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The Entropy of an Armco Iron under Irreversible Deformation

Anastasiia Kostina * and Oleg Plekhov

Institute of Continuous Media Mechanics UB RAS, Ak. Koroleva str.1, 614013, Perm, Russia;

E-Mail: poa@icmm.ru

* Author to whom correspondence should be addressed; E-Mail: kostina@icmm.ru;
Tel.: +7-3422-2378312; Fax: +7-3422-378487.

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Abstract: This work is devoted to the development of a theoretical approach for the calculation of entropy in metals under plastic deformation. The thermodynamic analysis of the plastic deformation process allowed us to obtain the expression for determination of the entropy production. The value of the entropy production in an Armco iron specimen under plastic deformation as calculated the basis of the proposed technique and infrared thermography data. This method also lets us define the inelastic strain caused by the initiation and growth of the defects which was used as the internal variable in the considered thermomechanical model from the experimental data. In order to verify the obtained results a theoretical analysis of the modeled situation was carried out.

Keywords: irreversible deformation; entropy of metals; infrared thermography

1. Introduction

The kinetics of the microstructure evolution of metallic materials have been the subject of experimental and theoretical studies for a long time. The available data indicate that the deformation of metals, especially plastic flow, is characterized by high dislocation activity and the emergence of specific mesodefect patterns. The evolution of these mesodefect structures, accompanied by rotation of mesovolumes of the material and material failure, leads to the generation of high internal stresses and, as a consequence, to the storage of energy in the metal. The problem of the stored energy is linked closely with the problem of the entropy of a plastic deformation. The calculation or measurement of the stored

energy of a plastic deformation during a mechanical test can provide useful information about the current state of the material structure and also help to develop new effective models and criteria for the irreversible deformation and failure of metals. The calculation of the entropy of plastically deformed metals can provide us more detailed information about the structure evolution and allows one to formulate a comprehensive thermodynamic model of the process of plastic deformation.

The problem of the measurement of the stored energy W_{st} of the plastic work $W_p = \sigma : (\varepsilon - \varepsilon^e)$ is widely covered in the literature [1–13]. One of the most detailed descriptions of this problem is given in [6]. An analysis of modern experimental data allows us to conclude that a generally accepted assumption, $W_{st} < 0.2 W_p$, often justified by citing the early work of Taylor *et al.* [7], works well only if calculations are carried out at a relative high strain and high strain rates [8,9]. For slow strain rates the energy balance in metals under plastic deformation is a complicated process [1,5,6] and the stored energy value is a function of the material state and a loading history. To describe the structural induced energy dissipation and storage processes in metals under plastic deformation we need both a new advanced experimental program and a new adequate theoretical model.

The structural investigations based on electron and optical microscopy, x-ray and neutron tomography and other techniques allow classification of the defective structures in metals and the study of their evolution in metals with different crystallographic structure. These studies provide an opportunity to estimate the values of the stored energy and entropy connected with the defect structure of materials [14–16].

Calorimetric and infrared (IR) thermography techniques give us another macroscopic way to study the structure evolution in metals under plastic deformation. Currently one of the most popular techniques for studying the energy balance in metals under plastic deformation is infrared thermography [1,10–13,17,18].

In published research works [12,18] a plastic strain was introduced as an internal thermodynamic variable describing the material structure evolution. However, from thermodynamic point of view, the plastic strain cannot be considered as an independent variable for describing the state of a thermodynamic system. One of the possible solutions to this problem was proposed in [19]. The main point of the proposal is an original model for the description of the damage kinetics based on a statistical description of the mesodefekt ensembles. Ensembles of mesodefects of different structural levels including microcracks and microshears are described by a second order tensor \mathbf{p} which can be considered as an additional strain caused by the generation and growth of mesodefects. This parameter can be considered as independent thermodynamic variable describing the structural changes in the material. Thus, the inelastic deformation can be divided into two parts: “pure” plastic (dissipative) deformation due to the dislocation movement and “structural” deformation due to the initiation and growth of the defects (non-dissipative). The evolution law for \mathbf{p} allows us to describe the energy balance in the material. More detailed information can be found in [20].

The aforementioned model was applied to the development of the theoretical approach for the entropy calculation in metals. Determination of this state function of the thermodynamic system permits to obtain other thermodynamic potentials and use them in theoretical models of the deformation behavior of metals. A number of studies are devoted to the entropy analysis in the frame of tribo-thermodynamics [21–24]. Klamecki [21] showed the existence of two different stationary entropy production rate states, connected

one of them with the structural changes due to defect initiation and the other one with the heat generation due to the defect annihilation. Bryant *et al.* [24] related entropy generation with the material degradation due to wear. Song *et al.* [23] proposed an approach to the calculation of the entropy generation due to plastic deformation during fretting. Beghi [25] proposed a way of the entropy production estimation in the case of elastic and at the beginning of the plastic regimes. He showed that the yield point is related to the transition of the entropy production rate from a low level (near to zero) to a higher level. Anand *et al.* [26] developed a thermo-mechanically coupled gradient theory of rate-independent single-crystal plasticity and obtained the expressions for the determination of the internal energy and entropy of cold work hardening.

In this work we propose a technique for estimation of the entropy production of metal sample under plastic deformation based on the original experimental data and results of the numerical simulation of the quasi-static tension test of an Armco iron specimen. These results allow us to calculate both the entropy rate during plastic deformation of the iron and the evolution of the earlier introduced internal thermodynamic variable (“structural” deformation) and increase the practical significance of the model proposed in [19].

2. The Entropy of the Defect Structure in Metals under Plastic Deformation

Following [27], thermomechanical processes can be described by a displacement vector field $\mathbf{u}(\mathbf{x}, t)$ and an absolute temperature field $T(\mathbf{x}, t)$, here \mathbf{x} is the position vector of a particle in a fixed reference configuration, and t is time. Thermodynamic processes are to obey the momentum balance equation and the first and second laws of thermodynamics. In the case of small deformations, these equations involve the following thermodynamic quantities: volumetric mass ρ , specific internal energy e , strain and stress tensors $\boldsymbol{\varepsilon}$ and $\boldsymbol{\sigma}$, heat supply r , heat flux vector \mathbf{q} , specific Helmholtz free energy F , and specific entropy S .

In particular, $\boldsymbol{\varepsilon} = \frac{1}{2}(\nabla \mathbf{u} + \nabla \mathbf{u}^T)$, $\boldsymbol{\sigma}$ is the Cauchy stress tensor and \mathbf{p} is the structural sensitivity parameter. For the sake of simplicity we will consider the corresponding scalar process.

We assume that the specific heat $c = -T(\partial^2 F / \partial T^2)$ depends on the variables p , ε^e , T . Bever *et al.* [6] discussed the possibility that structural defects associated with a cold work may alter the specific heat. Measurements of the specific heat of the annealed and heavily cold work samples have shown that differences in specific heat were never more than one percent. In our theory the cold work state of the material is related with the structural deformation. Following [12,26,28], we suppose that the specific heat of a process c depends on an elastic strain ε^e and temperature T and does not depend on the structural sensitive parameter p : $c = c(\varepsilon^e, T)$.

Then, the following relation holds: $\frac{\partial}{\partial p} \left(\frac{\partial^2 F}{\partial T^2} \right) = 0$ and the partial derivative $\frac{\partial F}{\partial p}$ can be represented as a linear function of T :

$$\frac{\partial F}{\partial p} = Tf(\varepsilon^e, p) + g(\varepsilon^e, p). \quad (1)$$

In addition, let us assume that the stress response function σ is also independent of the structural parameter p , then:

$$\frac{\partial \sigma}{\partial p} = \frac{\partial}{\partial \varepsilon^e} \left(\frac{\partial F}{\partial p} \right) = 0. \quad (2)$$

Taking into account (1), (2) can be written as:

$$\frac{\partial}{\partial \varepsilon^e} \frac{\partial F}{\partial p} = T \frac{\partial f(\varepsilon^e, p)}{\partial \varepsilon^e} + \frac{\partial g(\varepsilon^e, p)}{\partial \varepsilon^e} = 0. \quad (3)$$

Since expression (3) holds for the arbitrary T , we can write:

$$\frac{\partial f}{\partial \varepsilon^e} = \frac{\partial g}{\partial \varepsilon^e} = 0.$$

so (1) leads to the expression:

$$\frac{\partial F}{\partial p} = T f(p) + g(p), \quad (4)$$

where $\partial F / \partial p$ is a linear function of T and does not depend on the elastic strain.

Then we introduce functions $s_1(p)$, $\bar{E}_1(p)$, such that $s_1'(p) = -f(p)$, $\bar{E}_1'(p) = g(p)$ (here and after symbol “'” denotes the derivative).

Integration of (4) lets us obtain expressions for the free energy F , entropy S and stress response function σ :

$$F(\varepsilon^e, p, T) = -T s_1(p) + \bar{E}_1(p) + \Phi(\varepsilon^e, T), \quad (5)$$

$$S(\varepsilon^e, p, T) = -\frac{\partial F}{\partial T} = s_1(p) - \frac{\partial \Phi(\varepsilon^e, T)}{\partial T}, \quad (6)$$

$$\sigma(\varepsilon^e, T) = \frac{\partial F(\varepsilon^e, T)}{\partial \varepsilon^e} = \frac{\partial \Phi(\varepsilon^e, T)}{\partial \varepsilon^e}, \quad (7)$$

where function $s_1(p)$ is a structural entropy.

Furthermore, we follow common practice [12] and assume that the specific heat does not depend on the elastic strain at small elastic strain, hence $c = c(T)$, then from (5) we can obtain:

$$\frac{\partial}{\partial \varepsilon^e} \left(\frac{\partial^2 F}{\partial T^2} \right) = \frac{\partial}{\partial \varepsilon^e} \left(\frac{\partial^2 \Phi}{\partial T^2} \right) = 0.$$

Integration of the last equation permits us to obtain for the function $\Phi(\varepsilon^e, T)$ the following decomposition:

$$\Phi(\varepsilon^e, T) = \tilde{F}(T) - T s_2(\varepsilon^e) + \bar{E}_2(\varepsilon^e).$$

This leads (5), (6) and (7) to the expressions:

$$F(\varepsilon^e, p, T) = \tilde{F}(T) - T (s_1(p) + s_2(\varepsilon^e)) + \bar{E}_1(p) + \bar{E}_2(\varepsilon^e), \quad (8)$$

$$S(\varepsilon^e, p, T) = s_1(p) - \tilde{F}'(T) + s_2(\varepsilon^e), \quad (9)$$

$$\sigma(\varepsilon^e, T) = -Ts_2'(\varepsilon^e) + \bar{E}_2'(\varepsilon^e) = \bar{E}_2'(\varepsilon^e) + TM(\varepsilon^e), \quad M(\varepsilon^e) = -s_2'(\varepsilon^e), \quad (10)$$

where coefficient $M(\varepsilon^e) = -s_2'(\varepsilon^e)$ depends on the elastic strain only. Let us suppose that $\bar{E}_2''(\varepsilon^e) = E = \text{const}$ (Young's modulus) and $-s_2'(\varepsilon^e) = M = \text{const}$ (stress-temperature coefficient) and introduce the thermal expansion coefficient as $\alpha = -\frac{M}{E}$.

If we suggest that the specific heat [12,26] is a constant value at room temperature $c(T) = c = \text{const}$, then we can obtain an expression for the function $\tilde{F}(T)$ in the form $\tilde{F}(T) = -c \left(T \ln \frac{T}{T_0} - (T - T_0) \right)$. If we also suppose that in the reference configuration $\sigma = 0$ and $\varepsilon^e = 0$ at $T = T_0$, we can convert (8)–(10) to the relations:

$$F = c \left((T - T_0) - T \ln \frac{T}{T_0} \right) + \bar{E}_1(p) - Ts_1(p) + E(\varepsilon^e)^2 / 2 - (T - T_0)\alpha\varepsilon^e, \quad (11)$$

$$S = s_1(p) + c \ln \frac{T}{T_0} + \alpha E \varepsilon^e, \quad (12)$$

$$\sigma = E(\varepsilon^e - \alpha(T - T_0)). \quad (13)$$

The energy balance equation has the following form:

$$\dot{e} = \sigma \dot{\varepsilon} - q_x + r, \quad (14)$$

where $q_x = \partial q / \partial x$, upper dots mean time derivative. The free energy of the system can be defined as:

$$F = e - TS, \quad (15)$$

Substitution of (15) into (14) gives the first thermodynamic law in the form:

$$T\dot{S} = \sigma \dot{\varepsilon} - \dot{F} - S\dot{T} - q_x + r, \quad (16)$$

Time derivatives of Functions (11) and (12) can be defined as follows:

$$\dot{F} = \sigma \dot{\varepsilon} - \dot{W}^p - c\dot{T} \ln \frac{T}{T_0} + \dot{\bar{E}}_1(p) - \dot{T}s_1(p) - T\dot{s}_1(p) - E\dot{T}\alpha\varepsilon^e, \quad (17)$$

$$\dot{S} = \dot{s}_1(p) + c\frac{\dot{T}}{T} + \alpha E \dot{\varepsilon}^e, \quad (18)$$

Substitution of (17) and (18) in (16) gives the expression:

$$c\dot{T} + \alpha TE\dot{\varepsilon}^e = \dot{W}^p - \dot{\bar{E}}_1(p) - q_x + r = \dot{W}^p - \bar{E}_1'(p)\dot{p} - q_x + r. \quad (19)$$

where $\dot{W}^p = \sigma(\dot{p} + \dot{\varepsilon}^p)$ —the plastic work rate. Here, we take into account the assumption that the full strain rate ($\dot{\varepsilon}$) can be represented as the sum of three components: elastic strain rate ($\dot{\varepsilon}^e$), plastic strain rate ($\dot{\varepsilon}^p$) and structural strain rate (\dot{p}).

It was shown that the equation defining the fraction of the plastic work rate converted into heating β ($(1 - \beta)$ function characterizes the energy storage rate) in the plastic deformation process can be expressed in the form [29]:

$$\beta = 1 - \frac{F_p \dot{p} - T F_{Tp} \dot{p}}{\dot{W}^p}. \quad (20)$$

From (11) we can obtain expressions for the partial derivatives of the free energy:

$$F_p = \bar{E}_1'(p) - T s_1'(p)$$

and:

$$F_{Tp} = -s_1'(p).$$

Substitution of these expressions into (18) gives an equation for the β in the form:

$$\beta = 1 - \frac{\bar{E}_1'(p) \dot{p}}{\dot{W}^p}, \quad (21)$$

and from (21) we can obtain $\bar{E}'(p) \dot{p} = (1 - \beta) \dot{W}^p$. Substitution of this expression into (19) leads to:

$$c \frac{\dot{T}}{T} + \alpha E \dot{\varepsilon}^e = \beta \frac{\dot{W}^p}{T} - q_x + r. \quad (22)$$

By the use of (22), the entropy production of the system (18) can be expressed in the form:

$$\dot{S} = \dot{s}_1(p) + \beta \frac{\dot{W}^p}{T} - q_x + r. \quad (22)$$

The entropy associated with a structural parameter is quite small as has been discussed in [6] and can be neglected. Under this assumption $F_{Tp} = 0$ and we have the following equation for the entropy production:

$$\dot{S} = \beta \frac{\dot{W}^p}{T} - q_x + r. \quad (23)$$

or:

$$\dot{S} = c \frac{\dot{T}}{T} + \alpha E \dot{\varepsilon}^e. \quad (24)$$

These expressions were used for the experimental data processing. The entropy calculation allows us to determine the evolution of the structural sensitivity parameter. In [21] it was shown that the structural parameter \dot{p} is connected with the thermodynamic force $\sigma - \frac{\partial F}{\partial p}$ according to the following relation:

$$\dot{p} = \Gamma_p \left(\sigma - \frac{\partial F}{\partial p} \right), \quad (25)$$

where Γ_p —a kinetic coefficient.

Then, we can obtain from (25) a quadratic equation for \dot{p} :

$$\dot{p}^2 - \sigma \dot{p} + \frac{\partial F}{\partial p} \dot{p} \sim 0. \quad (26)$$

Substitution of (20) into (26) leads to the equation:

$$\dot{p}^2 - (\sigma - (1 - \beta)\sigma)\dot{p} + (1 - \beta)\sigma\dot{\epsilon}^p \sim 0.$$

Solution of this equation gives us two expressions for the structural parameter determination:

$$\dot{p}_1 \sim \frac{1}{2} \left[(\sigma - (1 - \beta)\sigma) - \sqrt{(\sigma - (1 - \beta)\sigma)^2 - 4(1 - \beta)\sigma\dot{\epsilon}^p} \right], \quad (27)$$

$$\dot{p}_2 \sim \frac{1}{2} \left[(\sigma - (1 - \beta)\sigma) + \sqrt{(\sigma - (1 - \beta)\sigma)^2 - 4(1 - \beta)\sigma\dot{\epsilon}^p} \right]. \quad (28)$$

To verify the expressions for the entropy production (24) and the structural parameter we will carry out a theoretical analysis of Equations (27) and (28).

3. Calculation of the Entropy Evolution under Plastic Deformation

Figure 1 shows the results (stress-strain curve, stored energy rate and temperature evolution) of the quasi-static tension test of the Armco iron specimen under a strain rate of 10^{-3} s^{-1} . The temperature evolution was recorded by infrared camera (FLIR SC5000, FLIR Systems Inc., Wilsonville, OR, USA). The details of the experiment can be found in [29]. For further consideration we will consider only the homogeneous part of the plastic strain (interval from 50 to 250 in Figure 1) and not take into account the inhomogeneous plastic strain due to the shear bands propagation.

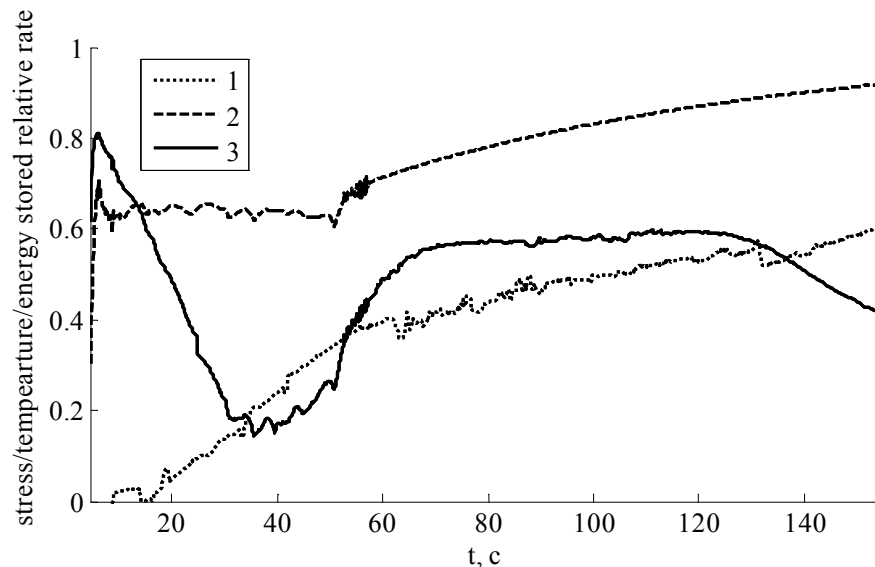


Figure 1. Temperature evolution (1), stress-strain curve (2), stored energy rate (3) for Armco iron specimen. Stress and temperature are normalized to their maximum values and the stored energy rate is normalized to current power applied to the specimen.

The increase of the energy storage rate on the initial stage of the plastic deformation can be connected with the initiation of new structural defects [1]. The energy storage rate decreases when movement and the annihilation of the structural defects become the leading processes of the plastic deformation. This means an increase in the dissipated energy portion. The special feature of the dissipated energy in Armco iron is the presence of the three linear parts on the temperature-time dependence. The second linear part

illustrates the formation of the plateaus on the energy storage rate. This feature was observed for all investigated strain rates.

Figure 2 presents the entropy production calculated with the use of (23) and (24). We can see that these lines are almost equal. Minor divergence can be explained by measurement noise. The stored energy was calculated with the use of the infrared thermography technique which permits us to define the energy storage rate $((1-\beta)$ function) in the deformation process and thus to estimate a portion of the energy expended in the defect structure formation in the material. The details of this technique can be found in [30].

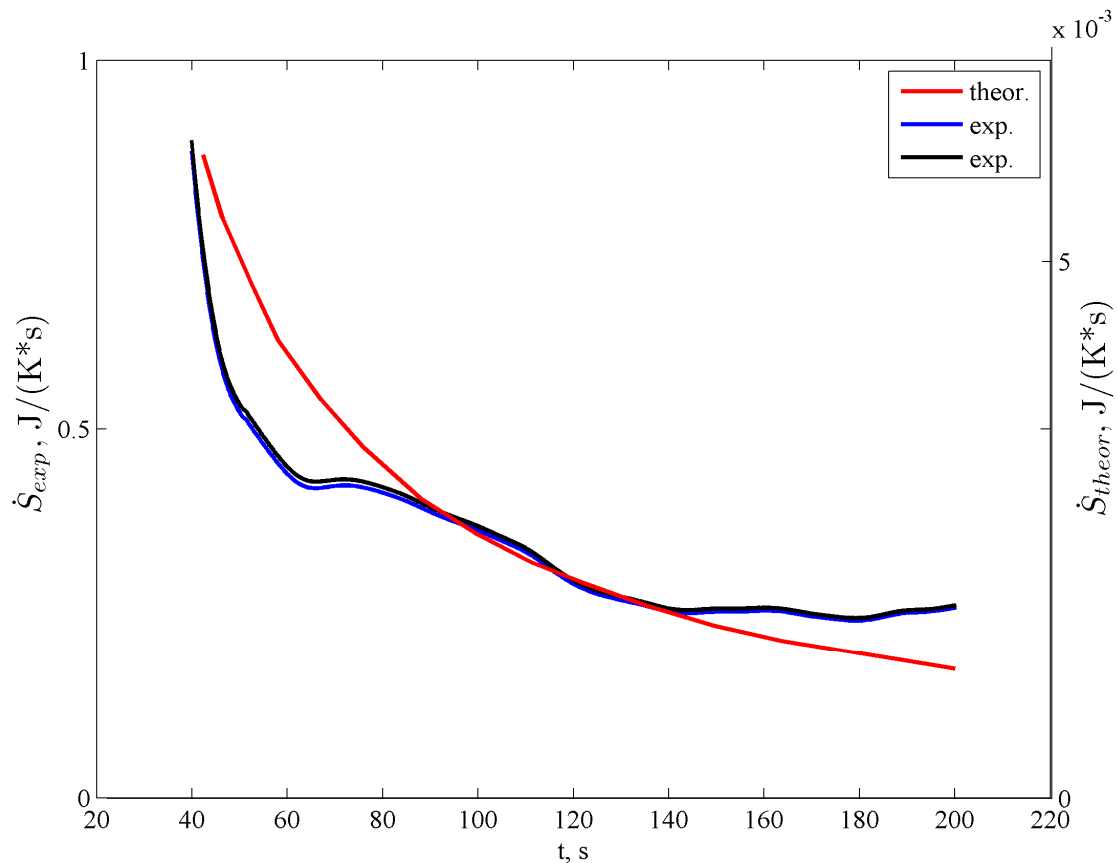


Figure 2. The entropy production (blue line—the entropy production calculated by the use of (23); black line—the entropy production calculated by the use of (24); red line—the entropy production obtained from theoretical analysis).

The evolution of the entropy up to the initial value $\delta S = \int_{t_1}^{t_2} \dot{S} dt$ (the increment of the entropy) is

presented in Figure 3 where entropy increases with time. The obtained results are in agreement with the fact that the entropy of the closed system increases under irreversible processes. This may be interpreted as the more disordered behavior of the defects when the system is closed to the failure.

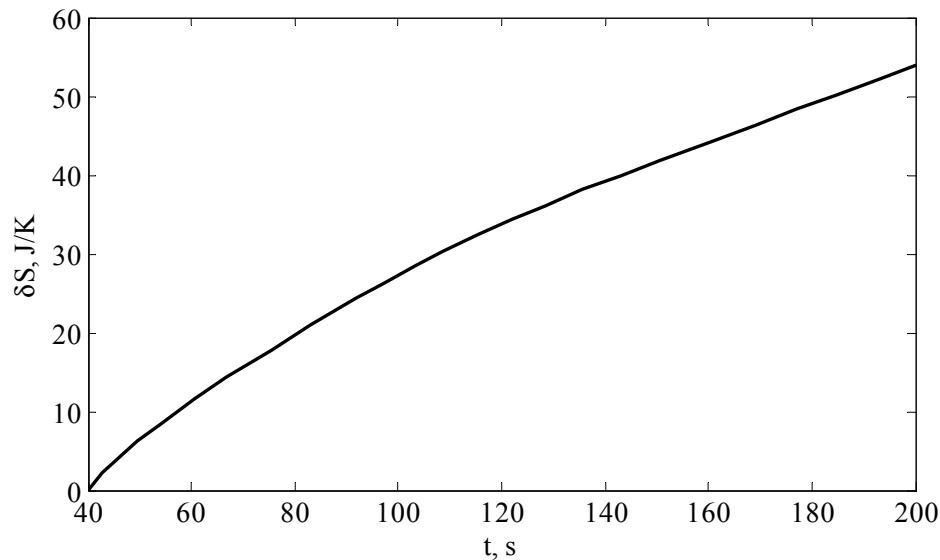


Figure 3. Value of the increment of the entropy *versus* time.

The entropy production curve (Figure 2) and the entropy curve (Figure 3) reflect the transition from plateaus to decreasing branch on the energy storage rate curve (Figure 1). The entropy production reaches a constant value when the energy storage rate decreases (the leading processes are dissipative). The linear segment on the entropy curve occurs in this case.

4. Calculation of the Structural Sensitivity Parameter Evolution under Plastic Deformation

In order to prove the Equations (24), (27) and (28), we will start from the qualitative analysis. The stored energy rate function usually has the increasing branch on the initial stage of a plastic deformation and after it reaches its maximum value the decreasing branch appears. Hence, as a first approximation for the qualitative analysis it can be written as a log-normal distribution function [31]:

$$1 - \beta(\varepsilon) = \frac{1}{\varepsilon k \sqrt{2\pi}} \text{Exp} \left(-(\ln \varepsilon - a)^2 / (2k^2) \right),$$

where $k = 1.9$, $a = 0$.

In this work we consider the entropy production under irreversible deformation and do not take into account the elastic region of the stress-strain curve. For simplicity, we will approximate the strain hardening and the plastic strain dependence upon strain by the linear functions with the appropriate slopes of the lines:

$$\varepsilon^p = \alpha \varepsilon,$$

$$\sigma = \varphi \varepsilon,$$

where $\alpha = 0.05$, $\varphi = 7 \times 10^8$.

The results of the substitution of these functions to (27) and (28) gives us two different solutions: decreasing and increasing. The first solution describes the relative weak defect evolution during the process of plastic deformation. The defect density decreases during the process and dissipation is provided by the movement and annihilation of structural defects. The second solution describes an

unbounded increase of the defect density and cannot be considered as an appropriate root. From the physical point of view the presence of the decreasing branch means that the growth rate of the defects is not constantly increasing quantity.

The value of the \dot{p} obtained from the theoretical analysis is presented in Figure 4 (red line). Results of the substitution of the real experimental data to (27) give similar trend and value for the structural strain rate (blue line). The $\dot{p}(t)$ function obtained from the theoretical analysis allow us to find the theoretical value of the entropy production $\dot{S}(t)$ (Figure 2, red line). The shape of this curve agrees with the experimental curve for the entropy production (Figure 2, blue and black lines). However, we can only use such theoretical approach for the qualitative analysis up to Γ_p .

Figure 4 (black line) shows the $\dot{p}(t)$ function obtained during the finite element simulation of a quasi-static process of the plastic deformation [32]. A quasistatic tension of a smooth Armco iron specimen under strain rate of 10^{-3} s^{-1} was considered. The numerical simulation was carried out in the Simulia Abaqus v6.13 package which was run under academic license. The user subroutine UMAT was used for the modeling. The results of the simulation agree with the results obtained with the use of the aforementioned approach.

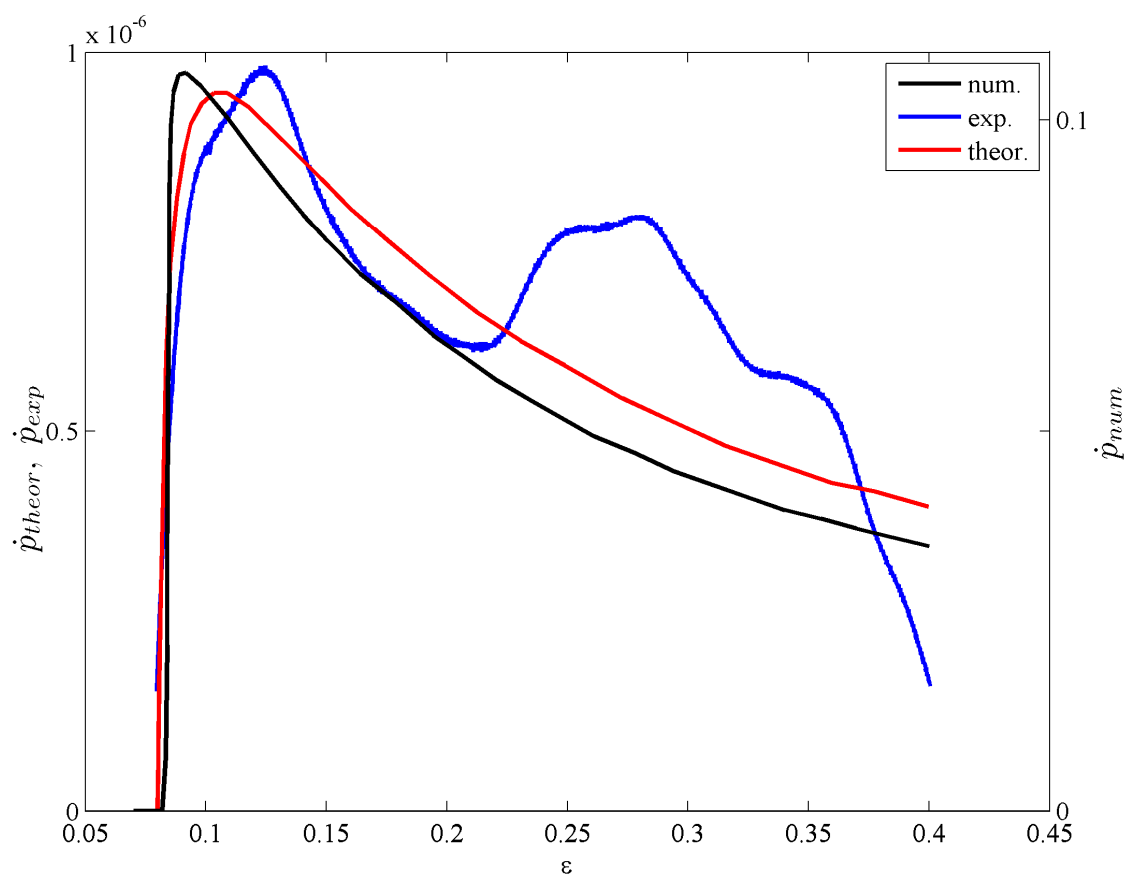


Figure 4. The dependence of \dot{p} upon strain (blue line—experimental results, red line—theoretical results, black line—numerical results).

4. Conclusions

The problem of the entropy of a plastic deformation is one of the most important problems in the description of the irreversible deformation of metals. The solution of this problem allows one to derive the form of thermodynamic potentials of media with defects and to develop a thermodynamic model of plastic deformation. Modern experimental techniques allow us to measure the temperature evolution under mechanical testing and open up interesting possibilities for studying the thermodynamics of plastic deformation. The theoretical analysis of the media with defects carried out in this paper coupled with the previous results of experimental investigation of temperature evolution under quasi-static tension test allow us: (i) to derive the analytical expression for the entropy production of metals under plastic deformation, (ii) to calculate the entropy production and the entropy up to initial value of an Armco iron specimen under quasi-static loading. The obtained curves for the increment of the entropy and the entropy production showed the connection with the energy storage rate curve. In order to verify the obtained results the modeled situation was considered. The comparative analysis of the modeled situation and the results of the experimental investigation of the temperature evolution of an Armco iron under quasi-static tension show a qualitative agreement. Also, the theoretical analysis gives us an opportunity for calculation of the earlier introduced structural strain rate [19] which was used as the internal variable in the considered thermomechanical model

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Author Contributions

The authors contributed equally to the theoretical analyses of the considered process and the paper writing. Both authors have read and approved the final manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

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