Quantitative characterization of solid lubricant transfer film quality

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ABSTRACT

Solid lubricant materials have become necessary in applications for which traditional lubrication approaches become impractical. These materials are often mated against a harder metallic counter surface (counterface) of higher surface energy. During sliding, wear fragments from the solid lubricant transfer to the counterface to form a protective barrier known as the transfer film. Historically, the coverage attributes of these transfer films have correlated strongly to the tribological performance of the solid lubricant. Although transfer film quality is often identified as a critical contributor to the success of a candidate solid lubricant, the community lacks a quantitative means to measure quality. Transfer film cohesion and adhesion are likely very important but they are also difficult to measure and not necessarily related to the visual features that have motivated the use of adjectives like 'quality', 'thin', 'uniform', and 'tenacious'. Area fraction and film thickness are more easily quantified, but to date, they have not proven to be robust predictors of tribological success. A recent visual study of transfer film evolution for a successful alumina–PTFE nanocomposite suggests that the characteristic *size of domains of exposed counterface* may correlate more closely with wear performance. This paper presents a method for quantifying transfer film quality based on this metric, which we call the free-space length (L_f). To illustrate the application of the method, we study the connection between the wear rate and free-space length for the transfer film of a well-studied alumina–PTFE system. The correlation to wear was best for the free-space length and worst for area fraction.

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1. Introduction

Solid lubricant materials are necessary in applications for which cost, complexity, contamination, and environmental constraints preclude more traditional fluid and grease lubrication strategies. Molybdenum disulfide, graphite, hexagonal boron nitride, and polytetrafluoroethylene (PTFE) are notable examples. Although bulk polymers generally lack sufficient lubricity or wear resistance for bearing applications, their tribological properties can be improved dramatically with the incorporation of fillers [1]; polymer composite solid lubricants with a wide range of physical properties are now commercially available and used in a variety of challenging industrial applications (*e.g.* space, mining, harvesting equipment, and wind turbines).

These materials are often mated against a harder metallic counter surface (counterface) of higher surface energy. During dry sliding, fragments of the solid lubricant adhere to and transfer to the counterface. As these fragments accumulate, they eventually form a film that protects the bulk solid lubricant from counterface asperities [2,3]; this film is known as the transfer film. The rate of transfer film removal and re-deposition represents the lower limit of the system wear rate [4], and the wear reducing effect of fillers is often discussed in terms of their ability to improve transfer film adhesion and otherwise reduce transfer film wear [3–8].

Most wear studies of filled polymers include some form of transfer film analysis. Bahadur et al. [9] found that the most successful wear-reducing fillers were those that promoted thin, uniform, and strongly adhered transfer films through mechanisms that include filler decomposition and mechanical interlocking of filler particles into counterface scratches. To our knowledge, only Schwartz and Bahadur have attempted to measure the bond strength of the transfer film [10]. They bonded a copper tab to the transfer film with cyanoacrylate and found that the wear rate decreased as the bond strength increased. The authors described poor performing transfer films as 'patchy', and 'non-uniform', while successful transfer films were described as 'uniform' and 'continuous'. Wang et al. found that various nanofillers significantly reduced the wear of polyetheretherketone (PEEK) and concluded that the wear reduction mechanism was tied to the filler's ability to improve characteristics of the transfer film [11–13]. The authors described the improved films as thin, uniform and tenacious. Li et al. [14], Chen et al. [15], Sawyer et al. [16], and

Bhimaraj et al. [17] made similar observations and drew similar links between the effects of nanofillers, wear resistance and transfer film quality. Krick et al. showed that the absence of environmental moisture disrupted transfer film formation and prevented an otherwise low wear alumina–PTFE nanocomposite from achieving low wear [18]; in other words, wear resistance required the high quality transfer film that forms in humid air.

Most transfer film analyses involve qualitative assessment using optical or electron microscopy [1,3,6,8,11,19–21]. However, there have been a number of efforts to quantify transfer film quality and correlate it to tribological performance. Wheeler used XPS to quantify the transfer film thickness of PTFE on steel in vacuum at varying speeds [22]. Jain and Bahadur [23] used infrared spectroscopy to quantify the transfer of low surface energy polymers to higher surface energy polymers. Blanchet et al. [24] used XPS to distinguish the transfer behaviors of unfilled and filled PTFE and Yang et al. [25] used X-ray yield measurements to quantify transfer film thickness of PTFE on stainless steel as a function of temperature. It could be argued that these methods were quantitative but indirect measures of transfer film thickness. Burris and Sawyer used optical profilometry to make direct measurements of transfer film thickness for PTFE, a PTFE micro-composite, and two PTFE nanocomposites [20]. The steady-state wear rate of these PTFE-based materials varied over three-orders of magnitude and was proportional to the cube of the transfer film thickness. Blanchet et al. made consistent observations with stylus profilometry on similar systems [26]. Laux and Schwartz used optical profilometry to correlate transfer film thickness to the wear resistance of unfilled PEEK [27,28]. They found no significant correlation between the two, and suggested that *film discontinuity* might be more strongly related to wear. Bowden et al. [29] proposed that friction is a linear function of the boundary film area fraction and it would therefore be reasonable to expect a similar relationship for wear. To our knowledge, only Bhimaraj et al. has conducted a quantitative study of the effect transfer film area fraction on wear [30]; there was no significant correlation

between transfer film area fraction and wear rate of polyethylene terephthalate (PET) nanocomposites.

Ye et al. [31] used quasi-*in-situ* methods to study the morphological evolution of a PTFE nanocomposite transfer film as the wear rate decreased by more than three orders of magnitude during run-in. The transfer film, which was initially thick, patchy, removed and replenished on a cycle-by-cycle basis, became thinner, more continuous, and longer lasting as the system transitioned to low wear. The results suggest that the characteristic size of 'transfer film-free' domains of the counterface may correlate more closely to wear performance than thickness or area fraction; this is one possible measure of film 'discontinuity' as discussed by Laux and Schwartz [27]. This paper presents a method for quantifying transfer film quality based on this metric, which we call the free-space length (L_f). To illustrate the application of the method, we study the connection between the wear rate and free-space length of the transfer film for a well-studied alumina–PTFE system.

2. Method

Transfer films lubricate by effectively modifying the mechanical and chemical properties of the counterface. Imagine a typical patchy transfer film like that shown in Fig. 1(a). The transfer film covered regions have reduced surface energy and hardness, which mitigate adhesive and abrasive wear, respectively. The bare or uncovered regions have high surface energy and will adhere most strongly to the solid lubricant. The size of this uncovered region limits the size of the potential adhered debris particle and, as Rabinowicz showed, smaller debris particles have increased ratios of surface energy to elastic energy, and are therefore less likely to be removed from the counterface [32]. Although thickness also reflects debris size, the lateral dimensions are more closely related to common visual cues (*e.g.* uniformity, coherence, homogeneity and continuity).
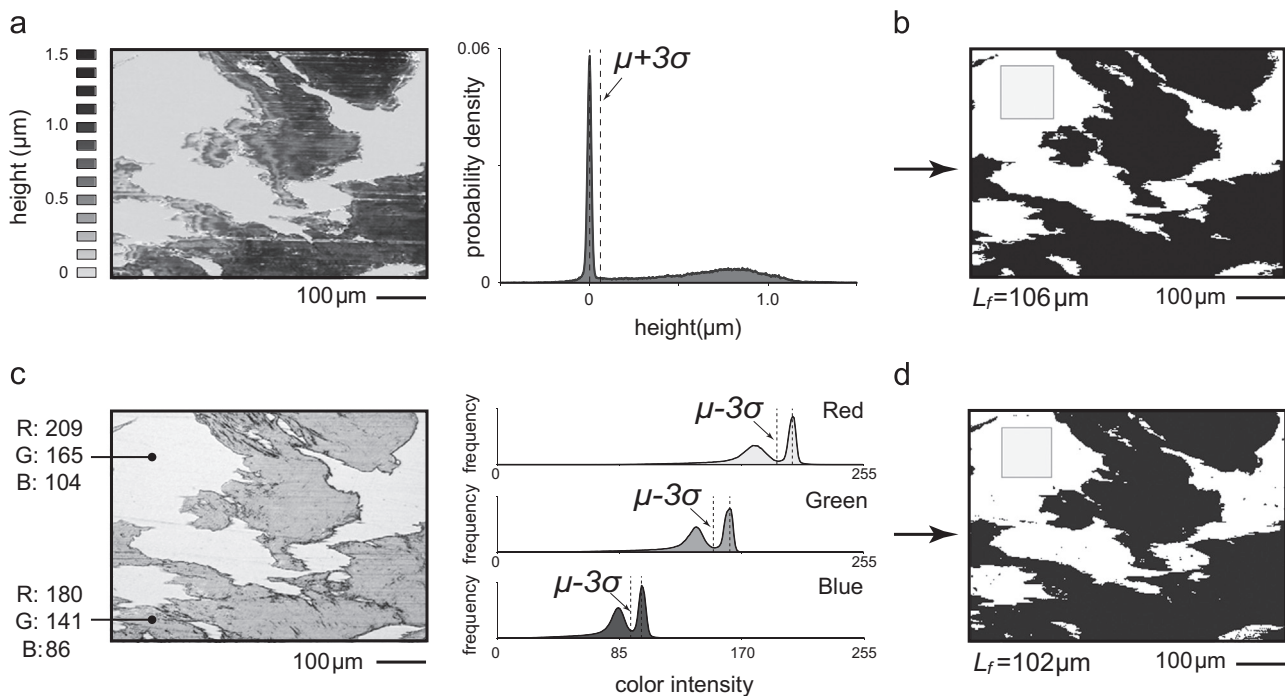

Fig. 1. Illustration of the conversion of a height map (a) or optical image (c) of a transfer film into black and white using thickness and color intensity (RGB or grayscale) thresholds, respectively. The corresponding height and color histograms are shown for reference. Each distribution is bimodal with one mode corresponding to the counterface and the other corresponding to the film. Transfer film in (a) is any pixel higher than 3σ above the mean, or $(\mu + 3\sigma)_{thickness}$; the same strategy is used for the optical image but the film has a lower intensity.