

Review

On the Thermodynamics of Friction and Wear—A Review

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Received: 17 March 2010; in revised form: 18 April 2010 / Accepted: 19 April 2010 / Published: 27 April 2010

Abstract: An extensive survey of the papers pertaining to the thermodynamic approach to tribosystems, particularly using the concept of entropy as a natural time base, is presented with a summary of the important contributions of leading researchers.

Keywords: friction; wear; energy dissipation; contact temperature; entropy production; non-equilibrium thermodynamics; self-organization

1. Introduction

Tribology—the science of lubrication, friction, and wear—deals with a diverse array of man-made and natural systems of interacting bodies in relative motion. As the principal cause of wear and energy dissipation, friction is responsible for consuming about one-third of the world's energy resources as humans attempt to overcome its barrier in one form or another [1].

Given that a process involving friction is always accompanied by transformation of energy, it is logical to attempt to develop a thermodynamic framework for studying its characteristics. Naturally, energy dissipation in friction processes represents an irreversible phenomenon, making the concept of the thermodynamic entropy production an ideal tool for probing into its complex behavior.

This paper presents a review of published works and the authors' perspective on the potential of using the thermodynamic approach to friction and wear in modeling sliding contacts in tribosystems. The outline of this paper is as follows: we begin, in Section 2, by reviewing the pertinent works on the energetic approach to friction. Interfacial temperature rise due to friction is covered in Section 3. Next, two generic characteristics of friction, *i.e.*, wear and formation of dissipative structures during friction, employing entropy production as a measure of the degradation are discussed in Sections 4 and 5.

2. Friction and Energy Dissipation

Pressing two bodies together and setting the contact in motion is always accompanied by dissipation of energy. The main factor that controls the behavior of energy generation within the contact of a sliding system is the interfacial friction. The frictional energy generated between contacting bodies is governed by the combination of applied load and velocity. Other contacting factors such as the material properties, relative velocity and size also influence the distribution and dissipation of the frictional energy. The friction and heat dissipation are, therefore, intimately related. In Section 2.1, we begin by describing the nature of energy dissipation due to friction within non-sliding contacts. By non-sliding contact, we mean rubbing between two surfaces with very small amplitude (typically of the order of microns) in oscillatory motion. This phenomenon is generally referred to as fretting wear. In Section 2.2, energy dissipation in contacting bodies in continuous relative motion is considered and its relation to wear is discussed.

2.1. Non-Sliding Contact

Known as fretting damage, repetitive, small-amplitude oscillatory motion between two contacting bodies degrades the contact surfaces due to repeated shear forces. The energy dissipation and associated surface damage in contacting bodies subjected to an oscillating tangential force has been known for decades, starting with the pioneering work of Cattaneo *et al.* [2]. Figure 1 shows a schematic of fretting contact subjected to normal load, P, and oscillatory tangential force, Q, within the contact region of 2a.

Figure 1. A schematic of two bodies in contact under fretting load.



Major advances were later made by Mindlin [3] and his co-authors [4], who are probably the first to theoretically derive an expression for energy dissipation during non-sliding contact. Their analysis of energy dissipation in fretting contact is extended by Mindlin and Deresiewicz [5] to include the effect of oscillation of an oblique force, inclined to the surface of contact. Deresiewicz [6] published a very comprehensive summary of the work of Mindlin. The preceding theoretical studies have been subjected to extensive experimental investigation by Johnson [7,8], Goodman and Bowie [9] and Goodman and Brown [10]. Johnson [8] concludes that the increase in energy dissipation in fretting contact results in increase in surface damage. The surface damage increases rapidly with the angle of obliquity, reaching the maximum when the force in tangential.

Earles [11] theoretically investigates the energy dissipation in nominally flat contacting bodies under fretting and postulates that comparable energy can be dissipated similar to that of visco-elastic materials. Subsequent works include publications by Earles and Philpot [12], Brown [13], Boothroys *et al.* [14], Rogers and Boothroyd [15] and Wilson *et al.* [16], among others.

Investigation on the assessment of energy dissipation during non-sliding contacts is followed by the theoretical work of Hanson *et al.* [17], who model an actual fretting fatigue experimental situation and estimate the energy dissipation under cyclic tangential loading. They estimate the energy dissipation by taking into account the hysteresis loop of tangential force *versus* tangential displacement. Their analysis shows that the energy dissipation drops off rapidly as the distance between the contact region and the boundary increases.

Extensive studies have been reported by Fouvry and collaborators [18–24] who analyzed the fretting phenomenon of contacting surfaces with particular attention to coated surfaces, on the basis of energy dissipation in a fretting contact. They argue [18] that energy dissipation is the primary parameter needed to quantify damage. A so-called global energy wear law is considered in their work, and it is confirmed that the wear volume could be correlated with the cumulative dissipated energy with a linear relationship as:

$$W = c_1 E_d + c_2 \tag{1}$$

where W is the wear volume, c_1 is energy wear coefficient, E_d is energy dissipation and c_2 is the residual volume. Figure 2 shows the wear volume as a function of the cumulated dissipated energy [18]. The results are presented for TiN and HSS (high-speed steels) *versus* alumina contacts, and both present a linear evolution.

Figure 2. Wear volume as a function of cumulative dissipated energy (reproduced from data in Fouvry *et al.* [18]).



They also conclude that the commonly used coefficient of friction (*i.e.*, $\mu = Q/P$) can overestimate the global friction behavior. Therefore, an energy friction coefficient of the following form is introduced to average the friction response:

$$\mu = E_{\rm d} / 4P \delta_0 \tag{2}$$

where δ_0 is the sliding amplitude. The energy approach proposed by Fouvry and co-workers to characterize fretting contact is formulated for non-adhesive wear tribosystems, displaying a weak influence of third body. However, in many applications the presence of third body is a concern.

The use of frictional energy dissipation approach for evaluating the behavior of non-sliding (fretting) triobsystem remains to be of great importance to researchers. Among them, are the works of Zhang *et al.* [25], Bureauy *et al.* [26], Attia *et al.* [27], Magaziner *et al.* [28], Dini and Hills [29], and Korsunsky and Kim [30].

2.2. Sliding Contact

Similar to non-sliding tribological systems, the irreversible energy dissipation due to frictional work in sliding systems has been of great interest to researchers. In the present paper, most significant and relevant works on energetic approach to sliding contact are reviewed.

Friction is an energy transformation process. Using a near-equilibrium analysis, one can demonstrate (see Section 4) how sliding energy is dissipated. Rymuza [31] considers friction as a process that transforms the external mechanical energy to the energy of internal processes. Rymuza proposes that the traditional 'laws' of friction is incapable of reflecting its energetic nature and suggests a new parameter called 'coefficient of friction losses' (CFL), so as to reflect both the dissipative nature of friction process and simultaneously provide a useful formulation for application in engineering practice. The definition of CFL is:

$$CFL = \frac{\text{energy of friction losses (caused by friction)}}{\text{input referenced energy represented by load × sliding speed}}$$

In comparison to traditional coefficient of friction, CFL depends not only on the properties of the contacting bodies, but also on load, sliding speed, environmental conditions, *etc.*,, However, as pointed out by Rymuza, this energetic approach lacks generality, in a sense that the CFL seems to enable easy and adequate description of friction process only in a macro scale. In Rymuza's work, the scale of observation of tribological process is fundamental. That is, the CFL is useful to estimate the energetic 'averaged' losses in the tribosystem being analyzed.

Chen and Li [32] extend the micro-scale dynamic model (MSDM) of wear simulation, originally proposed by Li *et al.* [33], followed by Elalam *et al.* [34], and Chen and Li [35,36]. They investigate the thermal aspects of friction processes with the emphasis on friction heating and its relation to plastic deformation. They perform a quantitative analysis of energy conversion and distribution during sliding. In their work, the first law of thermodynamics is applied to assess the energy distribution. The frictional energy is a part of mechanical energy lost during the sliding process. It includes three parts: strain energy, fracture energy, and thermal energy. They show that during sliding only a portion of friction energy is converted to heat and the remainder results in plastic deformation, micro-cracks, and a change in surface roughness. Figure 3 shows the distribution of generated heat by deformation with

distance from the contact surface (reproduced from the data presented in [32]). It can be seen that the heat is concentrated in the surface layer and that it sharply decreases with an increase in the distance from the contact surface. Similar results are presented by Neder *et al.* [37].





The energy analysis done by Chen and Li [32] is limited to the friction processes in which the change in the surface roughness is not a concern. As they point out, for simplicity of the analysis, the energy consumption corresponding to the change in surface morphology during the friction process is assumed negligible and there is no marked wear involved. This assumption may not be applicable during running-in operating period.

Energy analysis has gained considerable attention for assessment of tribosystems involving wear. Gershman and Bushe [38] consider wear as a generic and fundamental characteristic of friction; energy dissipates into the contacting bodies, giving rise to wear. Uetz and Fohl [39] have considered the energy dissipation by friction and its relationship to wear. Huq and Celis [40–42] propose a procedure to correlate the volumetric wear loss with the dissipated energy for unidirectional and bidirectional ball-on-flat configuration wear test for hard coatings like TiN and (Ti, Al)N, and multilayered (Ti, Al)N/TiN coatings. Their experimental work shows that wear volume is linearly correlated with dissipated energy and that the slope of the linear correlation is useful not only to compare the wear resistance of different materials, but also to compare the wear resistance in different environmental conditions. The effect of relative humidity on the wear of different coatings is investigated. Figure 4 shows an example of the results obtained in the work of Huq and Celis [42]. In this figure, fretting wear volume is plotted against the energy dissipation for multilayer (Ti, Al)N/TiN coating at different relative humidity. The linearity between wear volume and energy dissipation is substantially affected

by humidity of the surroundings. Huq and Celis [42] conclude that the more complexity in wear process arises when one considers the interactions not only between the contacting surfaces but also with the surroundings.

Figure 4. Fretting wear against dissipated energy for 140 nm (Ti, Al)N:140nm TiN multilayer coating at different relative humidity. Operating Conditions: 1N load and 10Hz frequency (reproduced from Huq and Celis, [42]).



A similar study on the energetic approach to wear is reported by Ramalho and Miranda [43]. Interestingly, they show that the energy dissipated in the contact is linearly correlated with the wear volume during sliding wear and that the results can be used to quantify the wear coefficient in Archard's equation. Archard's wear equation relates the wear volume W, to the normal load N, the sliding distance S, and the inverse of hardness H, through a proportionality constant K, often referred to as the wear coefficient (Archard [44]):

$$W = K \frac{SN}{H} \tag{3}$$

Amonton-Coulomb law of sliding friction relates the normal load *N*, to friction force *F*, as $F = \mu N$. Therefore, wear volume and friction force are directly related in accordance to the following relationship:

$$W \propto FS$$
 (4)

where the right hand side of expression (4) physically represents the work of friction force. Therefore, the volume of wear is directly proportional to the energy dissipated by friction. Figure 5 shows several experimental results reported by Ramalho and Miranda [43] for wear volume *versus* dissipated energy for different materials.

Figure 5. Wear volume against *SN* for three different materials (reproduced from Ramalho and Miranda, [43]).



Indeed, experimental results of Ramalho and Miranda [43] reveal that the rate of change in the wear volume is linearly related to the dissipated energy and that the slope of the line represents an estimation of the wear coefficient, K/H = W/SN.

The relationship between wear and dissipated energy has been of interest in the research community. Among them, the most recent and pertinent works are: Savkoor and Ouwerkerk [45], Gupta *et al.* [46], Kuhn and Balan [47], Scherge *et al.* [48], Shakhvorostov *et al.* [49], Larbi *et al.* [50], Li *et al.* [51], Abdel-Aal [52–54], Maeda *et al.* [55], Colaco *et al.* [56], and Nurnberg *et al.* [57]. The foregoing review of literatures on the state of the art pertaining to the energy approach to wear processes reveals that the frictional energy dissipation is a promising feature of tribosystems to characterize wear process. In fact, the linearity between wear and energy dissipation holds for both non-sliding and sliding tribosystems. The beauty of energy approaches to the wear process is that the effect of load, velocity, environmental conditions, *etc.*, on the wear rate can be accounted for by considering frictional energy dissipation within a contact. Further, the amount of energy dissipation can be considered as an indication of the changes in mechanical and metallurgical properties of contacting bodies. This is described in the next section.

3. Frictional Temperature Rise

It is generally accepted that most of frictional work during the wear process is converted into heat, which in turn, raises the interface temperature. The temperature distribution within the contacting bodies, and particularly the near-surface temperature in sliding systems, has been of great interest for many researchers during past decades; See for example, Alyabev *et al.* [58]; Ling [59], Kennedy [60,61], Greenwood and Alliston-Greiner [62], and Knothe and Liebelt [63]. Temperature rise at the

interface can be high enough so as to modify the mechanical and metallurgical properties of sliding surfaces—e.g., as accompanied by formation of oxide layer(s) on the surface or even by melting (Lim and Ashby [64])—and thus drastically changes the behavior of tribological systems (Welsh [65], Blau [66], Quinn [67]). Indeed, the flash temperature in a contact area of a sliding system involving wear of steel, could reach 750–800 °C, which in turn, might induce phase transformation, cause oxidation and reduce the wear resistance [68].

As discussed in Sections 4 and 5, the temperature, and particularly temperature gradient within the mating bodies plays an important role in assessment of entropy generation in a tribosystem. Both theoretical and experimental methods have been developed for determination of temperature rise at the contact surface. Blok [69] is credited to be the first researcher who proposed a model for determination of the temperature rise at the surfaces of contacting bodies under boundary lubricated condition. He considers the temperature rise due to a heat source whose dimension is small with regard to the body of the surface to which it is applied. In the model, two different shapes of heat source, *i.e.*, round and square, at low and high Peclet numbers are considered. In his seminal publications, Blok [69,70] highlights the concept of the flash temperature—the highest temperatures of short duration at the actual contact areas—and gives the formula to estimate its magnitude.

Jaeger [71] follows the concept of flash temperature proposed by Blok [69] to study the surface temperature rise at sliding interface for various shapes of a moving heat source with constant velocity in the surface of a semi-infinite medium. He, also, presents an approximate solution for interface temperature rise at the intermediate Peclet numbers.

Although Czichos [72] speculates that it is nearly impossible to obtain the surface temperature or temperature field during sliding by means of experimental methods, there are some experimental measurements of surface temperature reported in the literature. For example, Wang *et al.* [73] employ a thermal video system to capture a real-time record of the temperature distribution within sliding bodies in a pin-on-ring configuration. Figure 6 shows the measured temperature evolution at about 2 mm

and 10 mm from the sliding distance of the pin specimen (Wang *et al.* [73]). It can be seen that the temperature of the pin specimen rapidly increases with the sliding time during the early period of friction and wear. The temperature rise close to the surface increases to hundreds of degree Celsius in a few seconds during the very early period of sliding.

Wang *et al.* [73], also propose a mathematical model of temperature field in the surface layer during sliding friction and wear as follows:

$$T(x,t) = (1 - x/L)f_1(t) + (x/L)f_2(t) - K(t)\sin(\pi/L)x$$
(5)

which is based on the solution of one-dimensional transient heat conduction equation with K(t) given as:

$$K(t) = t^{1/2} / \pi \Big[A_0 T_0 \exp\left(-A_0 t^{1/2}\right) + A_0' T_0' \exp\left(-A_0' t^{1/2}\right) \Big]$$
(6)

where $f_1(t)$ and $f_2(t)$ are boundary conditions, A_0, A'_0 are constants that depend on the material properties and T_0, T'_0 are the temperature limits in the moving range. They show that the bulk surface temperature corresponding to the transition from mild wear mechanism to the severe wear mechanism is about 200 °C for steel 52100.

Figure 6. Measured temperature profiles of a pin of steel 52100: (a) at about 2 mm from sliding surface; (b) 10 mm from the surface (reproduced from Wang *et al.* [73]).



Additional notable experimental investigations on measurement of surface temperature during frictional contact include the work of Tian *et al.* [74,75] and Szolwinski *et al.* [76]. Tian *et al.* [74] use the thin-film thermocouple (TFTC) technique for polymer pin against Al₂O₃-coated glass in a dry oscillatory sliding system to measure the surface temperature. Szolwinski *et al.* [76] use an infrared thermographic technique to assess the magnitude and distribution of near-surface temperature within a fretting contact between an aluminum alloy cylinder and flat.

An interesting investigation is reported by Tian and Kennedy [77] to analyze the surface temperature rise for a semi-infinite body due to different moving heat sources for the entire range of Peclet number. Using the concept of partitioning heat between two contacting bodies, originally proposed by Blok [70], Tian and Kennedy [77] present the solutions of interface flash temperature for general sliding contact. Let Q represent the total heat flux generation at the interface. Then, the heat transferred to body 1 is ηQ , where η is the heat partitioning factor. The heat flux entering body 2 is $(1 - \eta)Q$. By matching the maximum surface temperature at real contact area, Tian and Kennedy [77] end up with the following expression for partitioning factor:

$$\eta = \frac{1}{1 + \frac{k_2}{k_1} \sqrt{\frac{1 + Pe_2}{1 + Pe_1}}}$$
(7)

where k_1 , k_2 and Pe_1 , Pe_2 are the thermal conductivity and Peclet number of first and second bodies, respectively. The concept of heat partitioning factor is very useful in analysis of a tribosystem. By having the total heat generation at the interface of contacting bodies, one can estimate the amount of heat flux entering bodies.

Recently, the authors of the present paper (Amiri *et al.* [78]) have carried out a series of sliding wear tests pertain to a ring-on-ring configuration for two sets of contacting materials: Bronze SAE 40 on Steel 4140 and 70–30 Brass on Steel 4140. Temperature evolution within contacting bodies during

wear tests are recorded using thermocouple. A thermocouple is placed close to interface (at 2.4 mm from sliding surface), giving reasonable values that can be taken as the representative of the interface temperature. Also, wear of the softer material is measured using a Linear Variable Differential Transformer (LVDT) sensor. The temperature rises during the early period of wear test, then, reaches a steady-state conditions; see Figure 6. The steady-state heat generation due to friction can be expressed as:

$$Q = \mu u N \tag{8}$$

where μ is the coefficient of friction, u is the velocity and N is the normal load. Using the concept of heat partitioning factor, the amount of heat transfers to the first body Q_I is:

$$Q_l = \eta \,\mu u \,N \tag{9}$$

Elimination of normal load N, between Equation (9) and Archard's law in Equation (3), results in:

$$\dot{w} = \left(\frac{K}{\eta \mu HA}\right) Q_I \tag{10}$$

where \dot{w} is the wear rate, $\dot{w} = \dot{W}/A$, and A is the area of contact. The results of the experiments show that there is a linear relationship between temperature rise at interface and the heat transferred to body 1. Figures 7 shows the results of temperature rise plotted against the heat dissipation.

Figure 7. Temperature rise ΔT , against the heat entering body 1, Q_1 (reproduced from Amiri *et al.* [78]).



The linearity between the temperature rise and heat entering body 1 can be expressed as:

$$\Delta T = \Psi Q_1 \tag{11}$$

where Ψ is the slope of the line. It is analytically shown by Özisik [79] that the constant Ψ can be evaluated from the solution of quasi-steady state heat conduction equation. Substitution of Q_I from Equation (11) into Equation (10) gives a relation for the wear coefficient *K* as follows:

Equation (12) offers a methodology for evaluation of the wear coefficient merely by measuring the contact temperature. This method provides a simple and effective technique to quantitatively characterize the wear behavior of a dry sliding system. Figure 8 shows the calculated wear coefficients for Bronze and Brass on Steel for different operating conditions (Amiri *et al.* [78]). The average value of wear coefficient obtained for the Brass on Steel pair is $K_{\text{brass}} = 4.3 \times 10^{-4}$ and for Bronze on Steel pair is $K_{\text{bronze}} = 2.02 \times 10^{-4}$. The published value of wear coefficient for Brass on Steel (Rothbart, [80]) and Bronze on Steel (Rabinowicz, [81]) are:

$$K_{\text{brass}} = 6 \times 10^{-4} \tag{13}$$

$$10^{-3} < K_{\text{bronze}} < 10^{-4}$$
 (14)

Figure 8. Wear coefficients for Brass and Bronze on Steel (reproduced from Amiri et al. [78]).



Comparison of the wear coefficients with the work of Rothbart [80] and Rabinowicz [81] shows very good agreement. It is, hence, concluded that temperature rise at the interface of the contacting bodies is of prominent importance in characterizing the behavior of the tribosystem. This method is particularly useful because of the availability of analytical and experimental results for predicting contact temperature under various operating conditions and configurations. Among them are the works of Kalin and Vizintin [82], Dwivedi [83,84], Chen and Li [32], Mansouri and Khonsari [85], Wen and Khonsari [86], and Bansal and Streator [87].

4. Entropy—Wear Relationship

According to Ramalho and Miranda [43], the friction energy is dissipated mainly through three processes: temperature rise of contacting bodies, generation of wear particles and entropy change due to material transformation in the interface. Relationship between energy dissipation with temperature rise and wear is discussed in previous sections. In this section, we focus our attention to characterization of friction process and wear within a thermodynamic framework. Here, we employ

entropy production as a measure of irreversibility to characterize degradation in a tribosystem, which manifests itself in the form of wear.

Manufacturing transforms raw materials into highly organized components, while aging and degradation—via irreversible processes such as wear, corrosion, *etc.*—tend to return these components back to their natural states (Ling *et al.* [88]). According to Gershman and Bushe [38], a friction process develops its characteristics and parameters with time. Therefore, above and beyond its initial and final stage, the way that this process evolves should be considered. Thermodynamic analysis of the evolution of two characteristics of friction process—*i.e.*, wear and formation of secondary structures—is discussed in this section and Section 5.

Perhaps Klamecki [89–92] is the first researcher who correctly describes the friction process based on the concepts of irreversible thermodynamics. He demonstrates [91] that the process of sliding of two bodies in contact with non-zero relative velocity is a non-equilibrium process. Using a near-equilibrium analysis via entropy production, he shows the occurrence of energy dissipation in sliding processes. Klamecki [89,90] shows that entropy can be defined to describe the state of the bodies in sliding contact and the definition of entropy can be generalized to include all pertinent energy dissipation mechanisms, particularly wear process. He studies the structure that develops near-surface regions of sliding surfaces, and postulates that when energy supplied to the system is not dissipated uniformly through the sliding bodies, the system response will be unstable and definite non-uniform structures are expected to develop [91]. The formation of such structures influences the properties and usefulness of the contacting bodies. Klamecki [92], further, analyzes the plastic deformation energy dissipation based on the model developed by Rigney and Hirth [93] and Heilmann and Rigney [94]. He proposes that the energy input into sliding system by friction, dissipates through two components of the total plastic deformation process. These components are: metallic structural change and heat generation, included in the analysis by means of an expression for entropy. The model proposed by Klamecki introduces a new school of thought for characterizing wear based on the thermodynamic response of the wearing system. However, as it is pointed out by Abdel-Aal [95], in the work of Klamecki, the entropy flow due to mass loss is not explicitly expressed so that a direct correlation between wear and entropy is not apparent.

Zmitrowicz [96–98] develops a complete mathematical framework to formulate a basic system of equations and boundary conditions for two bodies in contact and for third body (cf. Godet, [99]) in the interface. In the first paper (Zmitrowicz, [96]), he presents a model for formulation of the governing equations of mass, momentum, momentum of momentum, energy and entropy inequality for rubbing and wearing solids and third body. Also, a set of dependent variables describing properties of contacting bodies and third body is given from thermodynamics viewpoint. Zmitrowicz [97] also presents the constitutive equations and linearized theories for the contacting bodies and third body. Further, he introduces [98] the constitutive equations for friction, wear and frictional heat within the context of thermodynamical theory. The constants in constitutive equations are governed by two thermodynamic requirements: the second law of thermodynamics, and constraints of the energy dissipation at the frictional contact. Zmitrowicz [100] reviews the modeling of wear of materials, wear patterns and laws of wear.

Dai *et al.* [101], propose a model for analysis of fretting wear based on the irreversible thermodynamic and the concept of entropy balance and the stability of the irreversible processes. The

work of Dai *et al.* [101] is notable in the sense that, it is the first study to quantitatively characterize a tribosystem based on the irreversible thermodynamic concept, particularly for correlating wear and entropy generation during fretting wear test.

Doelling *et al.* [102] experimentally demonstrate that wear is correlated with entropy flow. Experimental wear tests pertain to a slider-rider configuration in boundary lubricated regime. The rider is a stationary copper specimen and the slider partner is a steel cylinder mounted on a rotating shaft. Continuous measurement of entropy flow is made by a calorimeter. Entropy flow is calculated using $S_n = \sum {}^n \Delta Q^{(n)}/T^{(n)}$, where $\Delta Q^{(n)}$ is the heat input to the rider during the *n*th time interval, and $T^{(n)}$ is the average absolute surface temperature of the rider during the *n*th time interval. Figure 9 shows their experimental results, plotted for normalized wear as a function of normalized entropy. Wear and entropy are normalized by the maximum value of the set in each test. Figure 9 demonstrates a strong relationship between normalized wear and normalized entropy flow, that is: Normalized wear = normalized entropy flow.

Doelling *et al.* [102], showed that the slope of Figure 9 is representative of wear coefficient in classical Archard's wear model (*K* in Equation (3)). They postulate that the Archard's wear law is a thermodynamic consequence and is subsumed in their generalized functional relationship between wear and entropy flow. It is discussed later, that at steady-state conditions the flow of entropy equals to the value of entropy production. It is important to note that, Doelling *et al.* [102] formulation is based on the entropy flow rather than entropy production. This important clarification was not made until the work of Bryant *et al.* [103] and Bryant [104].





As a continuation of Doelling *et al.* [102], Ling *et al.* [88] report a new set of experiments to the limiting case wherein there is neither wear nor production of entropy flow. They designate this set of experiments as Category II in comparison to their previous work as Category I. In Category I, the

normal load is so high, that only a small portion of it is carried by the lubricant. In contrast, in Category II, lubricating fluid carries all of normal load and consequently wear is zero. In Category II, the tests are in the mixed and hydrodynamic lubrication regime, as typically identified on the Stribeck curve. In the mixed lubrication regime, the wear and production of entropy flow are moderate. In contrast, in the hydrodynamic regime the both wear and entropy flow are nil.

Brahmeshwarkar [105] experimentally verified the concept proposed by Doelling *et al.* [102] and Ling *et al.* [88], by developing a relationship between the normalized wear and the normalized entropy flow in a totally different test set up, representing a different category of operating conditions. His work involves relating wear to production of entropy flow in a ring-on-ring configuration under dry sliding condition. A theoretical model that simulates thermal response in sliding contact is presented to verify the proposed relation. The model is based on the fact that sliding contact of two bodies would result in plastic deformation in the near surface region, referred to as the 'severely deformed region' (SDR). The plastic deformation results in heat generation in the SDR and subsequently gives rise to the temperature of contacting bodies. The heat generation at the interface is calculated considering plastic deformation in SDR region. The temperature distribution in contacting bodies due to heat generation in SDR region is then calculated. Having determined the heat generation and temperature distribution, he evaluates the entropy flow. The verification of his model, which basically involves comparison of the Archard's wear coefficient calculated using the theoretical model with the published values, reveals a comparable agreement.

Recently, a general theorem—the so called *degradation-entropy generation theorem*—was developed by Bryant *et al.* [103] that relates entropy generation to irreversible degradation, via generalized thermodynamic forces X and degradation forces Y. In an open system capable of exchanging heat and matter with its surroundings (see Figure 10), the change of entropy consists of sum of two parts (Prigogine, [106]):

$$dS = d_e S + d_i S \tag{15}$$

in which d_eS is the entropy exchange with surroundings and d_iS is the entropy produced internally by the system. The second law of thermodynamics states that the entropy production must be non negative, *i.e.*,:

$$\mathbf{d}_i S \ge 0 \tag{16}$$

Steady-state conditions are of great interest in analysis of a tribosystem. It is known that at the steady-state conditions, change of entropy, dS, does not depend on time. Therefore, it follows from Equations (15) and (16) that in stationary state:

$$d_e S + d_i S = 0;$$
 $d_e S = -d_i S < 0$ (17)

An interpretation of Equation (17) is that for an open system operating at steady-state conditions, the flow of entropy has to leave the system, *i.e.*, the equality of entropy generation and entropy flow at steady-state conditions. Further, the convenience of measurement of entropy flow in contrast to analytical prediction of entropy production has led some researchers to evaluate the tribosystem performance using entropy flow. This is the fact behind the assumption made by Doelling *et al.* [102] in correlating wear with entropy flow, instead of entropy production.



However, it is the internal entropy generation of a system that manifests the degradation of a system. To obtain an explicit expression for entropy production d_iS in terms of experimentally measurable quantities, one must invoke the concept of thermodynamic forces and thermodynamic flows (Kondepudi and Prigogine, [107]).

Suppose the system is divided into j = 1, 2, ..., n subsystems with dissipative processes p_j , where each $p_j = p_j(\zeta_j^k)$ depends on a set of time-dependent phenomenological variables $\zeta_j^k = \zeta_j^k(t)$, $k = 1, 2, ..., m_j$. The entropy production can be expressed as the sum of products of the thermodynamic forces and the corresponding thermodynamic flows:

$$\frac{d_i S}{dt} = \sum_j \sum_k \left(\frac{\partial_i S}{\partial p_j} \frac{\partial p_j}{\partial \zeta_j^k} \right) \frac{\partial \zeta_j^k}{\partial t} = \sum_j \sum_k X_j^k J_j^k$$
(18)

where X_j^k are the thermodynamic forces and J_j^k are the conjugate thermodynamic flows. Bryant *et al.* [103] introduce the concept of degradation forces to obtain an expression for degradation $w = w\{p_j(\zeta_j^k)\}$ as follows:

$$\frac{dw}{dt} = \sum_{j} \sum_{k} \left(\frac{\partial w}{\partial p_{j}} \frac{\partial p_{j}}{\partial \zeta_{j}^{k}} \right) \frac{\partial \zeta_{j}^{k}}{\partial t} = \sum_{j} \sum_{k} Y_{j}^{k} J_{j}^{k}$$
(19)

where Y_j^k are the degradation forces. Since Equations (18) and (19) share J_j^k , degradation coefficients can be defined as:

$$B_{j} = \frac{Y_{j}^{k}}{X_{j}^{k}} = \frac{\left(\frac{\partial w}{\partial p_{j}}\right)\left(\frac{\partial p_{j}}{\partial \zeta_{j}^{k}}\right)}{\left(\frac{\partial v}{\partial \gamma_{j}}\right)\left(\frac{\partial p_{j}}{\partial \zeta_{j}^{k}}\right)} = \frac{\partial w}{\partial_{i}S}\Big|_{p_{j}}$$
(20)

which suggests B_j measures how entropy production and degradation interact on the level of dissipative processes p_j . Bryant *et al.* [103] successfully applied their theorem to sliding systems involving wear and fretting. They conclude that the well-known Archard law for assessing wear in friction wear and energy-based models for fretting wear are the consequence of the laws of thermodynamics and the degradation-entropy generation theorem. It is worthwhile to mention that the degradation within an irreversible thermodynamic framework. The theorem expresses the rate of degradation and entropy generation by applying the chain rule and makes no assumption on the thermodynamic state of the system (Bryant [108]). Therefore, it can be applied to the systems

operating far from equilibrium. However, the applicability of the theorem in describing other type of degradation processes, *i.e.*, wear and fretting, are yet to be experimentally investigated.

Dai and Xue [109] review some fundamental problems in modeling friction and wear and development of thermodynamic approaches towards tribology problems. They point out that relating micro-, meso- and macro-scale features of friction can be obtained by introducing entropy as a key parameter to represent energy dissipation and material loss by using the theory of non-equilibrium thermodynamics.

5. Entropy and Self-Organization during Friction

Irreversible thermodynamic aspect of wear as an attribute of friction process is discussed in the preceding section. Here, another characteristic of friction process—the formation of dissipative structures during friction—is discussed. The process of formation of dissipative structures is called self-organization and it occurs under significant deviation from thermodynamic equilibrium state. The formation of dissipative structures during friction process results in reduction of wear rate. In development of wear resistance materials, technologies that help the system to form the dissipative structures faster are very desirable (cf. Gershman and Bushe [110]). Interests in selection of materials for contacting bodies that increase the intensity of non-equilibrium processes during friction are growing.

Self-organization has been widely studies in different disciplines such as mechanics, chemistry and biochemistry since the publication of the pioneering work by Glansdorff and Prigogine [111]. Several types of dissipative structures exist in fluid mechanics, chemical and biological reactions (Glansdorff [112]). A comparatively extensive literature survey is given by Gershman and Bushe [38]. Here, we present pertinent works done on the development of self-organization processes in the field of tribology and surface engineering.

It is well known that for a system at thermodynamic equilibrium state entropy has the maximum possible value whereas entropy production vanishes; that is, $d_iS/dt = 0$ in Equation (18). Therefore, it is inferred that $X_j^k = 0$, $J_j^k = 0$ (Prigogine, [113]). As stated earlier, a tribosystem is an open, non-equilibrium system. Hence, its entropy is lower than the equilibrium state and entropy production is not zero. The lower entropy of the system asserts an increase in orderliness and self-organization. Generally, a system does not reach equilibrium because of the interference of external elements. In a tribosystem, external elements such as load, velocity, temperature, humidity, *etc.*,, push the system to operate far from equilibrium. Therefore, to prevent entropy growth, a highly ordered intermediate body forms on the interface. This is commonly referred to as tribo-film. Formation of tribo-films is the response of the system to external stimulus to reduce wear rate. If tribo-films are not formed during friction, tribosystem stops the friction by seizure or jamming (Gershman and Bushe [38]).

Formation of tribo-films needs dissipation of energy. The required energy is spent for non-equilibrium processes instead of damaging the contact surfaces and producing wear particles. In tribo-films a dispersion of energy occurs. More than 90% of frictional energy is dissipated as heat in the thin layer of tribo-films (Garbar and Skorinin, [114]). This phenomenon has attracted attention of researchers in development of wear resistant coating which can resist heat generation at contact surfaces.

A very recent study on the thermodynamic approach to the self-organization and self-healing of coating during friction is presented by Nosonovsky *et al.* [115]. They postulate that the mechanisms of self-organization involve interactions with different characteristic length scales. For example, Figure 11 shows the self-healing of large-scale cracks and voids which contribute to the decrease of entropy at macroscale, while fracture of microcapsules increases disorder and entropy at the mesoscale. Hence, the decrease in entropy at macroscale is done at the expense of mesoscale. It is to be noted that microcapsules are microscale capsules embedded in the material and contain healing liquid. During a crack propagation process, the microcapsules fracture and release healing liquid to prevent crack propagation [115].

Figure 11. Crack healing by embedded capsules at the micro and meso scales (reproduced from Nosonovsky *et al.* [115]).



Nosonovsky *et al.* [115] propose that the net entropy of the entire system comprises of the entropies associated with structures and processes at different scale levels:

$$S_{\text{net}} = S_{\text{macro}} + S_{\text{meso}} + S_{\text{nano}}$$
(21)

where net, macro, meso and nano indicate the net entropy, macroscale, mesoscale and nonoscales component of entropy, respectively (Nosonovsky and Esche, [116]). Therefore during self-organization, the entropy production at one scale level can be compensated at another level. This means that a decrease in entropy at one level is accompanied by an increase in entropy at another level, so that the net entropy grows in accordance to the second law of thermodynamics [116]. Nosonovsky *et al.* [115] also propose a simple model that provides a criterion for self-healing. Analogous to the degradation parameter (*w* in Equation 19) introduced by Bryant *et al.* [103], Nosonovsky *et al.* [115] define a healing parameter ζ that characterizes the healing or decrease in degradation. A criterion for self-healing is proposed to account for self-healing that occurs when the rate of healing is higher than the rate of degradation. A system of simplified equations is given in their work to assess the rate of self-healing in a system; however, quantitative comparisons for experimental verification await further research.

Nosonovsky and Bhushan [117] study the entropy associated with multiscale features of degradation and self-organization processes in detail. They analyze the thermodynamics of biological

objects with abilities of adoption and self-healing from the view point of entropy production. Very interesting conclusions can be drawn from the analysis of the work of Nosonovsky and Bhushan [117] in which they account variation of coefficient of friction and thermal conductivity during friction analysis. Similar analysis has been carried out before their work by Gershman and Bushe [38]. From the thermodynamic stability analysis, the second-order variation of entropy about the stationary value, $\delta^2 S$, can be expressed as:

$$\frac{1}{2}\frac{\partial}{\partial t}\left(\delta^{2}S\right) = \sum_{i}\delta X_{i}\,\delta J_{i}$$
(22)

where δX_i and δJ_i , are the deviations of X_i and J_i from their stationary values, respectively. Depending on the detailed form of the flows J_i and forces X_i , the state of the system could be stable or unstable. Therefore, the right-hand side of Equation (22) could be negative or positive (Coveney [118]). For a thermodynamically stable system, the right-hand side of Equation (22) should be non-negative. In what follows, it is shown how dependence of coefficient of friction and thermal conductivity on load, N and velocity, u could either retain system in stable condition or drive the system away from equilibrium.

Assume that friction is the only source of energy dissipation in a system. According to Equation (18) the entropy production can be expressed as:

$$\frac{d_i S}{dt} = XJ \tag{23}$$

Considering $X = \nabla(1/T) = -\nabla T/T^2$ and $J = -k\nabla T = \mu Nu$, where μ and k are coefficient of friction and thermal conductivity, a phenomenological coefficient, L, can be defined as:

$$L = \frac{J}{X} = \frac{-k\nabla T}{-\nabla T/T^2} = kT^2$$
(24)

Assume that the coefficient of friction and thermal conductivity change during friction due to the change of a variable, like λ . In present analysis, λ can be sliding velocity, *u*, normal load, *N*, or temperature *T*. Using Equation (24) and assuming that thermodynamic flow, *J*, and force, *X*, depend on the variable λ , Equation (22) can be written as:

$$\delta X \delta J = \left(\frac{\partial X}{\partial \lambda} \delta \lambda\right) \left(\frac{\partial J}{\partial \lambda} \delta \lambda\right) = \left[\frac{1}{L} \left(\frac{\partial J}{\partial \lambda}\right)^2 - \frac{J}{L^2} \left(\frac{\partial J}{\partial \lambda}\right) \left(\frac{\partial L}{\partial \lambda}\right)\right] \left(\delta \lambda\right)^2$$
(25)

First, we consider the case in which the coefficient of friction and thermal conductivity depend on the sliding velocity, u. Equation (25) is, then, expressed as:

$$\delta X \delta J = \frac{N^2}{kT^2} \left(\frac{\partial \mu}{\partial u} u + \mu \right) \left(\frac{\partial \mu}{\partial u} u + \mu - \frac{\mu u}{k} \frac{\partial k}{\partial u} \right) (\delta u)^2$$
(26)

Self-organization occurs if the system loses its stability, *i.e.*, the right-hand side of Equation (26) becomes negative. One possible way is that the sign of the product $\frac{\partial \mu}{\partial u} \frac{\partial k}{\partial u}$ becomes positive. This means, the tribosystem is likely to enter the self-organization regime if the coefficient of friction and thermal conductivity simultaneously increase or decrease with velocity. Similar conclusion can be drawn if the coefficient of friction and thermal conductivity depend on normal load or temperature.

Equation (25) results in Equations (27) and (28) if the effect of load and temperature are considered, respectively:

$$\delta X \delta J = \frac{u^2}{kT^2} \left(\frac{\partial \mu}{\partial N} N + \mu \right) \left(\frac{\partial \mu}{\partial N} N + \mu - \frac{\mu N}{k} \frac{\partial k}{\partial N} \right) (\delta N)^2$$
(27)

$$\delta X \delta J = \left[\frac{N^2 u^2}{kT^2} \left(\frac{\partial \mu}{\partial T}\right)^2 - \frac{\mu N^2 u^2}{k^2 T^4} \left(\frac{\partial k}{\partial T}T^2 + 2kT\right) \left(\frac{\partial \mu}{\partial T}\right)\right] (\delta T)^2$$
(28)

From Equation (27), it can be concluded that the system loses its stability if the sign of the product $\frac{\partial \mu}{\partial N} \frac{\partial k}{\partial N}$ becomes positive. Similarly, Equation (28) implies the system is likely to lose it stability if the sign of the product $\frac{\partial \mu}{\partial T} \frac{\partial k}{\partial T}$ becomes positive. Therefore, if the system loses its stability from equilibrium state, there is a possibility that self-organization occurs and consequently friction and wear decrease. It is to be noted that the process of self-organizations during friction occurs if beside friction, one or more independent processes affect the system. As discussed above, variation of thermal conductivity during friction is an additional excitation to the system which tries to derive system away from equilibrium. Existence of more independent processes during friction enhances the non-equilibrium processes and increases the probability of self-organization.

Another interesting example is given by Greshman and Bushe [110] who theoretically investigate the lubricating effect of electric current on the friction process. This particular analysis, which is based on the non-equilibrium thermodynamics, is verified by experimental results. In the sliding of current collection materials, the entropy production in Equation (18) is comprised of two terms:

$$\frac{d_i S}{dt} = X_1 J_1 + X_2 J_2$$
(29)

The first term accounts for entropy production due to frictional heating, $X_1 = -\nabla T/T^2$ and $J_1 = -k\nabla T = \mu Nu$, and the second term for current collection entropy production, $X_2 = X_e/T$ and $J_2 = J_e$. X_e and J_e are voltage and electrical current, respectively. Therefore, the entropy production can be written as:

$$\frac{d_i S}{dt} = \frac{(\mu N u)^2}{kT^2} + \frac{X_e J_e}{T}$$
(30)

Gershman and Bushe [110] postulate that during stationary conditions, the mathematical condition of minimum coefficient of friction, μ , depending on electrical current, J_e is:

$$\left(\frac{d}{dJ_e}\right)\frac{d_iS}{dt} = \frac{2\mu(Nu)^2}{kT^2}\left(\frac{d\mu}{dJ_e}\right) + \frac{X_e}{T} = 0$$
(31)

Integration of Equation (31) leads to:

$$\mu = \sqrt{\mu_o^2 - \frac{X_e J_e T k}{(N u)^2}}$$
(32)

where μ_o is the coefficient of friction without electrical current. Equation (32) suggests that with application of electrical current, J_e , in frictional contact the coefficient of friction decreases. Also, the higher the voltage, X_e , the lower is the coefficient of friction. It is to be noted that in derivation of Equation (32), the entropy production, $d_i S/dt$, is first differentiated with respect to J_e and set to zero, then the coefficient of friction, μ , is integrated over J_e .

Abdel-Aal [52–54,119,120], has done an extensive investigation on the effect of thermal properties, particularly thermal conductivity, on the wear resistance of the surfaces during friction process. He defines a region on the frictional contacting surface, a so-called mechanically affected zone (MAZ) which transfers heat from high temperature asperity layers to low temperature sub-layers and postulates that wear is significantly influenced by the ability of MAZ to remove frictional heat away from the surface. The efficiency of MAZ depends on the themo-mechanical loading during the friction process. Abdel-Aal shows that the thermal conductivity of MAZ is influenced by the local distribution of strain rate, defined by an effective thermal conductivity K^* , as follows:

$$K^* = K_{\circ} \left(1 + \beta T \right) - \frac{3\dot{\varepsilon}K_b K_{\varepsilon}}{\nabla^2 T}$$
(33)

where K_o is the thermal conductivity at reference temperature (e.g., ambient temperature), β is the temperature coefficient of conductivity, $\dot{\varepsilon}$ is the strain rate, K_b and K_{ε} are bulk modulus and coefficient of thermal expansion, respectively [52–54]. Abdel-Aal concludes that the variation in strain rate can cause anisotropy in thermal conduction, which in turn, may lead to blockage of heat flow in MAZ. The accumulated energy within MAZ may result in formation of protective layer, *i.e.*, dissipative structure. Therefore, Abdel-Aal [54] postulates that formation of dissipative structures is the inherent response of the tribosystem to the external stimulus.

Kozyrev and Sedakova [121] perform a theoretical and experimental analysis within a non-equilibrium thermodynamic framework to derive a dependence of wear rate on the load in stationary state. They demonstrate that the linear increase in wear rate with an increased load may be interrupted as a result of reduction in wear rate. They observe a nonlinear behavior of wear W as a function of pressure p. That is, in a particular range of load, wear can decrease by increasing the pressure. This phenomenon is explained by tribological reactions that results in formation of wear resistant oxide layers. Kozyrev and Sedakova take into account the effect of another independent process which is the diffusion of material into tribo-film, beside the friction process. The diffusion process can be considered as an external element that lead to self-organization and wear reduction. Considering two dissipative processes, friction with forces and flows as $X_1 = -\nabla T/T^2$ and $J_1 = -k\nabla T = \mu Nu$ and diffusion, with $X_2 = -\nabla \varphi/T$ and $J_2 = -\gamma_D \nabla \varphi$, Equation (18) results in the following expression for entropy production:

$$\frac{d_i S}{dt} = \frac{(pu)^2 \,\mu^2 A^2}{kT^2} + \frac{\gamma_D (\nabla \varphi)^2}{T}$$
(34)

where φ is the chemical potential, γ_D is the transport coefficient, *p* and *A* are pressure and nominal area of contact, p = N/A. Kozyrev and Sedakova assume that in non-equilibrium stationary state, the wear of the tribo-film is proportional to γ_D and the product of *pu* is a characteristic of friction. Therefore, during stationary conditions, the analysis is performed for the conditions of minimum γ_D depending on *pu*. Hence, the mathematical condition for that is: Setting (35) to zero and integrating, we obtain:

$$\gamma_D = \gamma_{D0} - \frac{(pu)^2 \,\mu^2 A^2}{kT (\nabla \varphi)^2} \tag{36}$$

where γ_{D0} is the integration constant. Equation (36) indicates that under stationary conditions, as *pu* increases, γ_D decreases. It is to be mentioned that γ_D was assumed to be proportional to wear *W*. Therefore, under the stationary non-equilibrium conditions, Equation (36) offers a procedure for decreasing wear with increase in *pu*. Figure 12 shows the experimental wear results of the work of Kozyrev and Sedakova [121] for two different materials. Experimental wear tests pertain to a ring-plane configuration for two sets of contacting materials: F4K15M5 on 35KhM Steel and Sigma-3 on 35KhM Steel. Figure 12 shows that in some range of *pu*, the wear *W* does not grow with *pu* but even decreases. The length of this zone depends on material properties.

Figure 12. Wear as a function of *pu* for two different materials (reproduced from Kozyrev and Sedakova, [121]).



During the past decade, other researchers have successfully applied the concept of self-organization to characterize the behavior of tribosystem during friction process. Contributions include: Bershadsky [122], Kostetsky [123], Fox-Rabinovich *et al.* [124–126], Wilson and Alpas [127], Usychenko [128], Figueiredo *et al.* [129] and Kovalev *et al.* [130].

Recently, Fox-Rabinovich *et al.* [131] perform a theoretical study based on the irreversible thermodynamic concept to investigate the probability of the occurrence of the self-organization processes under complex conditions. The complex condition is defined as the severe operating condition (e.g., aggressive cutting conditions) along with a number of simultaneously processes that

occur during friction. It is postulated that by increasing the number of interrelated processes during friction, probability of initiation of the self-organization increases. As stated earlier in this section, self-organization can only occur if the system losses its thermodynamic stability. Fox-Rabinovich *et al.* [131] postulate that the probability of losing stability can be calculated as follows:

Probability of losing stability =
$$1 - 1/2n$$
 (37)

where *n* is the number of interrelated processes. Equation (37) suggests that by increasing the number of simultaneous processes (*i.e.*, increasing the complexity of the system), the possibility for self-organization increases (Fox-Rabinovich *et al.* [131]). As a result of self-organization, the wear rate decreases. Experiments are carried out by Fox-Rabinovich *et al.* [131] to verify the theory. Experiments pertain to dry high-speed end milling of tool steel on a complex nano-multilayered coating. The results of wear tests of multilayered coating are then compared to that of monolayered coating and this effect is related to the ability of the multilayered coating to form tribo-films during friction process.

In summary, the present work reviews the major applications of thermodynamics in the field of tribology, with particular interest in the energetic/entropic approach. The temperature rise due to the frictional heating at the interface of contacting bodies is an important parameter in machinery and components where the friction is a concern. Therefore, the subject has captured the attention of numerous scientific papers. However, practical applications of the entropic/thermodynamic approach in the field of tribology have not vet become widespread. We believe that the application of irreversible thermodynamics and specifically entropy generation provides new scientific research opportunities for engineering designs and developments of new materials with improved tribological performance. Further, the in-depth studies of the formation of dissipative structures in tribo-films can result in the development of materials that offer substantially improved resistance to wear. Future developments in surface engineering toward inventing wear resistant materials, should also take advantage of grand knowledge of self-organization within the non-equilibrium thermodynamic framework. It is believed that the concept of stability analysis provides a key, enabling factor for proper characterization of tribo-films. Tribo-films can form only if system losses its stability. That is, by changing the external stimulus-like load and velocity-one can increase the probability of formation tribo-films, thereby decreasing the wear rate. These concepts offer new and exciting research in the field of tribology for years to come.

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