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Energy Dissipation in Tribological Stressed Greases

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Abstract: Lubricating greases that are subject to a continuous friction process are in a non-equilibrium state. In processes far from equilibrium, there is a possibility that dissipative structures will form. In this work, the conjecture is pursued that this is also possible in loaded grease films. On the one hand, the shear process is considered in interaction with structural degradation, and on the other hand, the behavior of energy dissipation mechanisms is investigated. In the two models presented, it is shown that there are conditions under which it is possible to trigger self-organization processes. The next step must be the development of suitable experiments.

Keywords: lubricating grease; energy dissipation; entropy production

Citation: Kuhn, E. Energy Dissipation in Tribological Stressed Greases. *Processes* **2024**, *12*, 17. https://doi.org/10.3390/pr12010017

Academic Editor: Andrew S. Paluch

Received: 7 November 2023
Revised: 13 December 2023
Accepted: 15 December 2023
Published: 20 December 2023

1. Introduction

1.1. General Remarks

If lubricating greases are used, they form a friction body in the tribological contact, in addition to the two gap-limiting solids (in general). With a frictional energy balance of the entire contact, this provides a share and changes the overall situation of the tribological system through its behavior under stress.

The general lubricating grease behavior under frictional load, i.e., in the presence of a shearing process, has been investigated and documented in many studies, e.g., [1–5].

The lubricating grease is singled out and its behavior observed when a friction process is initiated.

The description of lubricating grease behavior under tribological stress by means of the investigation of entropy production and entropy transport has only recently been attempted in various works, for example [6–9].

In recent years, there have been a number of publications dealing with the formation of structures in solid-state friction and solid-state wear [10–13]. Processes are described that are initiated from within the system itself and lead to possible new structural formations. In particular, the formation of layers on the solid friction body surfaces in contact has been described in several papers [14,15].

In ASSENOVA one finds "Far from equilibrium, non-equilibrium due to energy and mass flows to and from the system becomes a source of order: a new space-time organization of the system or new structures (the so-called dissipative structures) can emerge" [16].

The process of the occurrence of instability and a self-organization initiated by it, as studied by Prigogine [17], shows the possibility of checking tribological processes via this criterion.The assumption is made that self-organized processes can also be triggered in tribologically stressed lubricating grease films and lead to a new structural state [18]. In order to counteract the difficulty of experimentally observing the structural formation in tribologically stressed lubricating grease films, indirect indicators for the triggering of self-organized processes are investigated in this work.

1.2. About Structural Degradation

The structural degradation of lubricating greases subjected to frictional loads is, on the one hand, a research topic that deals with the behavior of a very specific lubricant. On the other hand, this research topic can also be used to illustrate very general, fundamental behaviors of tribological systems.

The observation of various properties of lubricating greases during a friction process has led to the term structural degradation. This term has a negative intention that does not do justice to the process. Negative and positive are evaluative descriptions from the user's point of view. However, the perspective of the system "stressed lubricating grease" must be adopted in order to understand the processes taking place and their driving forces.

We can observe a "structural change" that represents something other than structural degradation. And this structural change comes with the question, why is this change happening?

The start of a friction process leads a tribological system, whether it comes from a thermodynamic equilibrium or a stationary state, into instability. The task of the system is to find conditions that eliminate this "disturbance" and prepare a path that leads to a stable state. Energy-dissipating mechanisms are initiated. In the case of solids, these can be material-removing wear processes; and in the case of lubricating grease, structure-changing processes. From the traditional cause-and-effect chain :

$$\text{Friction } (\textit{cause}) \longrightarrow \text{Wear } (\textit{effect})$$

now becomes

$$\text{Friction is the } \textit{cause} \text{ of instability (effect)} \longrightarrow \text{instability is the cause of wear, which then leads to} \longrightarrow \text{stability (stationary state) } (\textit{effect}).$$

In our case, the irreversible energy-dissipating wear process is the structural change and it is appropriate to use the term *grease wear* for this process. This has the same function as solid-state wear and, like the latter, leads to a stable process situation.

2. Materials and Methods

2.1. Grease Structure and Friction Process

Description of the structural development during a friction process in a lubricating grease fill has experimentally been unsuccessful or unknown. The formation of dissipative structures requires certain process conditions, and a tribological test is likely to pass through a limited range of the set test conditions. Presumably, observation is only possible during a shear test.

Comparative study of the structure or structural elements of a lubricating grease before and after tribological stress is simpler. In this context, the lubricating grease structure should be understood as the geometry and distribution of the solid (called thickener).

In our laboratory, we have different experimental possibilities for the investigation of the lubricating grease structure or structural elements. We use light microscopy (LM), scanning electron microscopy (SEM), interferometry (IFM), and, in collaboration with our scientific partner at the University of Huelva (group of Prof. Franco), atomic force microscopy (AFM).

The subject of this work is the possibility of self-organization in a stressed lubricating grease. The structure, which is defined as the geometry and distribution of the solid, is considered. Figures 1–4 are intended to show examples of different geometries and arrangements and also demonstrate the different methods of investigation.

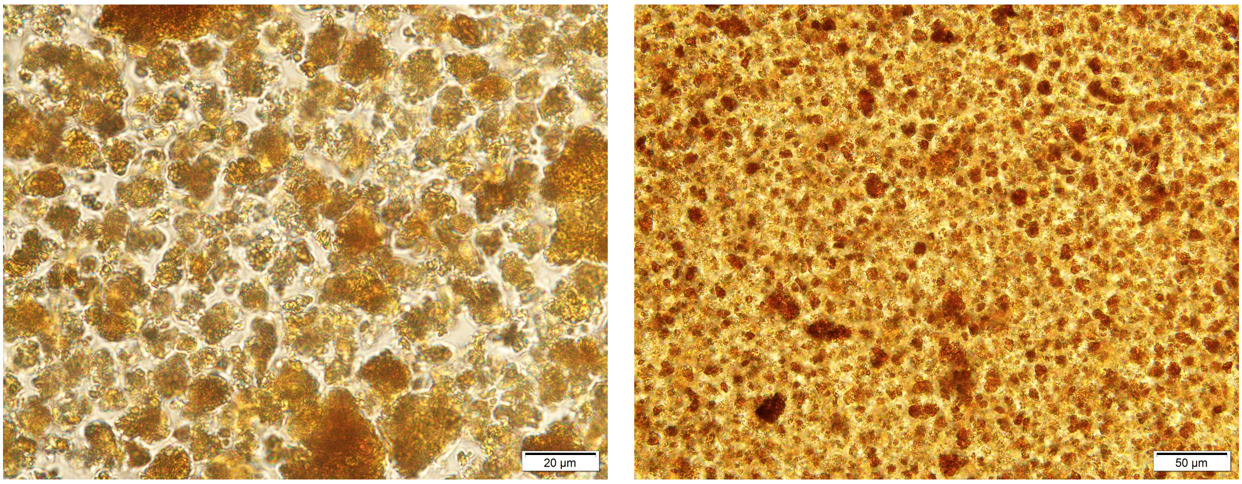

Figure 1. Two samples for transmitted light microscopy with a biogenic sample (solid and base oil).

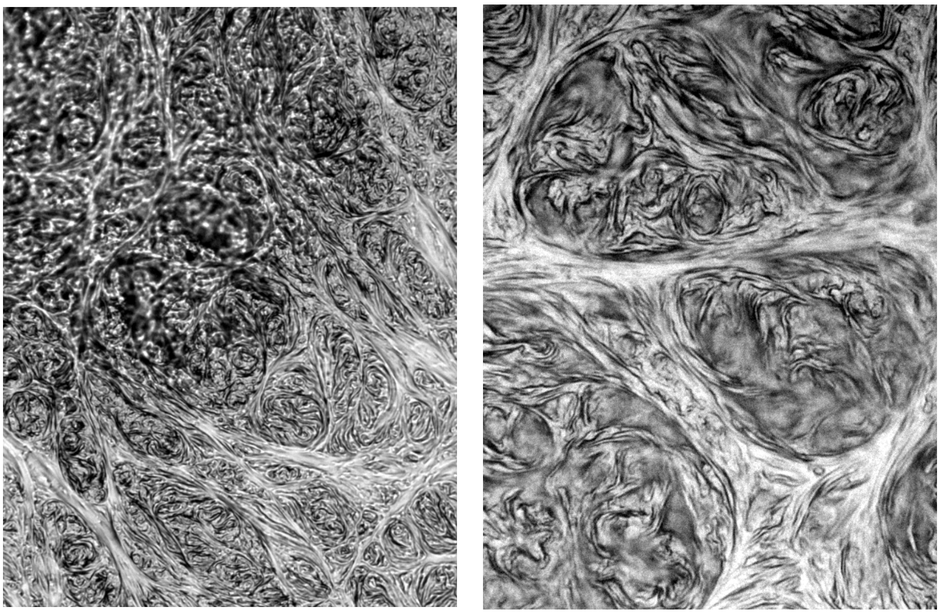

Figure 2. A sample with a Li-soap. Reflected light microscopy—grease structure covered by a thin oil coat.

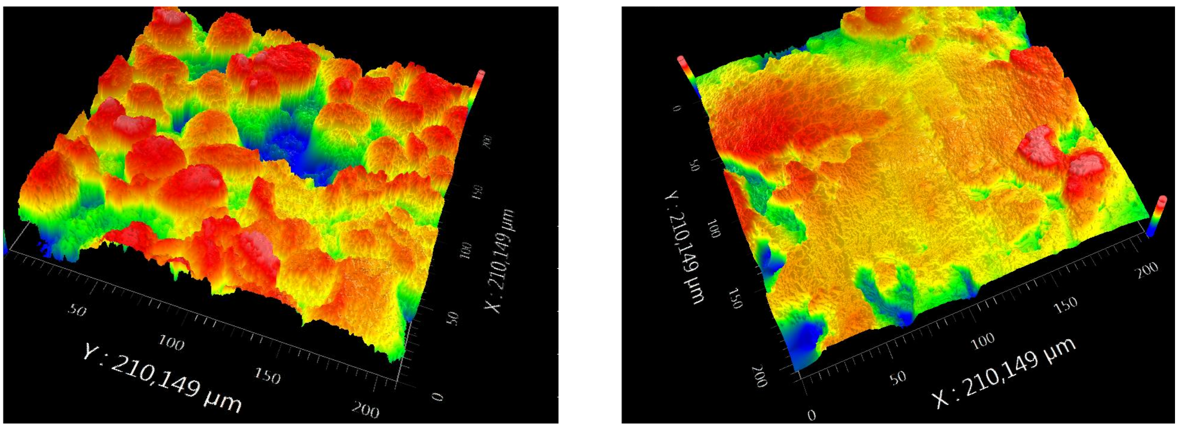

Figure 3. Interferometry of Ca-agglomerate (**left**) and an agglomerate from a polyurea sample (**right**).

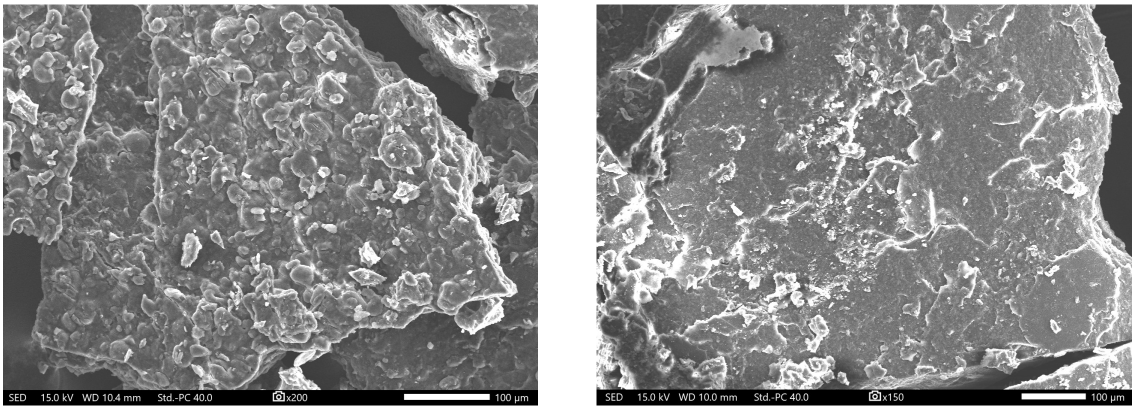

Figure 4. REM investigation of Ca-agglomerate (**left**) and an agglomerate from a polyurea sample (**right**).

A "simpler" method of observing the structural change is to examine the material before and after loading (Figures 5–7). The illustrations here are also exemplary. Of course, before and after does not mean observing any structural formation during the process.

In interferometric and scanning electron microscope examinations of the lubricating grease structure, the base oil and the solid are separated during sample preparation. Due to the resolution (SEM, IFM), we only see a small part of the solid structure. Agglomerates, i.e., larger particles formed e.g., from the Li fibrils, are then the sample material. In contrast, base oil and solid particles are recognizable in light microscopy.

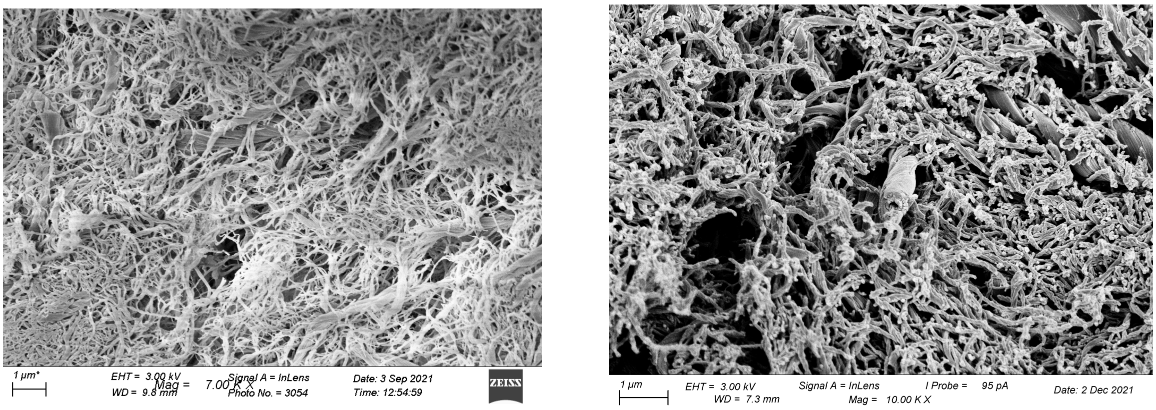

Figure 5. REM-investigation (**left**) Li-sample unstressed, (**right**) Li-sample after stress.

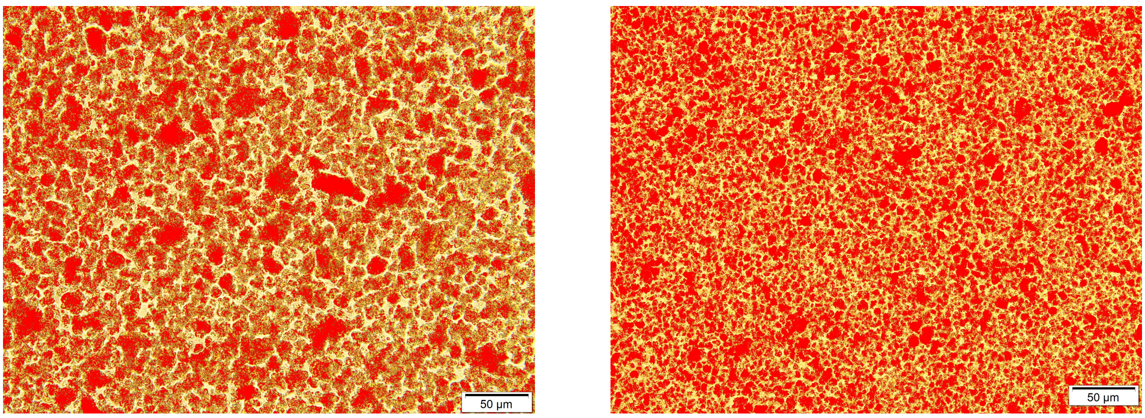

Figure 6. Transmitted light microscopy (**left**) grease sample before stress and (**right**) grease sample after stress.

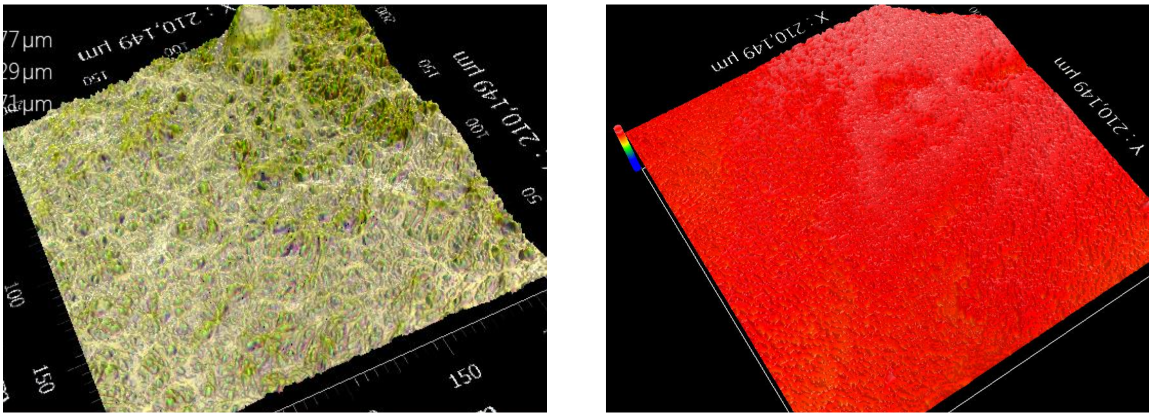

Figure 7. IFM-investigation (**left**) Li-sample unstressed, (**right**) Li-sample after stress.

The differences before and after a tribological stress are recognizable in the agglomerates, as well as in the lubricating grease structure. However, it is not possible to assess the structural state during the stress. This will be quite different from the more static structures after the end of the friction process.

Investigations along a τ vs. t curve, as presented by [8], also give only a limited real impression of the process structure.

2.2. Formation of a New Structure

Friction processes are non-equilibrium processes and are associated with the production of entropy [19]. They strive towards an equilibrium state characterized by maximum system entropy. This equilibrium state can be described using a certain structure in the stressed region of matter. Figure 8 illustrates the concept of the system under investigation.

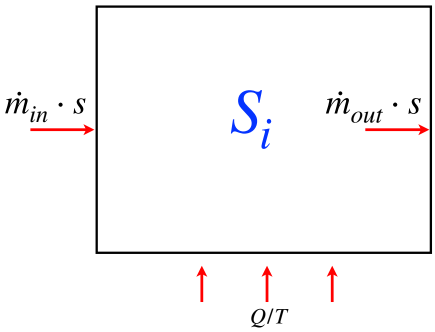

Figure 8. The stressed grease volume element as an open thermodynamic system. With S_i being the entropy production, m the mass that come in and goes out, and s the specific entropy.

If we assume a permanent friction process (steady state), i.e., a continuous supply of energy, this also leads to a permanent dissipation of energy. Quite different dissipation mechanisms can play a role. If the processes of energy supply and dissipation develop in an unbalanced manner due to the system conditions, the system shows itself as *instable*. This means that the change of the excess entropy is negative.

As already mentioned, the system "stressed grease" is hardly observable in the friction process with regard to a possible structural formation. An indication can be given by examining the occurrence of instabilities. Experimental work in the past has also led to the assumption of the possibility of a new structure formation in viscoelastic lubricants.

A test procedure was developed to describe the different structure degradation behaviors comparatively. In a rheometer test, after filling the plate–plate gap and a rest period, two test sections were realized. First, in a loading step, the sample under investigation was sheared (rotational mode) at a constant shear rate $\dot{\gamma} = const$. Immediately after the end of this stress phase, an oscillation measurement was started, i.e., an amplitude sweep was performed, which, after exceeding the linear viscoelastic range, passed into the plastic range of structural degradation and ended in the characteristic crossing point of the storage and loss modulus. An interesting result in the evaluation was the consideration of the energy expended in the oscillation test to reach the crossing point. In general, it is assumed that the structure of a grease completely changes at the crossing point. Thus, if comparatively little energy is expended, a strong structural degradation (wear) takes place during the stressing phase. If, in comparison, a lot of energy is required to reach the crossing point, this means that the structure is still stable after the shear stress.

If the reciprocal of this energy expended in the oscillation test is plotted on the ordinate and the intensity of the stress in the shear test (shear rate) is observed on the abscissa, an increasing curve for a grease sample is to be expected. With increasing stress, less energy is required to reach the crossing point, as the structure of the specimen is changed more and more in the shear test. However, grease samples can show a different behavior, and if one follows the considerations on possible structural formation under certain test conditions, it can be assumed here that new structures are formed (Figure 9).

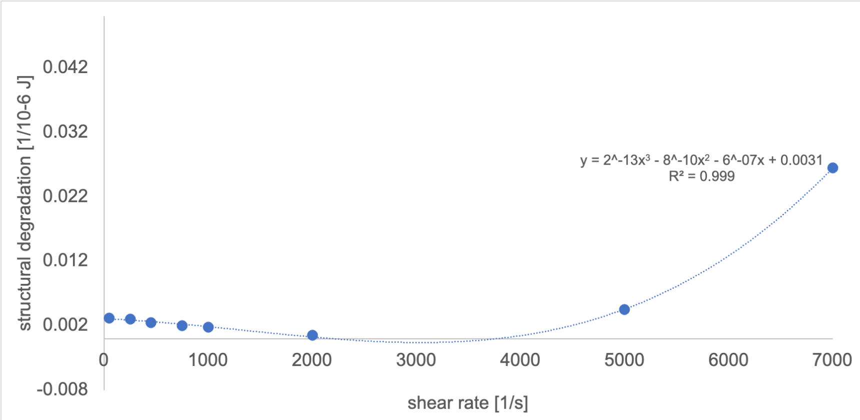

Figure 9. Decreasing structural degradation with increasing load (for smaller shear rates) in an investigated grease sample.

This assumption will now be followed up and indicators will be examined that enable instability and thus make self-organization probable. The analysis in [18] shows for the general case of lubricant friction

$$\frac{1}{2}\frac{\partial}{\partial t}(\delta^2 S) = \delta\left(\frac{\tau \cdot \dot{\gamma} \cdot V}{-\lambda \cdot T^2}\right)\delta(\tau \cdot \dot{\gamma} \cdot V) \tag{1}$$

that with the criterion

$$\frac{\partial \tau}{\partial \epsilon}\frac{\partial \lambda}{\partial \epsilon} < 0 \tag{2}$$

the occurrence of instability is probable. The time deviation of the function $(\delta^2 S)$ was used as a criterion for assessing whether possible instability can occur according to [17]. Stability is given with

$$\frac{1}{2}\frac{\partial}{\partial t}(\delta^2 S) > 0 \tag{3}$$

and the excess entropy production if friction is the only independent source of energy dissipation can be written as

$$\frac{1}{2}\frac{\partial}{\partial t}(\delta^2 S) = \delta X_h \delta J_h \tag{4}$$

Experimental work by Acar et al. [20] on the influence of solid content showed that, as the soap concentration in a Li sample increased, the critical deformation became smaller (Figure 10). This critical deformation γ_{critic} describes the transition from the elastic to the plastic range and can be quantified in an oscillation test with a rheometer (amplitude sweep). This seems explainable against the background that the bonds in the agglomerate are of a physical nature and during shearing the entire grease (base oil + thickener) is stressed.

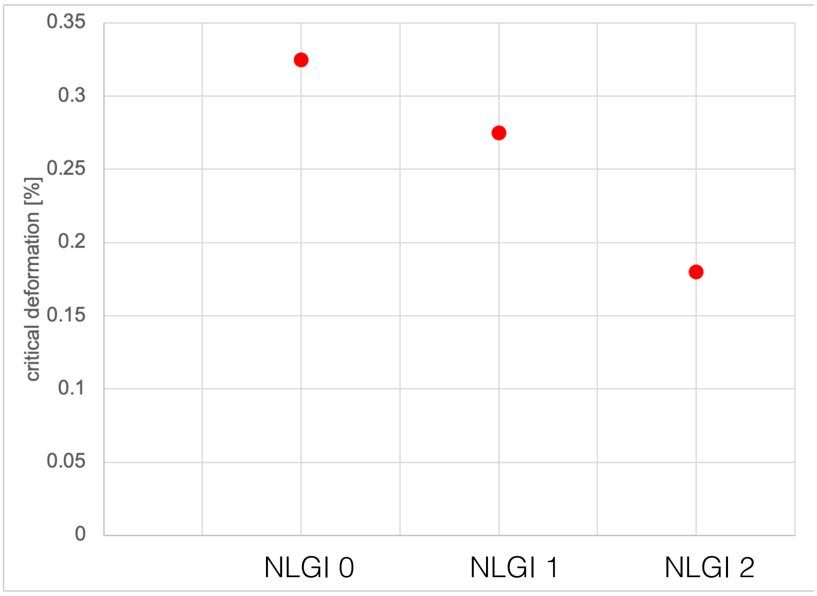

Figure 10. Decreasing critical deformation with increasing NLGI class (this corresponds to an increasing content of solids).

A more detailed investigation in [18] provided conditions for the initiation of instability. Several dissipation mechanisms were considered and, among others, a criterion with

$$\frac{\partial \gamma_{critic}}{\partial \epsilon} < 0 \quad \text{and} \quad \frac{\partial F_f}{\partial \epsilon} > 0 \tag{5}$$

was found.

This means that, with decreasing critical deformations γ_{critic} at increasing ϵ and simultaneously increasing fragmentation rate F_f of the agglomerates, there is a possibility of instability occurring. The parameter ϵ describes the distance to equilibrium. It can be seen that in this interpretation, increasing the solids content tends to lead to instability of the system.

The general shear of the base oil, fragmentation of agglomerates and coagulation of particles after a collision were considered in [18].

From the studies in [21], it can be concluded that with the increase in interconnected processes, the probability of loss of thermodynamic stability increases. As a direct consequence of this finding, the probability of self-organization also increases.

This fact will be further explored in the following section when considering the interaction of dissipation mechanisms.

2.3. Indirect Indications about the Possibilities of Forming New Structures

Apart from the described difficulty of the experimental proof of dissipative structures in a lubricating grease film, there is the possibility of finding indirect indications of a probable self-organized event from model observations.

The above-described circumstance of the favorable influence of a *higher solids content on the triggering of instabilities* is now investigated with other dependencies.

It should be taken into account that an increase in frictional energy due to an increase in deformation also leads to an increase in structural degradation (lubricating grease wear). This in turn results in a reduction in the frictional energy applied.

$$E_f = E_{f0} + E_{f\dot{\gamma}} - E_{fw} \tag{6}$$

with the frictional energy rate due to an increase in deformation

$$E_{f\dot{\gamma}} = (\tau \cdot \dot{\gamma} \cdot V) \quad \text{and} \quad \tau_{ostw} = k \cdot \dot{\gamma}^n \tag{7}$$

and a general assumption

$$E_{fw} = b \cdot W_{ear}^q \tag{8}$$

we use $W_{ear} = E_{f\dot{\gamma}}/E_P$, with the explanation that $E_P = T/B$ with B the degradation coefficient from Bryant et al. [22].

It should be remembered that we are looking for the possibility of the emergence of instability.

The Equation (6) is now

$$E_f = E_{f0} + (\tau_{ostw} \cdot \dot{\gamma} \cdot V) - b\left(\frac{E_f}{E_P}\right)^q \tag{9}$$

Equation (1) is now rewritten with

$$\frac{1}{2}\frac{\partial}{\partial t}(\delta^2 S) = \delta\left[\frac{k\dot{\gamma}^n\dot{\gamma}m}{\rho} - b\left(\frac{k\dot{\gamma}^n\dot{\gamma}m}{\rho E_P}\right)^q\right]\delta\left[-\frac{k\dot{\gamma}^n\dot{\gamma}m}{\rho\lambda T^2} + \frac{b}{\lambda T^2}\left(\frac{k\dot{\gamma}^n\dot{\gamma}m}{\rho E_P}\right)\right] \tag{10}$$

The parameter ϵ is introduced that describes the distance to equilibrium. The dependencies $\rho(\epsilon)$ and $\lambda(\epsilon)$ are examined.

$$\frac{1}{2}\frac{\partial}{\partial t}(\delta^2 S) = -\frac{1}{T2}\left[(\text{factor}_1)\left(\frac{\partial\rho}{\partial\epsilon}\right)^2 + \frac{1}{\lambda^2}(\text{factor}_2)\frac{\partial\rho}{\partial\epsilon}\frac{\partial\lambda}{\partial\epsilon}\right](\delta\epsilon)^2 \tag{11}$$

The factors can be written

factor$_1$

$$\left[-\frac{a}{\rho^2} + \frac{qba^q}{E_P^q}\cdot\frac{1}{\rho^{q+1}}\right]^2 \tag{12}$$

factor$_2$

$$\left[\frac{a^2}{\rho^4} - \frac{ba^{q+1}}{E_P^q\rho^{q+2}} - \frac{qba^{q+1}}{E_P^q\rho^{q+3}} + \frac{qb^2a^{2q}}{E_P^{2q}\rho^{2q+1}}\right] \tag{13}$$

a is summarized with

$$a = k \cdot \dot{\gamma}^{n+1} \cdot m \tag{14}$$

Unfortunately, no experiments could be performed for the quantitative determination of b and q. Thus, an estimation of the factors is not possible. But let us come back to the question of whether there are conditions in the considered interactions which show the influence of a higher solid content on the formation of new structures.

With assumptions for the factors, this shows that a condition exists to make the right side of the Equation (11) negative.

$$\frac{\partial\rho}{\partial\epsilon}\frac{\partial\lambda}{\partial\epsilon} > 0 \tag{15}$$

This shows that there are conditions where increasing the solid fraction (thickening) facilitates the possibility of self-organization. This can be determined in this estimative observation. Future experimental studies are required to make quantitative statements.

Another indication of indirect indicators for the formation of dissipative structures will now be investigated.

This hint came from PRIGOGINE [17] and KLAMECKI [19] and concerns the behavior of energy dissipation rates in a stressed system. A *cyclic behavior* indicates possibilities of the formation of new structures.

To study the relative energy dissipation of a selected mechanism under the influence of the relative dissipations of other mechanisms, KLAMECKI'S model ideas are modified and adapted to the considerations of the situation in a stressed grease film.

The following dissipation mechanisms are selected:

- thermal dissipation E_k;
- mechanical dissipation through fragmentation E_i;
- mechanical dissipation through coagulation (collision of particles) E_j

The following assumptions are made as influencing the energy dissipation rate of a mechanism:

- a mechanical dissipation always runs in parallel with a thermal dissipation;
- the energy dissipation rate is influenced by the amount of energy that is applied to the system $E_0 = E_k + E_i + E_j$;
- there is an influence of the energy dissipated up to now;
- the energy dissipation rate changes relative with the energy dissipated in the mechanism i in relation to the energy dissipated in j and k

The description of these considerations provides the following

$$\frac{dE_i}{dt} = zE_k E_i + a_i E_0 - b_i E_i^m + c_i E_i E_j E_k \tag{16}$$

The circumstance of mutual influence delivers

$$c_i = \left[1 - \frac{d_{ij} E_j^{pi} + d_{ik} E_k^{pk}}{d_{ii} E_i^{pi}}\right] \tag{17}$$

and for the other mechanisms

$$c_j = \left[1 - \frac{d_{ji} E_j^{pj} + d_{jk} E_k^{pk}}{d_{jj} E_i^{pj}}\right] \tag{18}$$

$$c_k = \left[1 - \frac{d_{ki} E_j^{pk} + d_{kj} E_k^{pk}}{d_{kk} E_i^{pk}}\right] \tag{19}$$

and

$$z_i = \left[1 - \frac{d_{ik} E_k^{pk}}{d_{ii} E_i^{pi}}\right] \tag{20}$$

$$z_j = \left[1 - \frac{d_{jjk} E_k^{pk}}{d_{jj} E_i^{pj}}\right] \tag{21}$$

This notation of the mutual influence of the mechanisms and their dissipation rate follows KLAMECKI. For example, it follows for the mechanism i from Equation (16)

$$\frac{dE_i}{dt} = \left[1 - \frac{d_{ik} E_k^{pk}}{d_{ii} E_i^{pi}}\right] E_i E_k + a_i E_0 - b_i E_i^m + \left[1 - \left(\frac{d_{ij} E_j^{pi} + d_{ik} E_k^{pk}}{d_{ii} E_i^{pi}}\right)\right] E_i E_j E_k \tag{22}$$

As long as an experimental investigation of the dissipation mechanisms is not possible, the introduced parameters d, p, a, b, and m remain unknown. To represent the energy dissipation behavior, a two-dimensional surface is stretched out and the slopes are placed in relation to each other. The focus here is on possible cyclical behavior.

There are an infinite number of solutions that depend on the initial value and the parameters listed. Experimental investigations could not be carried out, and an idea has to be developed first, for which experimental procedures would be suitable. A numerical calculation was performed using *Geogebra* from the University of Vienna for arbitrary parameters and initial values (Figure 11), to see if conditions exist that initiate the behavior we are looking for.

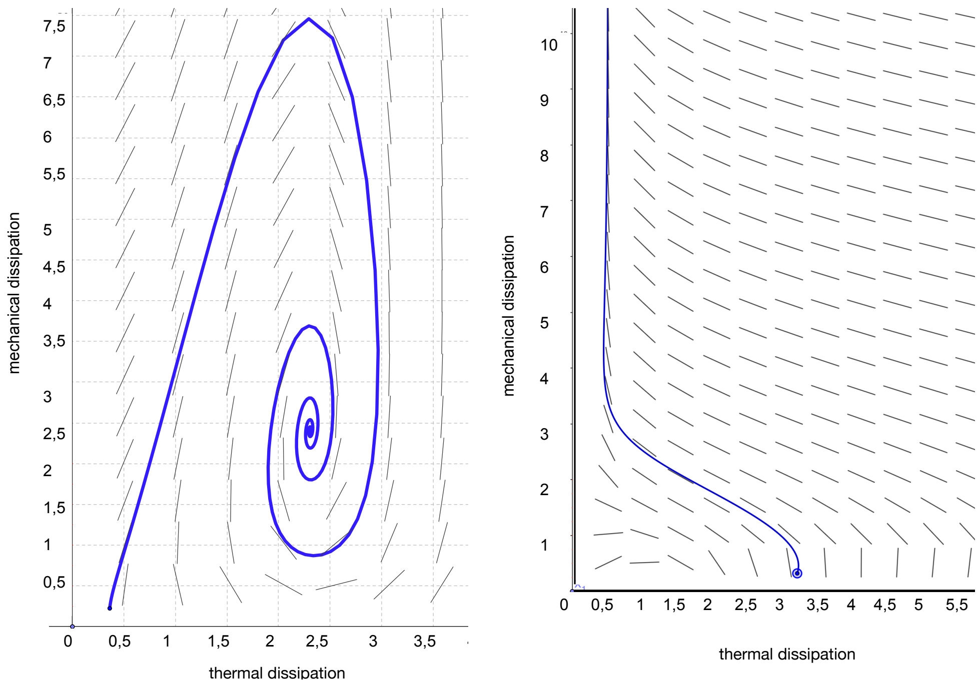

Figure 11. Interesting behavior of the energy dissipation rate for two selected mechanisms. (**left**) cyclic behavior as an indication of the possible formation of dissipative structures, (**right**) change in energy dissipation rate from thermal to mechanical dissipation.

3. Conclusions

The aim of this work was, on the one hand, to investigate the excess entropy rate under conditions that lead to instability and thus to the possibility of a self-organization process. This was attempted by considering an interaction between friction and structural degradation. On the other hand, the relative energy dissipation rates of selected interacting dissipative processes were brought into relation, in order to see whether, in principle, conditions exist that would cause a cyclic behavior. This behavior is also an indirect indicator for the formation of dissipative structures, i.e., for the triggering of self-organizing processes. The tribological system observed is a stressed grease volume, in which changes in the grease structure are observed. An open thermodynamic system is derived. When the interaction of friction and wear is taken into account, it can be seen that the previously found influence of the solid content on the formation of dissipative structures [20] can also be found under the conditions under consideration (Equation (6)). In the investigation of the three selected mutually influencing energy dissipation mechanisms, the investigation showed that there are conditions that produce a cyclic behavior. This is also an indicator [17] of the possible structural formation. The statement of the described models is of course relativized, due to the lack of experimental investigation. However, the possible descriptions and interpretations of the results (Figure 11) are extremely interesting from the point of view of the research question. The investigations presented here, along with other studies, also suggest the possibility of the formation of dissipative structures in an energetically loaded lubricating grease film. The information found makes it possible to better assess the very special tribological behavior of these viscoelastic lubricants.

Funding: This research received no external funding.

Data Availability Statement: No new data were created or analyzed in this study. Data sharing is not applicable to this article.

Conflicts of Interest: The author declares no conflict of interest.

Abbreviations

The following abbreviations are used in this manuscript:

E_f	applied friction energy [J]
E_0	amount of energy that is applied to the system [J]
F_f	fragmentation rate
S	entropy [J/K]
T	temperature [K]
V	volume [m^3]
(d, p,q)	parameter that describe the material behavior
k	consistency
m	mass [kg]
n	flow index
t	time [s]
(z, a, b, c)	are functions of the dissipation mechanisms
ϵ	parameter that describes the distance to equilibrium
λ	heat conductivity [W/(mK)]
ρ	density [kgm^{-1}]
τ	shear stress [Pa]
τ_{ostw}	shear stress, Ostwald-de Waele behavior [Pa]
$\dot{\gamma}$	shear rate [s^{-1}]

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