

Tribocorrosion Behavior of Aluminum Alloys

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Tribocorrosion is a material degradation process caused by the combined effect of wear and corrosion. The complexity of tribocorrosion lies in the fact that the chemical and mechanical attacks are not independent of each other but often act synergistically to cause accelerated failure.

Keywords: corrosion ; tribocorrosion ; aluminum alloys

1. Introduction

Tribocorrosion is a material degradation process caused by the combined effect of wear and corrosion ^{[1][2]}. The complexity of tribocorrosion lies in the fact that the chemical and mechanical attacks are not independent of each other but often act synergistically to cause accelerated failure ^{[3][4][5]}. The synergetic effect is most prominent for passive metals. Passive metals, which include important engineering metals such as stainless steels, aluminum (Al), and titanium alloys, spontaneously form a thin oxide film (passive film) when in contact with oxygen or water ^[6]. This passive film, sometimes only a few atomic layers thick, acts as a critical protective barrier against corrosion ^{[2][6]}. When mechanical wear occurs during corrosion, the passive film can be locally destroyed at the contacting asperities, with the ensuing depassivation leading to rapid localized corrosion and early component failure ^{[2][3][7][8][9]}. A better understanding of the tribocorrosion response of passive metals is required to extend the durability of these technologically important metals in complex service conditions.

2. Effects of Alloying and Grain Size on Wear Resistance of Al Alloys

2.1. Alloying Effects

Alloying leads to increased hardness and improved wear resistance of metals in most cases per Archard's law ^{[10][11]}. In addition to hardness increment, proper alloying additions can significantly lower the grain boundary energy and stabilize ultrafine microstructures in the tribolayer to suppress stress-assisted grain growth and improve the surface hardness and the overall wear resistance ^[12]. For example, Jiru and Sing ^[13] showed that surface alloying of Al with manganese (Mn) improved surface hardness, lowered the coefficient of friction, and reduced wear rate of aluminum by 30%. Dubourg et al. ^[14] found that the wear rate of Al–Cu alloys decreased with increasing Cu concentration from pin-on-disk wear tests under 2–4 N load and 0.05–0.2 m/s sliding speed. Du et al. ^[15] demonstrated a similar behavior in additively manufactured Al-5 wt.%Cu alloys via laser powder bed fusion, where the addition of ~1.5 wt.% Ti results in a reduction of wear rate from $9.79 \times 10^{-4} \text{ mm}^3/(\text{N}\cdot\text{m})$ to $3.05 \times 10^{-4} \text{ mm}^3/(\text{N}\cdot\text{m})$ after ball-on-disk wear tests under 10 N load for a sliding time of 30 min. In emerging high-entropy alloys, the wear coefficient of $\text{Al}_x\text{CoCrCuFeNi}$ alloys with 9–28.6 at.% Al decreases as the aluminum content increases ^[16]. Specifically, the wear coefficient of high aluminum content alloy is about one seventh of that of low aluminum content counterpart due to a change of the wear mechanism from delamination wear to oxidative wear with increasing aluminum content. Yu et al. ^[17] found that AlCoCrFeNiTi0.5 shows better dry wear resistance than wear-resistant steel AISI 52100 due to nano-sized Fe–Cr solid solution and Al–Ni–Ti rich intermetallic phases. However, the opposite has also been reported in terms of both hardness and wear resistance. For example, the addition of Zr, Mn, V, Cr, and other micro-elements to 7xxx series aluminum alloy (Al–Zn–Mg) results in a decrease of hardness ^[18]. De Rosso et al. ^[19] showed that the wear rate of Al–Si alloys decreases at higher Cr content but lower Si contents due to the smaller secondary dendritic arm spacings and higher hardness.

From a more general point of view, it is interesting to note that subsurface microstructure tend to be driven into a non-equilibrium status under constant wear and friction, leading to dynamically self-organized microstructures similar to those in ball-milling ^[20] or under ion irradiation ^[21]. In non-equilibrium systems, when several competing processes are present (such as dislocation accumulation due to sustained plastic deformation vs. stress-assisted grain growth and dynamic recovery), a steady state and equilibrium microstructure is not always achieved ^[22]. For example, Kasai et al. ^[23] performed wear tests of pin sliding against a brass disc in air and found that the Kelvin probe signal on the surface is

nearly periodic, indicating the periodic change of surface chemistry and structure and an absence of steady state. Another example of the lack of a steady state in wear is the well-known stick-slip frictional behavior observed in many elastomers when subjected to low sliding velocities and/or high normal pressures [24]. However, non-equilibrium surface microstructures, such as supersaturated solid solutions has also been reported in the tribolayer [25], where the level of supersaturation is far beyond the equilibrium solubility limit, similar to those observed during mechanically alloying and severe plastic deformation [26][27].

Alloying effects on wear of Al is complicated due to the various phase distribution and microstructure formation in different alloying systems. Generally speaking, higher alloy concentration leads to improved wear resistance but an optimum alloy concentration for the best wear resistance is not known *a priori*. For example, Jasim and Dwarakadasa [28] showed that the wear rate of Al–Si alloys decreases with Si content up to the eutectic composition and did not depend on the initial microstructure or the distribution of the silicon phase. Similar behavior is also observed in Al–Cu [14], Al–Cu–Fe [29], Al–Sn [30], etc. From a more general point of view, it is interesting to note that subsurface materials tend to be driven into a non-equilibrium status under constant wear and friction, leading to dynamically self-organized microstructures similar to those in ball-milling [20] or under ion irradiation [21]. In non-equilibrium systems, when several competing processes are present (such as dislocation accumulation due to sustained plastic deformation versus stress-assisted grain growth and dynamic recovery), a steady state is not always achieved [22]. As a result, a single optimum alloy concentration may not always exist.

2.2. Grain Size Effects

In addition to alloying, another important tunable microstructure feature is grain size [31]. Refined grain size often leads to increased hardness and improved wear resistance, per Archard's law [10], especially when abrasive wear is the dominating mechanism. For example, nanocrystalline Al with ~16 nm grain size exhibits a reduction of friction coefficient by about 55% compared to microcrystalline Al [32]. Farhat et al. [32] investigated the friction and wear characteristics of nanocrystalline Al as a function of grain size. They found that both mild and severe wear increased linearly with the square root of grain size, following a modified Archard-type of relationship. However, when high stress is applied during wear, the large amount of plastic deformation at the contact surface leads to stress-assisted grain growth due to the high mobility of nanocrystalline grain boundaries and results in wear resistance higher than what would be expected based on hardness alone [8][33].

It should be pointed out that since these wear tests were performed at room temperature and moderate sliding speed, frictional heating was ruled out as the driving force for the observed grain growth [8]. Another deviation from Archard's law is that the optimum grain size for wear resistance does not coincide with the grain size for the highest hardness at the Hall–Petch crossover. The optimum grain size for wear is not known *a priori*. For example, Jeng et al. [33] showed that the hardness of nanocrystalline Ni reaches a maximum at grain size of 15 nm, while the optimum wear rate is reached at grain size of 30 nm. Further reducing the grain size does not alter the wear rate much. From a more general point of view, it is interesting to note that subsurface materials tend to be driven into a non-equilibrium status under constant wear and friction, leading to dynamically self-organized microstructures similar to those in ball-milling [20] or under ion irradiation [21]. In non-equilibrium systems, when several competing processes are present (such as dislocation accumulation due to sustained plastic deformation versus stress-assisted grain growth and dynamic recovery), a steady state is not always achieved [22]. As a result, a single optimum grain size may not always exist.

3. Effects of Alloying and Grain Size on Corrosion Resistance of Al Alloys

3.1. How Does Alloying Affect Corrosion of Al Alloys?

Al and Al alloys form an amorphous semiconducting passive film in neutral (pH 4–9) solutions [34]. This passive film is still vulnerable to the local attack of different halide ions. The adsorption of halide ions on the surface leads to localized breakdown of the passive film and eventually leads to pitting and crevice corrosion of Al. Unlike in wear-only conditions, alloying may affect corrosion resistance of Al in different ways depending on the specific alloying elements, as reviewed by Szklarska–Smialowska [35]. Small quantities of Sn, In, Hg, Ga, and Zn are detrimental to Al corrosion, as they reduce the passive potential region and shift the corrosion and pitting potentials in the negative direction [36]. Thus, these transition metals (TMs) lead to high anodic current density and uniform active surface corrosion. Other TMs, such as Cu, Mo, Mn, W, Nb, Cr, Ta, V, and Zr, improve corrosion resistance and decrease pitting susceptibility of Al by increasing the overpotential for anodic dissolution and decreasing metastable pit initiation and growth rates [37]. For example, Mraied et al. [38] showed that increasing the Mn concentration of Al–Mn alloys from 0–20.5 at.% resulted in higher corrosion resistance and the formation of much smaller pit sizes at higher Mn content. Unlike pure Al (A0), which clearly showed

through thickness circular “pits” of ~120 µm in diameter after 24 h of immersion in 0.01 M NaCl solution, Al–Mn alloys did not reveal any marked localized attack after 108 h of immersion. Instead, a network of conspicuous grooves developed on alloy A20 with ~ 100 nm in depth. Corrosion mechanisms of Al has been reviewed from the perspectives of acid–base interactions [39], chloride ion interactions with surface oxide film [40], critical steps involved in localized corrosion and stressed corrosion [41], and the effects of corrosion inhibitors [42], etc. Specifically, the review by Olugbade [43] discusses the effects of plastic deformation, including ultrasonic shot peening, equal-channel angular processing, constrained groove pressing, etc., and addresses the impacts of deformation on corrosion, which shares some similarity with tribocorrosion for the wear-induced gradient subsurface microstructure [44].

3.2. Corrosion Resistance Depend on Grain Size

Unlike in wear-only conditions, grain size may affect corrosion resistance in different ways depending on the specific corrosion mode. For the technologically important case of corrosion resistance of passive metals, it is thought that grain size influences corrosion resistance by affecting the rate of ion transport through the passive film, as well as the passive film dissolution rate [45]. The nanocrystalline materials involve a much higher linear boundary intersection density at the surface compared to their microcrystalline counterparts. This high density of grain boundaries leads to higher average surface reactivity through increased electron state density and diffusional defect transport. Those factors are believed to lead to higher stability and better adherence of the passive film on nanocrystalline materials [45]. For passive metals, including Al [46][47], Ti [48], Ti alloys [49], and steel [50][51], it has been shown that indeed reducing grain size leads to improved corrosion resistance. However, it should be pointed out that these results are drawn from a wide variety of test conditions that often do not isolate the effect of other microstructural changes imparted by the grain refining process from the effect of grain size decrease alone. These microstructural changes include, among others, a secondary phase refinement, texture evolution, and residual stress build-up. As experimental focus and conditions vary among various studies, reported findings on grain size effects to date often tend to be contradictory. Orłowska et al. [52] studied the effect of grain size on the corrosion behavior of commercially pure Al processed by multi-turn equal channel angular pressing and upsetting. Despite the significant change of grain size from ~11 µm to ~669 nm after severe plastic deformation, grain refinement was found to show negligible effects on the pitting behavior. Instead, Fe-containing particles, which are pit nucleation sites, are determined to be the crucial factor for the observed corrosion behavior. Since there is no difference in size and number of these particles after the severe plastic deformation, the electrochemical parameters showed very similar values for samples with different grain sizes. Olugbade reviewed the corrosion rate of Al alloys processed by equal channel angular pressing (ECAP) and summarized that the corrosion current density generally decreases at smaller grain size. Ralston et al. [46] studied corrosion resistance of high-purity Al with grain sizes from ~100 to 2000 µm processed using different plastic deformation methods. They found, albeit with considerable remaining uncertainty, that corrosion resistance increased with the reciprocal square root of grain size in a manner analogous to the Hall–Petch relationship. Similar behavior was found for nanocrystalline Al and Al alloys [47][53][54]. However, the opposite behavior has been reported by others [55][56]. Eizadjou et al. [55] showed that Al with grain size of ~380 nm produced by accumulative roll bonding (ARB) was more susceptible to corrosion than coarse grained Al with ~ 33.8 µm grain size, a result that was interpreted as due to a high density of defects and inhomogeneities in the passive film. Similar contradictory results have also been reported in Ti alloys [49][57] and Mg alloys [58][59].

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