**Effect of creep parameters on the steady-state flow stress of pure metals**

**processed by high-pressure torsion**

Roberto B. Figueiredo1,\*, Kaveh Edalati2,\* and Terence G. Langdon 3

1 Department of Metallurgical and Materials Engineering, Universidade Federal de Minas Gerais, Belo Horizonte, MG 31270-901, Brazil

2 WPI, International Institute for Carbon-Neutral Energy Research (WPI-I2CNER), Kyushu University, Fukuoka 819-0395, Japan

3 Materials Research Group, Department of Mechanical Engineering, University of Southampton, Southampton SO17 1BJ, U.K.

**Abstract**

The correlation between the flow stress and grain size for severely deformed metals remains undefined because the conventional Hall-Petch relationship ignores the expected contributions from thermally-activated phenomena in nanomaterials and also it fails to explain the reported strain softening of metals having low melting temperatures. In this study, the contribution of thermally-activated creep mechanisms to the room temperature flow stress is evaluated for 31 pure metals processed by high-pressure torsion. The steady-state grain size and the hardness of these metals are first compared to theoretical predictions from high temperature creep mechanisms. It is shown that these mechanisms are not able to predict the flow stress, although the data from metals with low melting temperatures fall close to the theoretical prediction for high-temperature grain boundary sliding suggesting a possible explanation for the unusual softening of these metals. Nevertheless, a detailed analysis demonstrates that a modified model for grain boundary sliding at low temperature provides the capability of correctly predicting the flow stresses for metals having both high and low melting temperatures. The results confirm the significance of thermally-activated phenomena in determining the flow stress of nanomaterials processed by severe plastic deformation.

**Keywords:** creep; grain boundary sliding; high-pressure torsion; severe plastic deformation (SPD); ultrafine-grained materials.

\*Corresponding authors: figueiredo@demet.ufmg.br / kaveh.edalati@kyudai.jp

1. **Introduction:**

An understanding of the parameters that control the mechanical strength of solid materials has been a major topic of research over many decades in materials science and engineering. It is widely understood that atomic bond parameters and crystalline structure affect some mechanical properties such as the elastic modulus. Although it is known that these parameters also affect the yield stress and plastic deformation of high purity single crystals, the correlations are not clear for polycrystalline alloys since other parameters, such as impurities, defects in the crystalline structure and second phase particles, appear to be important [[1](#_ENREF_1)].

The generation of lattice defects by plastic deformation is a well-established strategy for controlling the yield stress but over the last two decades the introduction of severe plastic deformation (SPD) processing techniques [[2-4](#_ENREF_2)] have provided an opportunity for producing virtually unlimited quantities of defects in the crystalline structure of materials. This could lead to an amorphization of the structure but it is now known that dynamic recovery and recrystallization takes place and controls the maximum densities of defects for each material. For example, there is a minimum grain size and a maximum steady-state strength for each material subjected to SPD [[5-7](#_ENREF_5)].

Several reports have attempted to correlate the minimum grain size and the steady-state flow stress of different materials to their fundamental properties [[5-9](#_ENREF_5)]. An evaluation of the data confirms the expected trends such that the steady-state flow stress increases with increasing atomic bond energy and the minimum grain size decreases with increasing activation energy for diffusion. Despite these reports, there is no generally acceptable correlation between the steady-state flow stress, the minimum grain size and the fundamental properties of SPD-processed materials because of uncertainties regarding the deformation mechanisms.

The concept that materials processed by SPD reach a condition of deformation in which the rate of generation of defects equals the rate of defect recovery leads directly to the proposal that there is steady-state deformation through the occurrence of dynamic recovery and recrystallization mechanisms. Furthermore, it is widely accepted that materials undergo steady-state deformation through creep at high homologous temperatures. It is also known that thermally-activated phenomena become active at lower homologous temperatures in very fine-grained materials [[10](#_ENREF_10), [11](#_ENREF_11)] and very high diffusion rates, which are significant in the occurrence of creep, have been specifically reported in materials processed by SPD [[12](#_ENREF_12), [13](#_ENREF_13)]. Furthermore, the grain sizes of materials processed by SPD are small, usually <1 μm, and deformation mechanisms typically observed in high temperature creep, such as grain boundary sliding, have been reported extensively at low temperatures [[14-17](#_ENREF_14)]. There are also reports of SPD-induced softening at room temperature in low melting point materials such as Al, Mg, In, Sn, Zn and Pb where this is different from the general trend of hardening observed in metals with higher melting points [[18](#_ENREF_18)]. Although the Hall-Petch mechanism can generally explain the hardening of metals with high melting points, it cannot account for the creep-like softening phenomena reported in fine-grained metals with low melting temperatures.

Recent reports characterizing the low temperature deformation mechanisms of fine-grained materials have included *in situ* observations of grain rotation [[19](#_ENREF_19)] and dislocation absorption at grain boundaries [[20](#_ENREF_20)]. Thus, these observations confirm that creep deformation can play a significant role in the deformation of fine-grained materials even at low temperatures.

In practice, high temperature creep mechanisms are usually described through an equation of the form [[21-23](#_ENREF_21)]:

$\dot{ε}=\frac{ADGb}{kT}\left(\frac{σ}{G}\right)^{n}\left(\frac{b}{d}\right)^{p}$ (1)

where $\dot{ε}$ is the strain rate, *A* is a constant, *D* is the diffusion coefficient, *G* is the shear modulus, *b* is the Burgers vector, *k* is Boltzmann’s constant, *T* is the absolute temperature, $σ$ is the effective stress, *d* is the spatial grain size and *n* and *p* are the stress and the inverse grain size exponents, respectively. The constants *A*, *n* and *p* and the type of diffusion are all dependent upon the creep mechanism. Therefore, the creep kinetics depend on the temperature, stress and grain size, the fundamental properties of the material and on the deformation mechanism in terms of dislocation climb and glide, grain boundary sliding and Coble and Nabarro-Herring diffusion creep.

Despite the validity of Eq. (1) for a wide range of materials and creep phenomena, it was shown recently that the rate controlling equation for grain boundary sliding must be adjusted in order to reasonably account for the large stresses developed during low temperature deformation [[24](#_ENREF_24)]. This adjustment leads to a rate equation which incorporates grain boundary diffusion, $D\_{gb}$, and is written in the form [24]:

$\dot{ε}=\frac{10δD\_{gb}}{d^{3}}\left[exp\left(\frac{2dσ^{2}b^{2}}{3GkT}\right)-1\right]$ (2)

Although there are anticipated contributions from creep mechanisms, including grain boundary sliding, to the deformation of SPD-processed metals, there have been few attempts to evaluate the steady-state flow stress of these materials by considering the creep parameters. Recognizing this deficiency, the present research was initiated to evaluate the available data from 31 different pure metals subjected to the same SPD processing technique of high-pressure torsion (HPT) and then to compare their steady-state strength to the general creep equation and to the mechanism of grain boundary sliding. Creep studies usually focus on a single material tested under different conditions of temperature and stress whereas the variables in the present work relate to the fundamental properties of the different materials. The analysis is focused specifically on materials of high purity in order to reduce any effects from impurities to the deformation behavior.

1. **Experimental data**

Pure metals were selected where their steady-state hardness after HPT processing is readily available. These materials had different crystal structures including bcc (vanadium, chromium, iron, niobium, molybdenum, tantalum and tungsten), fcc (aluminum, nickel, copper, palladium, silver, platinum, gold and lead), hcp (magnesium, scandium, titanium, cobalt, zinc, zirconium, hafnium and rhenium), tetragonal (indium and tin), rhombohedral (bismuth), diamond cubic (silicon and germanium), hexagonal (tellurium and neodymium) and complex cubic (manganese). For each material, the melting temperature, Burgers vector, shear modulus, lattice diffusion and grain boundary diffusion coefficients and the steady-state grain size and strength were collected from the literature and these data are summarized in Table 1 [[5](#_ENREF_5), [8](#_ENREF_8), [18](#_ENREF_18), [25-52](#_ENREF_25)]. Several points should be noted regarding this summary. First, the Burgers vector of Mn is not readily available since its crystal structure is not well documented and therefore the minimum interatomic distance was taken as *b* for this material [[52](#_ENREF_52)]. Second, the pre-exponential value and the activation energy for grain boundary diffusion for some materials were not readily available and therefore the present analysis considered estimated values from general trends available in the literature [[49](#_ENREF_49), [51](#_ENREF_51)]. Third, the data for the purity level, grain size and the hardness observed after HPT processing of the different materials were collected from reports published in the literature on HPT processing.

Table 1 – Summary of data for melting temperature (*Tm*), homologous temperature at 298 K (298/*Tm*), Burgers vector (*b*), shear modulus (*G*) at 298 K, pre-exponential (*D0,l*) and activation energy (*Ql*) for lattice self-diffusion, pre-exponential (*δD0,gb*) and activation energy (*Qgb*) for grain boundary diffusion, purity, grain size (*ds*) and hardness (*Hv*) for different materials processed by HPT.

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Metal** | ***Tm*** | **298/*Tm*** | ***b*** | ***G*** | ***D0,l*** | ***Ql*** | ***δD0,gb*** | ***Qgb*** | **Purity** | ***dS*** | ***Hv*** |
|  | (K) [[26-28](#_ENREF_26)] |  | (nm)[[25](#_ENREF_25)] | (GPa) [[26-28](#_ENREF_26)] | (10-14m2s-1)[[29](#_ENREF_29), [30](#_ENREF_30)] | (kJ.mol-1)[[29](#_ENREF_29), [30](#_ENREF_30)] | (10-14m3s-1) | (kJ.mol-1) | (%) | (μm) | (GPa)[[8](#_ENREF_8)] |
| 12Mg | 922 | 0.32 | 0.3197 | 17.3 | 1.75 | 138 | 500[[31](#_ENREF_31)] | 92[[31](#_ENREF_31)] | 99.9 | 1.0[[32](#_ENREF_32)] | 0.3420 |
| 13Al | 933 | 0.32 | 0.2864 | 26.2 | 0.176 | 126 | 5[[31](#_ENREF_31)] | 84[[31](#_ENREF_31)] | 99.99 | 1.9[[33](#_ENREF_33)] | 0.313 |
| 14Si | 1685 | 0.18 | 0.2352 | 39.7 | 20 | 424 | 2.25[[46](#_ENREF_46)] | 135[[46](#_ENREF_46)] | 99.999 | 0.017[[34](#_ENREF_34)] | 6.355 |
| 21Sc | 1811 | 0.16 | 0.3256 | 29.1 | 0.5 | 261d | 1.5a | 155a | 99.999 | --- | 0.981 |
| 22Ti | 1940 | 0.15 | 0.2896 | 45.6 | 0.66 | 169 | 450[[47](#_ENREF_47)] | 187[[47](#_ENREF_47)] | 99.4 | 0.2[[35](#_ENREF_35)] | 2.599 |
| 23V | 2175 | 0.14 | 0.2618 | 46.7 | 0.36 | 308 | 5[[31](#_ENREF_31)] | 209[[31](#_ENREF_31)] | 99.9 | 0.33[[36](#_ENREF_36)] | 2.354 |
| 24Cr | 2133 | 0.14 | 0.2498 | 115 | 1.6 | 339 | 0.5[[31](#_ENREF_31)] | 192[[31](#_ENREF_31)] | 99.9 | 0.2[[37](#_ENREF_37)] | 4.756 |
| 25Mn | 1517 | 0.20 | 0.225b  | 79.5 | 0.88 | 225e | 3.9c | 125c | 99.9 | --- | 4.086 |
| 26Fe | 1809 | 0.16 | 0.2482 | 81.6 | 1.9 | 240 | 110[[31](#_ENREF_31)] | 174[[31](#_ENREF_31)] | 99.96 | 0.2[[50](#_ENREF_50)] | 3.02 |
| 27Co | 1768 | 0.17 | 0.2497 | 82 | 0.55 | 289 | 1.5a | 151a | 99.99 | 0.12[[38](#_ENREF_38)] | 3.53 |
| 28Ni | 1728 | 0.17 | 0.2492 | 75 | 1.77 | 285 | 0.35[[31](#_ENREF_31)] | 115[[31](#_ENREF_31)] | 99.996 | 0.24[[18](#_ENREF_18)] | 3.021 |
| 29Cu | 1357 | 0.22 | 0.2556 | 48.3 | 0.35 | 204 | 0.5[[31](#_ENREF_31)] | 104[[31](#_ENREF_31)] | 99.99 | 0.37[[18](#_ENREF_18)] | 1.298 |
| 30Zn | 693 | 0.43 | 0.2665 | 41.9 | 0.13 | 92 | 1.3[[31](#_ENREF_31)] | 61[[31](#_ENREF_31)] | 99.99 | 5.1[[18](#_ENREF_18)] | 0.362 |
| 32Ge | 1210 | 0.25 | 0.2450 | 29.6 | 25 | 318 | 0.02[[48](#_ENREF_48)] | 133[[48](#_ENREF_48)] | 99.999 | 0.024[[39](#_ENREF_39)] | 2.986 |
| 40Zr | 2125 | 0.14 | 0.3179 | 35 | 0.0021 | 113 | 74[[47](#_ENREF_47)] | 188[[47](#_ENREF_47)] | 99.9 | 0.2[[40](#_ENREF_40)] | 2.532 |
| 41Nb | 2740 | 0.11 | 0.2864 | 37.5 | 1.1 | 402 | 5[[31](#_ENREF_31)] | 263[[31](#_ENREF_31)] | 99.9 | 0.24[[37](#_ENREF_37)] | 2.354 |
| 42Mo | 2888 | 0.10 | 0.2725 | 125.6 | 2.77 | 465 | 5.5[[31](#_ENREF_31)] | 263[[31](#_ENREF_31)] | 99.9 | 0.34[[36](#_ENREF_36)] | 6.669 |
| 46Pd | 1825 | 0.16 | 0.2751 | 43.6 | 0.205 | 266 | 0.97a | 138a | 99.9 | 0.22[[18](#_ENREF_18)] | 2.127 |
| 47Ag | 1234 | 0.24 | 0.2889 | 27 | 0.278 | 182 | 0.45[[31](#_ENREF_31)] | 90[[31](#_ENREF_31)] | 99.99 | 0.48[[41](#_ENREF_41)] | 0.941 |
| 49In | 429 | 0.69 | 0.3251 | 3.7 | 2.7 | 79 | 3.9c | 35c | 99.999 | 320[[18](#_ENREF_18)] | 0.014 |
| 50Sn | 505 | 0.59 | 0.3022 | 18.4 | 10.7 | 105 | 0.3[[49](#_ENREF_49)] | 40[[49](#_ENREF_49)] | 99.9 | 135[[18](#_ENREF_18)] | 0.068 |
| 52Te | 722 | 0.41 | 0.2884 | 17 | 130 | 169 | 3.9c | 60c | 99.99 | --- | 0.723 |
| 60Nd | 1294 | 0.23 | 0.3628 | 16.3 | 0.5 | 186d | 3.9c | 107c | 99.9 | --- | 0.720 |
| 72Hf | 2500 | 0.12 | 0.3127 | 56 | 0.073 | 174 | 35[[47](#_ENREF_47)] | 212[[47](#_ENREF_47)] | 99.99 | 0.18[[42](#_ENREF_42)] | 3.481 |
| 73Ta | 3253 | 0.09 | 0.2856 | 69 | 0.124 | 413 | 5.7[[31](#_ENREF_31)] | 280[[31](#_ENREF_31)] | 99.9 | 0.18[[37](#_ENREF_37)] | 4.132 |
| 74W | 3683 | 0.08 | 0.2741 | 160.6 | 15.3 | 626 | 33[[31](#_ENREF_31)] | 385[[31](#_ENREF_31)] | 99 | 0.1[[43](#_ENREF_43)] | 9.086 |
| 75Re | 3453 | 0.09 | 0.2741 | 181 | 1 | 511 | 1.5a | 295a | 99.9 | 0.1 [[44](#_ENREF_44)] | 7.262 |
| 78Pt | 2042 | 0.14 | 0.2775 | 61 | 0.22 | 279 | 0.97a | 154a | 99.9 | --- | 2.501 |
| 79Au | 1336 | 0.22 | 0.2884 | 27.7 | 0.107 | 177 | 0.019[[49](#_ENREF_49)] | 112[[49](#_ENREF_49)] | 99.999 | 0.52[[41](#_ENREF_41)] | 0.804 |
| 82Pb | 600 | 0.50 | 0.3500 | 5.6 | 1.37 | 109 | 8[[31](#_ENREF_31)] | 66[[31](#_ENREF_31)] | 99.999 | 100[[18](#_ENREF_18)] | 0.051 |
| 83Bi | 545 | 0.55 | 0.3071 | 12.8 | 0.88 | 81e | 3.9c | 45c | 99.99 | 20 [[45](#_ENREF_45)] | 0.137 |

aAdapted from suggested general equations [[51](#_ENREF_51)] for fcc (*δD0=9.7×10-15; Qgb = 75.4×Tm*) and hcp (*δD0=1.5×10-14; Qgb = 85.4×Tm*) metals

bTaken as the minimum interatomic distance [[52](#_ENREF_52)]

cThe values for *δD0=3.9×10-14* and *Qgb = 82.5×Tm* were estimated from average correlations [[51](#_ENREF_51)] between bcc., fcc and hcp structures.

dAdapted from a suggested general equation [[49](#_ENREF_49)] for hcp (D0,l=0.5×10-14; Ql = 144×Tm)

eThe values for *D0,l=0.88×10-14* and *Ql = 148×Tm* were estimated from average correlations [[49](#_ENREF_49)] between bcc, fcc and hcp structures.

1. **General creep analysis**

Equation (1) can be rearranged to isolate the flow stress by expressing in the form:

$σ=\left[\frac{\dot{ε}kTG^{n}}{ADGb}\left(\frac{d}{b}\right)^{p}\right]^{^{1}/\_{n}}$ (3)

It is then possible to evaluate the correlations between $σ$ and the fundamental parameters *b*, *G* and the activation energy for diffusion. Thus, Fig. 1 shows the effective stress for each material after HPT processing, considered as H/3 [[53](#_ENREF_53)] where H is the hardness, plotted as a function of these various parameters. It is reasonable to anticipate that diffusion plays a significant role in the deformation of materials processed by SPD [[6-9](#_ENREF_6)]. The graphs show that the trends generally agree with the equation for creep since the flow stress increases with decreasing Burgers vector and with increasing shear modulus and activation energy for grain boundary diffusion. The flow stress also increases with increasing activation energy for lattice diffusion (not shown here). Nevertheless, the scatter in the $σ×b$ plot in Fig. 1(a) is very large since materials with similar Burgers vectors may display flow stresses that vary by up to 2 orders of magnitude. The scatter is also quite large in the $σ×G$ and $σ×Q\_{gb}$ plots in Figs. 1(b) and 1(c), respectively. For example, Zn and Si have similar values for the shear modulus but there is an order of magnitude in difference in the flow stress for these two materials after HPT. Also, although the activation energy for grain boundary diffusion in Nb is over twice the value for Ni, the former displays a slightly lower strength than the latter. Thus, it is clear from this analysis that the strength of materials processed by HPT cannot be simply predicted by any of these parameters alone.



Figure 1 – Flow stress plotted as a function of (a) the Burgers vector, (b) the shear modulus and (c) the activation energy for grain boundary diffusion.

In practice, the general creep relationship at Eq. (1) can be rewritten to isolate the normalized stress in the form:

$\left(\frac{σ}{G}\right)^{n}=\frac{\dot{ε}kT}{AbDG}\left(\frac{b}{d}\right)^{p}$ (4)

In order to estimate the exponent *n*, it is useful to plot the flow stress normalized by the shear modulus as a function of the strain rate normalized by the temperature, Burgers vector, diffusion coefficient, shear modulus and grain size. Figure 2 shows examples of such plots considering (a,b) lattice self-diffusion, *Dl*, and (c,d) grain boundary diffusion, *Dgb*, and different values for the grain size exponent, *p*. A strain rate of 10-3 s-1 and a testing temperature of 298 K were considered for all materials in the present analysis where these values correspond to quasi-static hardness testing at room temperature (RT). Thus, in practice, the normalized strain rate accounts for the effect of the diffusion coefficients and the shear modulus from the different materials. In order to include the grain boundary diffusion, *Dgb*, the grain boundary width, δ, is used in place of the Burgers vector.

The contribution of grain size to the flow mechanism is considered negligible (*p* = 0) in Fig. 2(a) and it is apparent that the experimental data span over a large range of values. This large range for the data is attributed to the large differences in the lattice self-diffusion coefficient of the different materials. Inspection of the data suggests two basic trends. For normalized strain rates lower than 1020, which mainly correspond to metals with low melting temperatures, the normalized stress increases with an increase in the normalized strain rate and it is possible to estimate a value of *n* ≈ 15 in this range. This corresponds to a strain rate sensitivity of ~0.07. The normalized stress saturates at ~0.02 for materials with normalized strain rates larger than ~1020 and this suggests a negligible strain rate sensitivity so that the contribution from thermally-activated phenomena appears to be negligible for metals located in this range. The value of *n* is larger than the values usually observed in creep mechanisms since a value of *n* ≈ 4.5 is typical for a dislocation climb mechanism [[22](#_ENREF_22), [54](#_ENREF_54)].

The normalized stress is plotted as a function of normalized strain rates considering the contribution of the grain size in the plots in Figs. 2(b), 2(c) and 2(d). Figures 2(b) and 2(c) investigate the contribution of grain boundary sliding considering coarse grains and fine grains, respectively. It has been shown that grain boundary sliding in coarse-grained materials, designated GBScg, is associated with lattice self-diffusion, an inverse grain size exponent of 1 and a stress exponent of 3 whereas grain boundary sliding in fine-grained materials, designated GBSfg, is associated with grain boundary diffusion, an inverse grain size exponent of 2 and a stress exponent of 2 [[21](#_ENREF_21)]. The theoretical predictions for these mechanisms are also plotted for comparison. It is apparent that the experimental data do not follow a constant trend in these plots and they do not, in general, agree with the theoretical predictions for high temperature grain boundary sliding. However, the data for metals with low melting points fall close to the theoretical predictions for grain boundary sliding in fine-grained materials which suggests that grain boundary sliding should at least partly contribute to the flow stress and the documented softening behavior of these materials.

Finally, the data are plotted as a function of the normalized strain rate considering grain boundary diffusion and a grain size exponent of 3 in Fig. 2(d) where this is similar to the behavior expected for diffusion creep according to the Coble mechanism [[55](#_ENREF_55)]. Again, the data do not follow a constant trend and they do not agree with the theoretical predictions but, as in Fig. 2(c), the data for metals with low melting points fall closer to the theoretical line for Coble creep.



Figure 2 – Normalized stress plotted as a function of the normalized strain rate using the general creep equation by considering (a) *p* = 0 and *Q* = *Q*l for dislocation climb, (b) *p* = 1 and *Q* = *Q*l for grain boundary sliding in coarse-grained materials, (c) *p* = 2 and *Q* = *Q*gb for grain boundary sliding in fine-grained materials and (d) *p* = 3 and *Q* = *Q*gb for Coble creep.

1. **Grain size contribution**

Some high temperature creep mechanisms are not dependent upon grain size (*p* = 0) while others display positive values for *p*, where positive values of *p* mean that the flow stress increases with increasing grain size. However, the present data shows that the grain size follows an inverse trend in relation to the flow stress. For example, in Fig. 3 the grain sizes of the different materials processed by HPT are plotted as a function of the shear modulus and the activation energy for grain boundary diffusion. This shows that the grain size decreases with increasing *G* and *Qgb* where these trends are the opposite to those observed with the flow stress.



Figure 3 – Grain size plotted as a function of (a) the shear modulus and (b) the activation energy for grain boundary diffusion.

The inverse trends between flow stress and grain size, although contradicting the high-temperature creep mechanisms, are expected for low temperature grain boundary sliding and the explanation is given in the next section. The mechanism of grain refinement during SPD processing is attributed to the formation of dislocation cells which gradually evolve to subgrain boundaries and then to high-angle grain boundaries with increasing deformation. Additional grain refinement may take place due to geometrical distortions of the grains caused by the continuous shearing. Therefore, the final grain sizes are expected to be similar to, or possibly smaller than, the average stable subgrain sizes. It is now well-established that the average subgrain size, λ, is inversely proportional to the applied stress so that the following relationship is valid for many materials [[56](#_ENREF_56), [57](#_ENREF_57)].

$\frac{σ}{G}=20\frac{b}{λ}$ (5)

Therefore, the grain size of a material processed by HPT is expected to be inversely proportional to its strength. Figure 4 shows the normalized flow stress plotted as a function of the normalized grain size. The prediction from Eq. (5) is also shown for comparison. The results demonstrate that the majority of materials follow the expected trend such that the grain sizes are similar to, or smaller than, the stable subgrain size. Five materials, Bi, In, Pb, Sn and Zn, display grain sizes larger than the stable subgrain size. This is attributed to the occurrence of grain boundary migration and grain growth after HPT processing. These materials have low melting points so that processing at RT is compatible with high temperature processing. In addition, it should be noted that grain boundary migration is expected to be especially fast in these materials due to their high purity and low melting point.

Careful inspection of Fig. 4 shows that materials with high melting temperatures, such as Co, Re and W and the materials with covalent bonding (Ge and Si), have grain sizes that are significantly smaller than the stable subgrain size. This suggests that the rate of recovery is lower in these materials.



Figure 4 – Normalized stress plotted as a function of the normalized grain size.

1. **Comparison to the mechanism for grain boundary sliding**

The analysis so far shows that the flow stress data for the different materials follow the expected trends with respect to *b*, *G* and *Qgb*. However, the data for any grain size do not follow the expected trend for high temperature creep in which grain refinement is usually associated with softening. Inspection of Fig. 4 shows also that the normalized stress decreases with increasing normalized grain size and this is in agreement with the well-known trend of grain refinement hardening that occurs in most materials at RT. Therefore, the deformation mechanism associated with the steady-state deformation of materials processed by HPT should incorporate this trend of grain refinement hardening at RT.

It was shown earlier that the mechanism of high-temperature grain boundary sliding [[21](#_ENREF_21)] may be adapted to account for the large stresses during low temperature deformation [24]. This mechanism is based on the assumption that grain boundary sliding activates slip systems in adjacent grains at triple junctions of grain boundary ledges. In the absence of any barriers due to subgrain boundaries, these dislocations then glide and pile up to the opposite grain boundary so that the rate-controlling event is the climb of dislocations at the head of the pile-up. At high temperatures, the occurrence of fast grain boundary diffusion prevents a large increase in the density of defects generated by the climb of dislocations. This condition is associated with the exponents of *n* = 2 and *p* = 2 in the general creep relationship given by Eq. (1). However, the conditions of slower diffusion and high stresses at low temperatures can increase the density of defects generated by the climb of dislocations and the rate-controlling mechanisms is no longer described by the general creep equation. The flow stress for this mechanism is estimated from the relationship [[24](#_ENREF_24)]:

$σ=\sqrt{\frac{3GkT}{2db^{2}}ln\left(\frac{\dot{ε}d^{3}}{10δD\_{gb}}+1\right)}$ (6)

It follows that, under conditions of large strain rates, large grain sizes and/or low coefficients of grain boundary diffusion, the value within the logarithmic *ln* function is large so that small variations in grain size have little or no effect on the outcome of this function. In this case, it follows that the flow stress is approximately proportional to the inverse of the square root of the grain size so that this is the expected trend for room temperature deformation of most materials. By contrast, the flow stress is proportional to the grain size under conditions where $^{\dot{ε}d^{3}}/\_{10δD\_{gb}}\ll 1$ which is usually observed during the high temperature deformation of fine-grained materials. It is important to note that low melting point materials display reasonably large coefficient of grain boundary diffusion at RT and this means that the mechanism of grain boundary sliding can predict grain refinement softening at RT for these materials. This is consistent with reports of grain refinement softening in Al [[58](#_ENREF_58), [59](#_ENREF_59)], Mg [[17](#_ENREF_17), [60-62](#_ENREF_60)] and Zn [[63](#_ENREF_63)].

Figure 5 shows the flow stress of the different materials plotted as a function of the right-hand side of Eq. (6). It is observed that the data follow a single trend and they also agree with the estimated values from the mechanism of grain boundary sliding where this agreement is achieved without the use of any adjustable parameters. This agreement confirms that a single mechanism can fully explain de deformation behavior of multiple materials having different crystalline structures and different fundamental properties.

It should be noted that the agreement with the theoretical prediction is very good despite inaccuracies in the determinations of the grain size. It is important to note that the prediction from Eq. (6) considers the spatial grain size whereas most experimental techniques are based on two-dimensional evaluations which usually underestimate the true grain sizes. The flow stress often contains a contribution of a threshold stress at low temperatures where the threshold stress varies depending on the material. The contribution from the threshold stress to the overall flow stress is expected to be larger in samples having larger grain sizes. Also, the relationship $σ=H/3$ is expected to provide a good estimate of the flow stress in fine-grained materials which do not undergo strain hardening. However, the grain sizes are larger than the subgrain sizes in materials with low melting temperatures and these metals are expected to display some strain hardening. Therefore, the slightly larger values of the experimental flow stresses in Bi, In, Pb and Sn are attributed to the significance of a threshold stress and the hardness test effect.

Taken altogether, the current study confirms that the creep parameters and thermally-activated phenomena have a considerable contribution to the flow stress of nanomaterials processed by SPD. This conclusion provides a clear explanation for the softening behavior observed in metals with low melting points [[18](#_ENREF_18)]. This conclusion is in line with the reported low temperature superplasticity in SPD-processed materials [[14-17](#_ENREF_14)] as well as with *in situ* observations of thermally-activated phenomena in nanomaterials [[19](#_ENREF_19)].



Figure 5 – Flow stress plotted as a function of the predicted flow stress for the mechanism of grain boundary sliding as given in Eq. (6).

1. **Summary and conclusions**

1. The experimental hardness and grain size data from 31 different pure materials processed by HPT were used to evaluate the occurrence of steady-state deformation by considering the creep parameters.

2. The flow stress follows the expected trends from creep mechanisms where it increases with decreasing Burgers vector and with increasing shear modulus and activation energy for diffusion.

3. The grain size follows different trends to that observed for the flow stress. This is the opposite of the expected trend in high temperature creep but agrees with the predictions of low temperature grain boundary sliding. The general inverse relationship between the normalized grain size and the normalized flow stress also agrees with the usual observation of grain refinement hardening for most materials at RT.

4. A comparison between the steady-state grain size and hardness with predictions from high temperature creep mechanisms, shows that the data for metals with low melting temperature fall close to the theoretical prediction for high-temperature grain boundary sliding suggesting a possible explanation for the softening of these metals.

5. In general, the results show there is excellent agreement between published experimental data for all materials and the predictions from the mechanism of low temperature grain boundary sliding.

1. **Acknowledgements**

RBF was supported by CNPq (grant No. 302445/2018-8) and FAPEMIG (grant No. TEC-PPM-00324-17). KE thanks Prof. Zenji Horita of Kyushu University for discussion and the MEXT, Japan, for scientific research grants 19H05176 and 21H00150. TGL was supported by the European Research Council under ERC Grant Agreement No. 267464-SPDMETALS.

1. **Data availability**

The raw/processed data required to reproduce these findings cannot be shared at this

time due to technical or time limitations.

**References**

[1] G.E. Dieter, Mechanical Metallurgy, 3rd ed., McGraw-Hill Book Company, London.

[2] R.Z. Valiev, Y. Estrin, Z. Horita, T.G. Langdon, M.J. Zehetbauer, Y.T. Zhu,Fundamentals of Superior Properties in Bulk NanoSPD Materials, Materials Research Letters, 4 (2016) 1-21.

[3] R.Z. Valiev, Y. Estrin, Z. Horita, T.G. Langdon, M.J. Zehetbauer, Y.T. Zhu,Producing bulk ultrafine-grained materials by severe plastic deformation, JOM, 58 (4) (2006) 33-39.

[4] R.Z. Valiev, R.K. Islamgaliev, I.V. Alexandrov,Bulk nanostructured materials from severe plastic deformation, Progress in Materials Science, 45 (2000) 103-187.

[5] K. Edalati, Z. Horita,High-pressure torsion of pure metals: Influence of atomic bond parameters and stacking fault energy on grain size and correlation with hardness, Acta Materialia, 59 (2011) 6831-6836.

[6] F.A. Mohamed, S.S. Dheda,On the minimum grain size obtainable by high-pressure torsion, Materials Science and Engineering: A, 558 (2012) 59-63.

[7] F.A. Mohamed, S.S. Dheda,On the minimum grain size obtainable by equal channel angular pressing, Materials Science and Engineering: A, 580 (2013) 227-230.

[8] K. Edalati, Z. Horita,Correlations between hardness and atomic bond parameters of pure metals and semi-metals after processing by high-pressure torsion, Scripta Materialia, 64 (2011) 161-164.

[9] M.J. Starink, X. Cheng, S. Yang,Hardening of pure metals by high-pressure torsion: A physically based model employing volume-averaged defect evolutions, Acta Materialia, 61 (2013) 183-192.

[10] A.H. Chokshi, A. Rosen, J. Karch, H. Gleiter,On the validity of the hall-petch relationship in nanocrystalline materials, Scripta Metallurgica, 23 (1989) 1679-1683.

[11] B. Cai, Q.P. Kong, L. Lu, K. Lu,Low temperature creep of nanocrystalline pure copper, Materials Science and Engineering: A, 286 (2000) 188-192.

[12] S.V. Divinski, G. Reglitz, H. Rösner, Y. Estrin, G. Wilde,Ultra-fast diffusion channels in pure Ni severely deformed by equal-channel angular pressing, Acta Materialia, 59 (2011) 1974-1985.

[13] G. Wilde, J. Ribbe, G. Reglitz, M. Wegner, H. Rösner, Y. Estrin, M. Zehetbauer, D. Setman, S. Divinski,Plasticity and Grain Boundary Diffusion at Small Grain Sizes, Advanced Engineering Materials, 12 (2010) 758-764.

[14] K. Edalati, Z. Horita, R.Z. Valiev,Transition from poor ductility to room-temperature superplasticity in a nanostructured aluminum alloy, Scientific Reports, 8 (2018) 6740.

[15] R.B. Figueiredo, S. Sabbaghianrad, A. Giwa, J.R. Greer, T.G. Langdon,Evidence for exceptional low temperature ductility in polycrystalline magnesium processed by severe plastic deformation, Acta Materialia, 122 (2017) 322-331.

[16] N.Q. Chinh, P. Szommer, Z. Horita, T.G. Langdon,Experimental evidence for grain‐boundary sliding in ultrafine‐grained aluminum processed by severe plastic deformation, Advanced Materials, 18 (2006) 34-39.

[17] H. Somekawa, T. Mukai,Hall–Petch breakdown in fine-grained pure magnesium at low strain rates, Metallurgical and Materials Transactions A, 46 (2015) 894-902.

[18] K. Edalati, Z. Horita,Significance of homologous temperature in softening behavior and grain size of pure metals processed by high-pressure torsion, Materials Science and Engineering: A, 528 (2011) 7514-7523.

[19] L. Wang, J. Teng, P. Liu, A. Hirata, E. Ma, Z. Zhang, M. Chen, X. Han,Grain rotation mediated by grain boundary dislocations in nanocrystalline platinum, Nature Communications, 5 (2014) 4402.

[20] H. Li, S. Gao, Y. Tomota, S. Ii, N. Tsuji, T. Ohmura,Mechanical response of dislocation interaction with grain boundary in ultrafine-grained interstitial-free steel, Acta Materialia, 206 (2021) 116621.

[21] T.G. Langdon,A unified approach to grain boundary sliding in creep and superplasticity, Acta Metallurgica et Materialia, 42 (1994) 2437-2443.

[22] T.G. Langdon,Identifying creep mechanisms in plastic flow, Zeitschrift fuer Metallkunde/Materials Research and Advanced Techniques, 96 (2005) 522-531.

[23] R.B. Figueiredo, T.G. Langdon,Analysis of the creep behavior of fine-grained AZ31 magnesium alloy, Materials Science and Engineering: A, 787 (2020) 139489.

[24] R.B. Figueiredo, T.G. Langdon,Deformation mechanisms in ultrafine-grained metals with an emphasis on the Hall-Petch relationship and strain rate sensitivity, Journal of Materials Research and Technology, 14 (2021) 137-159.

[25] B.D. Cullity, Elements of X-ray diffraction, 2nd ed., Addison-Wesley, London, 1978.

[26] A. Buch, Short handbook of metal elements properties and elastic properties of pure metals, 3rd ed., Krzysztof Biesaga, Warsaw, 2005.

[27] A. Buch, Pure metals properties, a scientific-technical handbook, ASM International/Freund Publishing House, London and Tel Aviv, 1999.

[28] Metals Handbook. Properties and selection of nonferrous alloys and special-purpose materials, ASM International, Metals Park, Ohio, 1990.

[29] P. Shewmon, Diffusion in Solids, 2nd ed., The Minerals, Metals & Materials Society, Warrendale, PA, 1989.

[30] H. Mehrer, Diffusion in Solid Metals and Alloys, Numerical Data and Functional Relationships in Science and Technology, Springer-Verlag, Berlin, 1990.

[31] H.J. Frost;, M.F. Ashby, Deformation-Mechanism Maps: The Plasticity and Creep of Metals and Ceramics, Pergamon Press, Oxford, U.K., 1982.

[32] K. Edalati, A. Yamamoto, Z. Horita, T. Ishihara,High-pressure torsion of pure magnesium: Evolution of mechanical properties, microstructures and hydrogen storage capacity with equivalent strain, Scripta Materialia, 64 (2011) 880-883.

[33] K. Edalati, Y. Ito, K. Suehiro, Z. Horita,Softening of high purity aluminum and copper processed by high pressure torsion, International Journal of Materials Research, 100 (2009) 1668-1673.

[34] R.K. Islamgaliev, R. Kuzel, S.N. Mikov, A.V. Igo, J. Burianek, F. Chmelik, R.Z. Valiev,Structure of silicon processed by severe plastic deformation, Materials Science and Engineering: A, 266 (1999) 205-210.

[35] K. Edalati, E. Matsubara, Z. Horita,Processing Pure Ti by High-Pressure Torsion in Wide Ranges of Pressures and Strain, Metallurgical and Materials Transactions A, 40 (2009) 2079-2086.

[36] S. Lee, K. Edalati, Z. Horita,Microstructures and Mechanical Properties of Pure V and Mo Processed by High-Pressure Torsion, Materials Transactions, 51 (2010) 1072-1079.

[37] S. Lee, Z. Horita,High-Pressure Torsion for Pure Chromium and Niobium, Materials Transactions, 53 (2012) 38-45.

[38] K. Edalati, S. Toh, M. Arita, M. Watanabe, Z. Horita,High-pressure torsion of pure cobalt: hcp-fcc phase transformations and twinning during severe plastic deformation, Applied Physics Letters, 102 (2013) 181902.

[39] R.K. Islamgaliev, R. Kuzel, E.D. Obraztsova, J. Burianek, F. Chmelik, R.Z. Valiev,TEM, XRD and Raman scattering of germanium processed by severe deformation, Materials Science and Engineering: A, 249 (1998) 152-157.

[40] K. Edalati, Z. Horita, S. Yagi, E. Matsubara,Allotropic phase transformation of pure zirconium by high-pressure torsion, Materials Science and Engineering: A, 523 (2009) 277-281.

[41] H. Matsunaga, Z. Horita,Softening and Microstructural Coarsening without Twin Formation in FCC Metals with Low Stacking Fault Energy after Processing by High-Pressure Torsion, Materials Transactions, 50 (2009) 1633-1637.

[42] K. Edalati, Z. Horita, Y. Mine,High-pressure torsion of hafnium, Materials Science and Engineering: A, 527 (2010) 2136-2141.

[43] Q. Wei, H.T. Zhang, B.E. Schuster, K.T. Ramesh, R.Z. Valiev, L.J. Kecskes, R.J. Dowding, L. Magness, K. Cho,Microstructure and mechanical properties of super-strong nanocrystalline tungsten processed by high-pressure torsion, Acta Materialia, 54 (2006) 4079-4089.

[44] M. Mito, H. Matsui, K. Tsuruta, T. Yamaguchi, K. Nakamura, H. Deguchi, N. Shirakawa, H. Adachi, T. Yamasaki, H. Iwaoka, Y. Ikoma, Z. Horita,Large enhancement of superconducting transition temperature in single-element superconducting rhenium by shear strain, Scientific Reports, 6 (2016) 36337.

[45] K. Edalati, D.J. Lee, T. Nagaoka, M. Arita, H.S. Kim, Z. Horita, R. Pippan,Real Hydrostatic Pressure in High-Pressure Torsion Measured by Bismuth Phase Transformations and FEM Simulations, Materials Transactions, 57 (2016) 533-538.

[46] P. Keblinski, D. Wolf, S.R. Phillpot, H. Gleiter,Continuous thermodynamic-equilibriumglass transition in high-energy grain boundaries?, Philosophical Magazine Letters, 76 (1997) 143-152.

[47] C. Herzig, S. Divinski, Y. Mishin,Bulk and interface boundary diffusion in group IV hexagonal close-packed metals and alloys, Metallurgical and Materials Transactions A, 33 (2002) 765-775.

[48] A. Almazouzi, E.G. Moya, J. Bernardini,Bulk and Grain Boundary Diffusion of Ge and Sn in Intrinsic Germanium, Defect and Diffusion Forum, 143-147 (1997) 1047-1052.

[49] A.M. Brown, M.F. Ashby,Correlations for diffusion constants, Acta Metallurgica, 28 (1980) 1085-1101.

[50] K. Edalati, T. Fujioka, Z. Horita,Evolution of Mechanical Properties and Microstructures with Equivalent Strain in Pure Fe Processed by High Pressure Torsion, Materials Transactions, 50 (2009) 44-50.

[51] W. Gust, S. Mayer, A. Bögel, B. Predel,Generalized representation of grain boundary self-diffusion data, J. Phys. Colloques, 46 (1985) C4-537-C534-544.

[52] A.J. Bradley, J. Thewlis,The crystal structure of alpha-manganese, Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character, 115 (1927) 456-471.

[53] D. Tabor,A Simple Theory of Static and Dynamic Hardness, Proceedings of the Royal Society of London Series a-Mathematical and Physical Sciences, 192 (1948) 247-274.

[54] J. Weertman,Steady‐State Creep through Dislocation Climb, Journal of Applied Physics, 28 (1957) 362-364.

[55] R.L. Coble,A model for boundary diffusion controlled creep in polycrystalline materials, Journal of Applied Physics, 34 (1963) 1679-1682.

[56] J.E. Bird, A.K. Mukherjee, J.E. Dorn, Quantitative Relation Between Properties and Microstructure, Israel Universities Press, Jerusalem, Israel, 1969.

[57] W.R. Cannon, T.G. Langdon,Creep of ceramics, Journal of Materials Science, 23 (1988) 1-20.

[58] A. Mohammadi, N.A. Enikeev, M.Y. Murashkin, M. Arita, K. Edalati,Examination of inverse Hall-Petch relation in nanostructured aluminum alloys by ultra-severe plastic deformation, Journal of Materials Science & Technology, 91 (2021) 78-89.

[59] Y. Ito, K. Edalati, Z. Horita,High-pressure torsion of aluminum with ultrahigh purity (99.9999%) and occurrence of inverse Hall-Petch relationship, Materials Science and Engineering: A, 679 (2017) 428-434.

[60] R. Zheng, J.-P. Du, S. Gao, H. Somekawa, S. Ogata, N. Tsuji,Transition of dominant deformation mode in bulk polycrystalline pure Mg by ultra-grain refinement down to sub-micrometer, Acta Materialia, 198 (2020) 35-46.

[61] M.M. Castro, P.H.R. Pereira, A. Isaac, T.G. Langdon, R.B. Figueiredo,Inverse Hall–Petch behaviour in an AZ91 alloy and in an AZ91–Al2O3 composite consolidated by high-pressure torsion, Advanced Engineering Materials, n/a (2019) 1900894.

[62] A.P. Carvalho, R.B. Figueiredo,The Effect of Ultragrain Refinement on the Strength and Strain Rate Sensitivity of a ZK60 Magnesium Alloy, Advanced Engineering Materials, n/a 2100846.

[63] H. Naziri, R. Pearce,The effect of grain size on workhardening and superplasticity in Zn/0.4% A1 alloy, Scripta Metallurgica, 3 (1969) 811-814.