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# Effect of friction on the microstructure of compacted solid additive blends for polymers

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ABSTRACT

The present study is concerned with the characterization of the heat generation during the pelletizing process that produces a shell/core pellet structure. Shell formation contributes to the mechanical durability of compacted pelletized additive blends and alleviates health and environmental issues by handling powder additives. A model system comprised of erucamide (low melting point component) and silica was selected for the investigation. The thermal conductivity of the model system was investigated as a function of the erucamide content. The frictional heat generated during the pelletizing process was studied by conducting tribological experiments. The results show that thermal conductivity behaves according to the rule of mixtures. Frictional heat was correlated to the depth of the shell formation examined by Scanning Electron Microscopy after testing. Temperature distributions using semi-infinite solid approximation with constant surface temperature show that the inside boundary of the shell microstructure is almost always above 70 °C. Differential Scanning Calorimetry experiments show that erucamide, the low-melting component, has an onset of melting at temperatures around 70 °C, well below its melting point (83 °C). This suggests that frictional heating generated at the surface raises temperature until onset of melting of erucamide takes place which leads to the formation of a shell/core microstructure.



## 1. Introduction

Additives are used to modify the processing and properties of polymers [1,2]. For years the industry has been working with powd ers as the main form for additive delivery during the extrusion of polymers. However, there are disadvantages associated with handling powder additives, such as health and environmental risks. The compaction of additive blends is a new venture in the polymers industry which can provide promising improvements in the processing of the industrial practice of additives. The specific advantages of compacted additive blends over powder form are their superior flow properties, minimal dust generation during handling and transport, reduced health hazards associated with dust exposure of personnel, lower risk of dust explosion, formulation flexibility, cost savings due to less handling equipment, purchasing inventory control and logistics [3].

A novel approach for the preparation of compacted additive blends is to make compacted pelletized additive blends using a solids-state compaction process [4]. In this process at least one of the additives is a high-melting additive with a melting temperature above 300 °C and at least one additive is a low-melting additive with a melting temperature below about 90 °C. Typical high-melting additives include silica, talc, diatomaceous earth, and titanium dioxide. Typical low-melting additives include erucamide, oleamide, and glycerol monostearate. The additives in their original form are mixed together. Then, the mixture is pelletized to form compacted cylindrical pellets in a pellet mill that consists of rollers and a perforated die, with each part serving an instrumental role in the process. During this process heat is generated due to friction between the die and material [5], resulting in an increase in temperature until it reaches a constant value during steady-state. Since the process for preparing compacted pelletized additive blends is a fairly new development, few studies have focused on it. To date, the effects of compression ratio and critical binder concentration on resulting mechanical durability and microstructure of resulting erucamide–silica pellets have been investigated [6]. However, there are still several unknowns associated with the process. More specifically, thermal properties and frictional heat generation are believed to be key contributing factors leading to the high durability of compacted pellets of additive blends made using this process which helps them maintain their shape and density.

The ASTM E1225-09 comparative method is a standard technique to measure the thermal conductivity of homogeneous solids in the range of 0.2 W $m^{-1}$ $K^{-1}$–200 W $m^{-1}$ $K^{-1}$ over the temperature range of 90–1300 K [7]. In general, a specimen is placed under a load between two references with known thermal conductivity. A steady state temperature gradient is established and the thermal conductivity is calculated by using the measured gradient in the references and their thermal conductivity.

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**List of symbols**

| | |
|---|---|
| $\rho$ | density (kg/m$^3$) |
| $m$ | mass (kg) |
| $D$ | diameter of the die chamber (m), |
| $H$ | height (m) |
| $k_{eff}$ | effective thermal conductivity (W m$^{-1}$ K$^{-1}$) |
| $k_{ref}$ | known reference thermal conductivity (W m$^{-1}$ K$^{-1}$) |
| $\frac{\Delta T}{\Delta Z}$ | temperature gradient (K/m) |
| $d$ | contact diameter (m) |
| $N$ | number of laps. |
| $t$ | adjusted actual time (s) |
| $\mu$ | coefficient of friction (CoF) |
| $Q$ | total average frictional heat (J) |
| $V$ | linear velocity (m/s), |
| $\delta$ | thermal penetration depth (μm) |
| $\alpha$ | thermal diffusivity (m$^2$/s) |

Hadley utilized a method similar to the ASTM E1225-09 which he referred to as the steady-state comparator method to measure the effective thermal conductivities of consolidated binary mixtures of metal powders [8]. Experimental measurements of the effective thermal conductivities yielded results between 0.0369 W m$^{-1}$ K$^{-1}$ and 36.81 W m$^{-1}$ K$^{-1}$, depending on metal, composition, and sample form.

Leung et al. built a thermal conductivity analyzer that utilized the ASTM E1225-04 comparative method to measure the effective thermal conductivity of polymer composites with heterogeneous structures [9]. Experimental measurements of the effective thermal conductivities yielded results between 0.22 W m$^{-1}$ K$^{-1}$ and 1.89 W m$^{-1}$ K$^{-1}$.

Various idealized models have been developed to estimate the frictional heat generated between two sliding surfaces contacting each other [10]. However, due to the inherent complexity of the contact area, contact stress, surface roughness, wear, and more, idealized analytical solutions may be limited and provide only initial estimates [11]. With the development of advanced computer power modeling and simulation, more complex analytical and finite element solutions are starting to be implemented [12]. The circular contact analysis with one body in motion is an idealized model where a protuberance on a body forms a circular contact area with the flat surface of a second body. This can be utilized in applications such as friction testing with a tribometer [2,10].

The overall objective of this study is to provide insight into the thermal processes that are involved in the solid-state compaction process of solid additive blends and how they affect the resulting microstructure. Simple thermal transport mechanisms can be modeled using simple geometries, such as semi-infinite bodies, and thin films to yield exact or approximated analytical solutions. These demand knowledge of physical and thermophysical properties, such as density, thermal conductivity, and thermal capacity, of the materials involved. Although these properties may change as a function of temperature, it is a good approximation to use constant values for solids and molten states. When dealing with systems comprised of a collection of particulate solids, much more complex analysis is required. However, one way in which a rigorous treatment may be avoided is by applying analytical solutions of thermal heat transfer problems by replacing the overall heat transfer properties with "effective" values [13]. Therefore, the specific goal of this paper is to study the correlation between frictional heating and the resulting shell/core microstructure of compacted solid additive blends for polymers.

## 2. Experimental section

### 2.1. Materials

A model system was selected for the study based on two requirements: (i) the system must be comprised of one "ideal" low-melting additive and one "high-melting" additive; (ii) the combination should be as applicable to the polypropylene/polyethylene resins that are used in the polymers industry. A binary model system comprised of erucamide and silica was selected in the present study. "Crodamide ER Bead" is a commercially available erucamide from Croda. It is a vegetable oil based linear fatty acid amide in the form of beads with an average particle size of about 1.5 μm, melting point of 83.0 °C, and a bulk density of 570 kg/m$^3$. It is used as a slip agent in polyolefins film processing. "Gasil AB725" is commercially available silica from PQ Corporation. It is synthetic amorphous silica in fine powder form with an average particle size of 5.3 μm, high melting point, and a bulk density of 220 kg/m$^3$. It is used as an antiblock agent for polyolefins film processing.

### 2.2. Sample preparation

Compacted samples 50.4 mm in diameter and about 5 mm in thickness were prepared for thermal conductivity measurements and tribological testing using a custom built laboratory single press pelletizing apparatus. It is comprised of a hardened steel cylinder die with a removable bottom and a piston with a tight fit. The piston is pushed using a hydraulic mechanism.

Appropriate amounts of erucamide and silica were weighed separately using an analytical scale in order to yield a total sample weight of about 10 g. The two components were pulverized by a mortar and pestle to simulate the action that occurs inside of a pelletizing unit. After crushing, both materials were mixed in order to make a homogenous powder.

Four different samples with varying composition of erucamide and silica were prepared according to the procedure detailed above as detailed in Table 1.

Samples for thermal conductivity measurements and frictional testing were compacted in the laboratory single pelletizer. Mold release was sprayed on all the surfaces of the hardened steel chamber, the additive powder mixture was loaded into the die and the piston was inserted. The die assembly was placed on top of the driving mechanism and pressure was then applied. Samples were slowly compacted to a desired final density of 850–1005 kg/m$^3$, depending on composition. The density ($\rho$) of the material inside the die was calculated in-situ using the following equation :

$$\rho = \frac{4 \times m}{\pi \cdot D^2 \times H} \quad (1)$$

where $m$ is the sample mass, $D$ is the diameter of the die chamber, and $H$ is the sample height. Once the desired density was achieved, pressure was released and the compacted sample was removed

**Table 1**
Composition of erucamide and silica mixtures.

| Sample | Erucamide (wt%) | Silica (wt%) |
|---|---|---|
| 1 | 25.0 | 75.0 |
| 2 | 50.0 | 50.0 |
| 3 | 75.0 | 25.0 |
| 4 | 100.0 | 0.0 |