 27 nm and 22 nm for copper, and 174 nm, 107 nm, 82 nm, 58 nm, 43 nm for titanium samples. Once the parameters c and δ are determined, the values of σ_y and A_∞ can therefore be identified for the ultrafine and nanocrystalline grains of each material. The results of the identification procedure are presented in Tab. 1.

The curves of σ_y and A_∞, as function of $\frac{1}{\sqrt{d}}$, for the aluminum, copper and titanium are depicted in Figs. 1, 2, and 3 respectively. Based on these adjusted values, the parameters of Eq. (3), $\{\sigma_{y_0}, k_0, k_1, k_2\}$ (Figs. 1(a), 2(a), and 3(a)), and of Eq. (4), $\left\{A_{\infty_0}, k_0^A, k_1^A, k_2^A\right\}$ (Figs. 1(b), 2(b), and 3(b)) are identified. Obtained values are presented respectively in Tabs. 2 and 3.

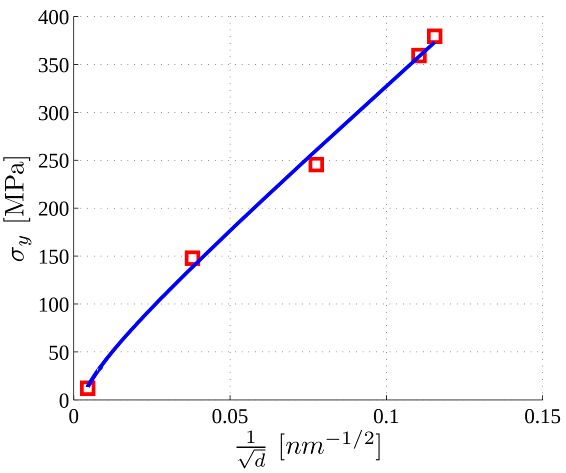
[1] These experimental results were originally presented in the works (Khan, Farrokh & Takacs, 2008a,b), respectively for aluminum and copper.

Table 1 – Adjusted parameters of Eq. (1), for aluminum, copper and titanium.

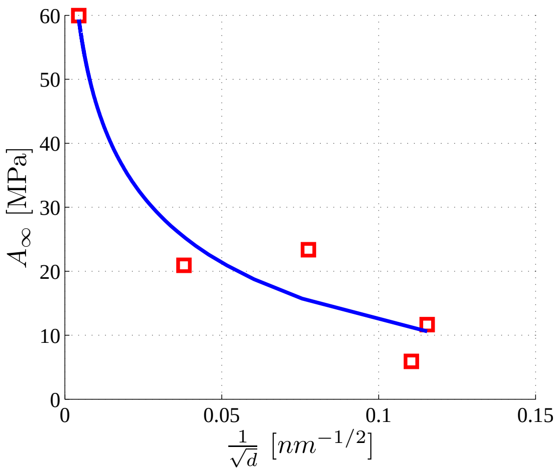
Mat.	d [nm]	σ_y [MPa]	A_∞ [MPa]	c [−]	δ [−]
Aluminum	5×10^4	12.1	60.0	2.1	44.6
	693	147.7	20.9	–	–
	166	245.4	23.3	–	–
	82	359.2	5.9	–	–
	75	379.3	11.7	–	–
Copper	5×10^4	22.7	114.1	0.9	11.6
	720	263.3	96.3	–	–
	350	287.8	93.5	–	–
	118	358.6	84.2	–	–
	51	567.9	68.7	–	–
	32	681.0	39.2	–	–
	27	697.9	60.4	–	–
	22	728.3	63.6	–	–
Titanium	2×10^4	347.6	96.2	10.9	38.4
	174	500.1	118.5	–	–
	107	847.4	128.5	–	–
	82	1190.3	113.6	–	–
	58	1598.9	137.9	–	–
	43	1850.2	85.0	–	–

Analyzes of results depicted in Figs. 1, 2, and 3, and in Tabs. 2, 3, indicated that the initial yield stress increases with grain refinement for all considered materials. However, differently to what is observed for the FCC materials (aluminum and copper) the titanium saturation hardening A_∞ increases with grain refinement. In addition, considering the FCC materials, the terms proportional to d could be neglected in the adjustment of both Eqs. (3) and (4). This means that the values of parameters k_2 and k_2^A are close to zero for the aluminum and copper. As regards the titanium calibration, it is possible to neglect the terms proportional to $\ln\left(\frac{d_0}{d}\right)$, i.e. $k_0 \approx 0$ in Eq. (3). It should be underlined that considering $k_0^A \approx 0$ and $k_2^A \approx 0$ in Eq. (4) the expression of A_∞ reduces to that originally given by the *Hall-Petch* equation. It should also be mentioned that the point referent to the grain size $d = 43\,\text{nm}$ was excluded from the analysis since the material failed under low deformation level.

Once the constitutive model has been adjusted, it can be employed in subsequent analyses for correlation with experi-



(a)



(b)

Figure 1 – Graphs for the aluminum: (a) σ_y vs. $\frac{1}{\sqrt{d}}$; (b) A_∞ vs. $\frac{1}{\sqrt{d}}$.

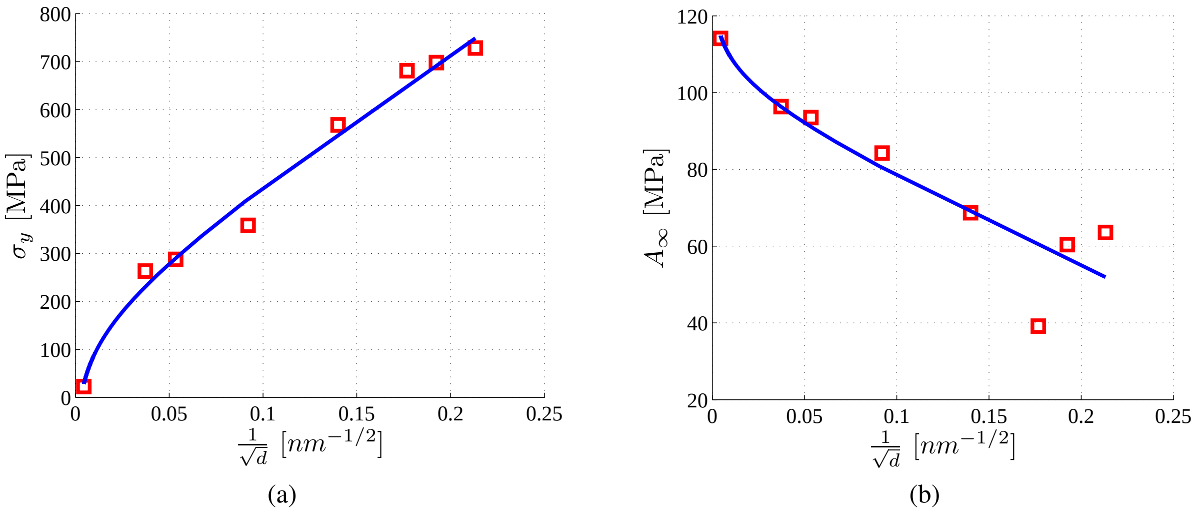


Figure 2 – Graphs for the copper: (a) σ_y vs. $\frac{1}{\sqrt{d}}$; (b) A_∞ vs. $\frac{1}{\sqrt{d}}$.

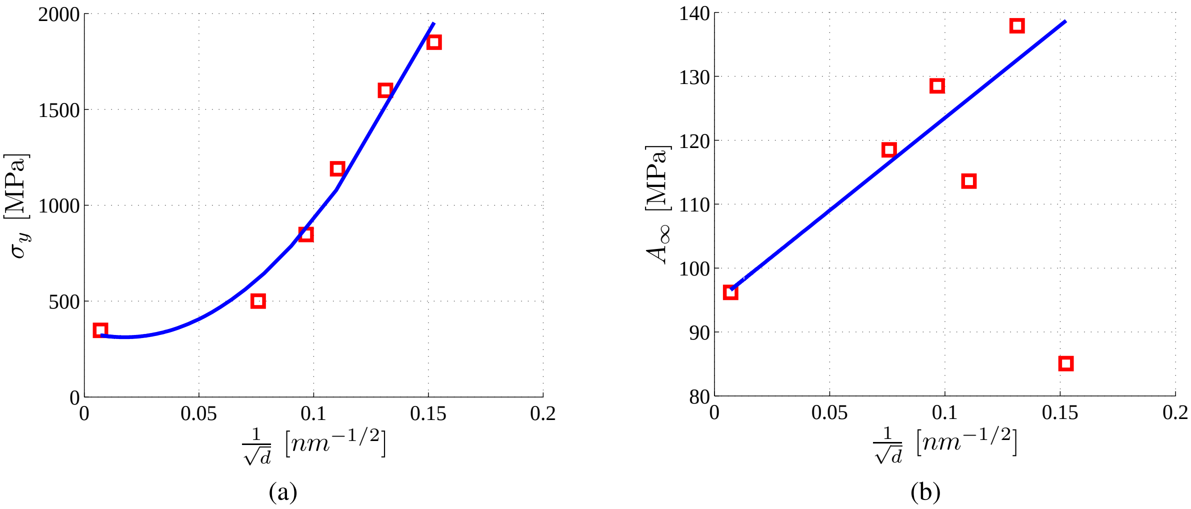


Figure 3 – Graphs for the titanium: (a) σ_y vs. $\frac{1}{\sqrt{d}}$; (b) A_∞ vs. $\frac{1}{\sqrt{d}}$.

ments. The predicted stress-strain curves are compared with reference experimental data. Figures 4, 5, and 6 present such curves for the aluminum, copper and titanium, respectively. It is clearly observed from Fig. 4 that the constitutive model is able to represent the material behavior from a coarse to a nanometric grain size for the whole considered deformation range. The worst scenario is for the grain of size 693 nm, for which the model presented a slightly smaller *quasi*-linear hardening when compared to the experimental response.

Figure 5 also shows a good concordance between the model representation and experiments. Although a discrepancy is observed for the grain sizes of 720 nm and 118 nm, the model demonstrated its aptitude to adequately predict the material

Table 2 – Adjusted parameters of Eq. (3)**, for aluminum, copper and titanium.**

Mat.	σ_{y_0} [MPa]	k_0 $[-]$	k_1 $[\sqrt{\text{nm}}.\text{MPa}]$	k_2 $\left[\frac{\text{MPa}}{\text{nm}}\right]$
Al	$0,86$	$-7,1$	2823	0
Cu	$17,55$	$-29,5$	2359	0
Ti	$339,8$	0	-3176	9×10^4

Table 3 – Adjusted parameters of Eq. (4), for aluminum, copper and titanium.

Mat.	A_{∞_0} [MPa]	k_0^A $[-]$	k_1^A $\left[\sqrt{\text{nm}}.\text{MPa}\right]$	k_2^A $\left[\frac{\text{MPa}}{\text{nm}}\right]$
Al	$59,1$	$-8,3$	$-48,64$	0
Cu	$115,7$	$-2,8$	196	0
Ti	$94,5$	0	$-289,6$	0

response for all the strain range and a wide range of grain sizes. For the FCC materials considered in this work, it becomes clear from the analysis of Figs. 4 and 5 that the grain refinement produces: (i) an increasing in the yield strength; and (ii) a reduction in the amount and rate of hardening. Proposed model can predict this last effect by the dependence of the saturation hardening parameter A_∞ on the grain size, what also influences the *quasi*-linear hardening rate $A_\infty c$ in advanced stages of deformation.

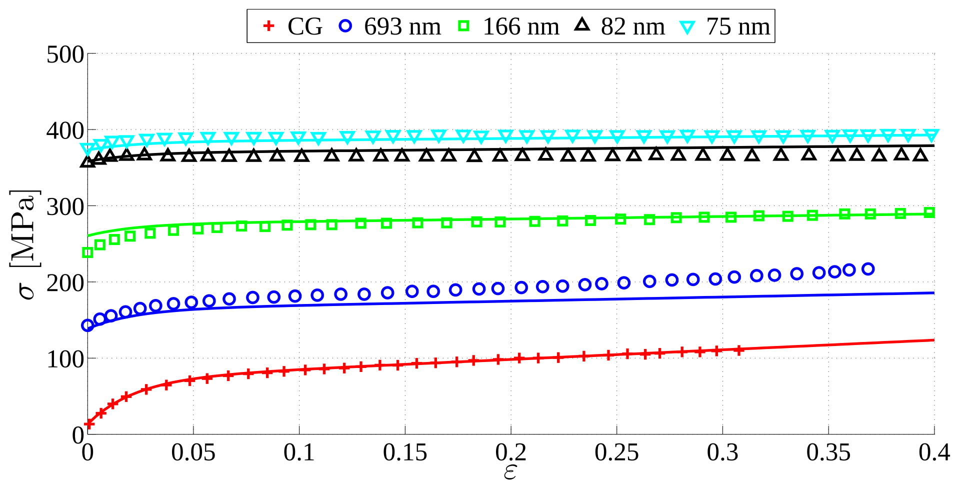


Figure 4 – Stress-strain curves for aluminum with different grain sizes. Comparison between model prediction (continuum lines) and experimental data (Khan, Farrokh & Takacs, 2008b; Farrokh & Khan, 2009) (points).

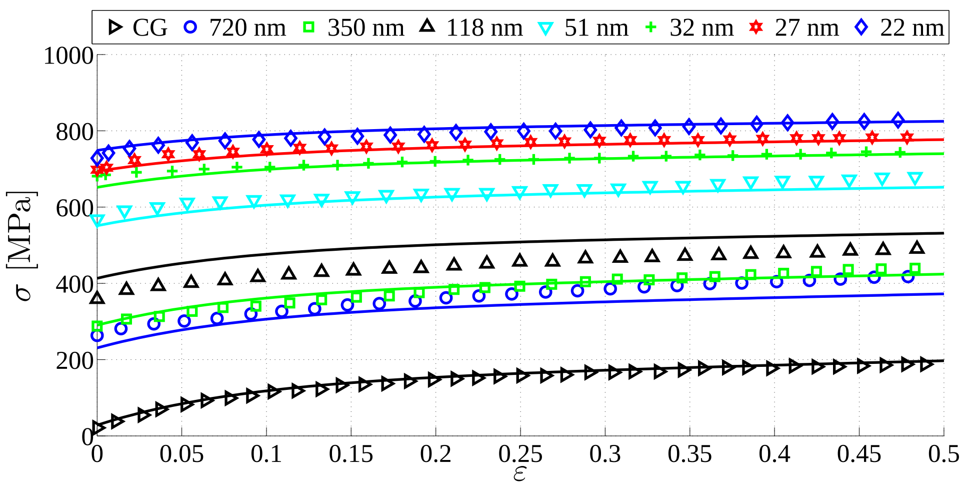


Figure 5 – Stress-strain curves for copper with different grain sizes. Comparison between model prediction (continuum lines) and experimental data (Khan, Farrokh & Takacs, 2008a; Farrokh & Khan, 2009) (points).

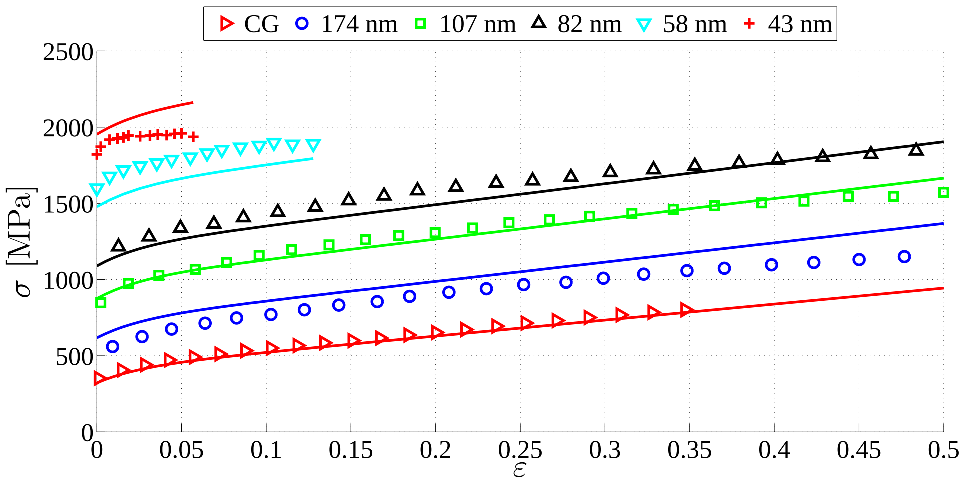


Figure 6 – Stress-strain curves for titanium with different grain sizes. Comparison between model prediction (continuum lines) and experimental data (Liu et al., 2015; Ahn et al., 2015) (points).

Quite similar conclusions may be drawn regarding titanium material, graphs depicted in Fig. 6, for the case of coarse grain and the grains of sizes of 174 nm, 107 nm, 82 nm. A higher discrepancy is observed for the size of 58 nm. However, the model is fairly capturing the material response for the grain size 43 nm. Work (Liu et al., 2015) has already reported similar difficulties when attempting to adjust the *Khan-Liang-Farrokh* for titanium considering the grain size of 43 nm. The authors mentioned that such gaps could be caused by the higher level of impurities in the titanium samples due to the long period of mechanical milling. Interestingly noting, the experimental measurements for titanium sizes of 174 nm, 107 nm, 82 nm indicate a slight reduction in the hardening rate when strain level exceeds 0.30. This feature is not accounted for by the proposed model since the nonlinear terms of Eq. (1), $A_\infty\left[1-\exp\left(-\delta\varepsilon\right)\right]$, are no more evolving and the hardening increase is controlled by the constant hardening rate term $A_\infty c\varepsilon$.

In summary, the model developed here provides results similar to the *Khan-Liang-Farrokh* model for the aluminum, copper (Farrokh & Khan, 2009) and titanium (Liu et al., 2015) under *quasi*-static conditions and at room temperature. However, the constitutive model proposed in this work is not limited to materials with ultrafine or nanocrystalline grains, being able to be used for predicting the behaviour of the same materials but with coarse grains.

CONCLUSION

In the present work a *quasi*-static rigid-plastic model was formulated and adjusted. This model considers grain size effects on material hardening. The constitutive model was calibrated considering experimental data from literature for pure aluminum, copper and titanium. The model proved to be suitable to predict experimental data, within all considered strain range and for a wide grain size range, from coarse-grained to nanocrystalline samples. In general, the present model has a great potential to serve as basis to further investigations, in which grain size effects be considered, e.g., in dynamic loading conditions at different temperature levels. However, the employed hardening rule given in Eq. (1) was derived in an heuristic reasoning, considering physical aspects related to FCC metals (dos Santos et al., 2016). Due to this reason, present hardening law was not able to predict the hardening rate reduction in the 174 nm, 107 nm, 82 nm grain-sized titanium response after a given strainning. This feature can acomplished in the present hardening law by allowing the parameter c to depend on the grain size, or considering micromechanical features associated with HCP metals, e.g., modifying the hardening equation in order to account for twin formation.

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