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Room temperature spallation of α-alumina films grown by oxidation

S. Wang [a,b], C.M. Harvey [a,*], B. Wang [a]

[a] Department of Aeronautical and Automotive Engineering, Loughborough University, Loughborough, Leicestershire LE11 3TU, UK
[b] School of Machinery and Equipment Engineering, Hebei University of Engineering, Handan, China

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A B S T R A C T

Tolpygo and Clarke (2000) presented an excellent experimental study on the room temperature circular spallation of α-alumina films grown by oxidation on Fe-Cr-Al alloy. Their observations are remarkable and thought-provoking and are worthy of mechanical interpretation. The present work hypothesizes that pockets of energy concentration (PECs) exist due to dynamic and non-uniform plastic relaxation or creep in the film and Fe-Cr-Al alloy substrate during cooling. PECs may be the driving energy for room temperature spallation failure. Based on this hypothesis, an analytical mechanical model is developed in this work to predict the spallation behavior, including the separation nucleation, stable and unstable growth, and final spallation and kinking off. The predictions from the developed model are compared against experimental results and excellent agreement is observed. The work reveals a completely new failure mechanism of thin layer materials.

1. Introduction

Tolpygo and Clarke [1,2] presented an excellent experimental study on the room temperature spallation failure of α-alumina films grown by oxidation on Fe-Cr-Al alloy. Their observations are remarkable and thought-provoking. Refs. [1,2] convincingly demonstrate a thin film spallation process with an unknown mechanical mechanism. To help readers understanding the present work, a detailed introduction to Refs. [1,2] is thought to be necessary. In their work, α-Al_2O_3 films of different thicknesses were formed on the surface of Fe-Cr-Al heat-resistant alloy substrates of different thicknesses by oxidizing them at 1200 °C for different time periods. Then, the film-substrate material systems were cooled to room temperature at different cooling rates. Cooling causes an increase of compressive in-plane residual stress in the α-Al_2O_3 films due to thermal expansion mismatch between the films and the substrates. Their major observations were as follows: No separation or spallation failure occurs during cooling at any rate. For specimens cooled to room temperature at rates in the range 5°–200 °C min^{-1}, circular interfacial separations develop, apparently spontaneously, at a constant compressive residual stress far below the critical buckling stress: The separations nucleate, grow in separation distance and propagate radially. After a period of slow and stable growth, some of these separations then grow abruptly and the oxide spalls off. For specimens cooled at extremely slow cooling rates (≤2 °C min^{-1}) and at very fast cooling rates (≥500 °C min^{-1}), no separation or spallation occurs at any point.

* Corresponding author.
E-mail addresses: s.wang@lboro.ac.uk (S. Wang), c.m.harvey@lboro.ac.uk (C.M. Harvey), b.wang2@lboro.ac.uk (B. Wang).

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Nomenclature

A	amplitude of upward deflection of film bubble
E	Young's modulus of film
G, G_I, G_{II}	total, mode I and mode II ERRs
G_{Ic}, G_{IIc}	film-substrate pure mode I and II interface fracture toughness
G_c	film-substrate mode-dependent interface fracture toughness
G_{cf}	fracture toughness of film
h	thickness of film
M_{rB}, N_{rBe}	crack tip radial bending moment and effective force
r	radial coordinate of circular film bubble
R_B	radius of circular-edged delamination
U_0	strain energy of film bubble before separation
U_a	'bubble energy'; increase in combined strain energy and surface energy due to bubble separation
U_b	bending strain energy of film bubble
U_i	in-plane strain energy of film bubble
U_s	surface energy of delaminated surfaces of film bubble
w	upward deflection of film bubble
z	out-of-plane coordinate
α	buckling correction factor (e.g. due to initial imperfection)
β	kink-off angle
ε_0, σ_0	uniform residual compressive strain and stress in film
$\varepsilon_r^R, \sigma_r^R$	averaged radial relaxation strain and stress due to bending deflection
$\varepsilon_\theta^R, \sigma_\theta^R$	averaged circumferential relaxation strain and stress due to bending deflection
θ	circumferential coordinate of circular film bubble
ν	Poisson's ratio of film
ψ	ratio of film-substrate pure mode II and I interface fracture toughness

1.1. Tolpygo and Clarke's proposed explanations

Various explanations for the phenomenon were proposed and thoroughly and insightfully examined by Tolpygo and Clarke [1,2]. One category of these explanations was the flaw or imperfection hypothesis [3,4], which attempted to explain the nucleation and growth of the separations. The hypothesized flaw consisted of pre-existing separations, cavities or other large defects; or pre-existing inclusions such as Zr-containing oxides; or impurity segregations at the oxide-substrate interface due to the slow cooling rates. Each possible type of flaw was explored in turn. Optical microscopy studies showed that no discernible interfacial separations or spallation existed in any of the specimens when examined immediately after cooling to room temperature. Also, when examining the exposed metal surface after spallation, scanning electron microscopy studies did not reveal any interfacial cavities or voids except for areas near sharp edges at the periphery of the specimens. Some craters on the metal surface were formed by Zr-rich oxide particles in the film but they were largely similar in size to the film thickness. They therefore could not have provided a flaw that was large enough to result in film buckling. Furthermore, these Zr-rich oxide particles were found to resist separation propagation, maintaining stable separation and preventing spallation. Regarding impurity segregation, some impurities such as sulphur, carbon or phosphorous may be expected to segregate at the interface due to the gradual decrease of solubility of the metal during slow cooling. Little difference in segregation was found between slow and fast cooling rates however. This flaw hypothesis was therefore invalidated.

In the second hypothesis, the time-dependent growth behavior of the separations was explained by stress corrosion due to moisture [5]. To have a convincing invalidation of this hypothesis, some slowly-cooled specimens were placed in a sealed container in a purified nitrogen atmosphere with zero humidity. Spallation was still as prevalent as during regular exposure in ambient atmosphere [1].

Cooling rates affect the separation and spallation behavior, as shown by all the specimens. A third hypothesis, therefore, was that metal plastic strain during cooling is the key factor governing the spallation as it is directly related to the cooling rates. Carefully designed experiments, however, showed that the metal plastic strain during cooling was not sufficient to cause spallation of the film [1].

Several other hypotheses were also considered in Refs. [1,2]: Condensation of equilibrium thermal vacancies at the interface during cooling, diffusion of hydrogen or carbon monoxide from the metal to the film causing disruption to the film at room temperature, and metal embrittlement or hardening near the interface. Tolpygo and Clarke [1], however, stated that none of these hypotheses is consistent with all the experimental results. Readers are strongly recommended to read their work [1,2] for a thorough understanding of the above descriptions. A more recent study [6] on the same topic presents some contradictory observations. A major one is that impurity segregation at the film metal interface is indeed a key factor on the separation and spallation of the film.

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