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Journal of the European Ceramic Society 26 (2006) 27–36
www.elsevier.com/locate/jeurceramsoc

Multilayer coatings with improved reliability produced by aqueous electrophoretic deposition

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Received 26 May 2004; received in revised form 19 October 2004; accepted 29 October 2004
Available online 30 December 2004

Abstract

Laminates in the system Al_2O_3/Y-TZP have been produced by an aqueous electrophoretic deposition (EPD) process combined with the use of thermogelling polysaccharides. Mixtures with relative volume ratios of Al_2O_3 to Y-TZP of 95/5, 60/40 and 0/100 have been studied in terms of colloidal stability, gelation behaviour and EPD kinetics. A graded symmetrical architecture, with external layers of A95Z5, and intermediate layers of A60Z40 sandwiching a layer of A0Z100 has been designed by controlling the sequential EPD kinetics of each suspension. The fracture strength of monoliths and laminates has been studied by three point bending and Weibull analysis. Even though the multilayer does not present strength values significantly higher than those of the A95Z5 monolith it presents enhanced reliability, as demonstrated by a higher Weibull modulus. Reliability of the laminate, under critical fracture conditions such as those involved in strength testing, is mostly due to a strong effect of the reduced thickness of the A95Z5 layers as compared to total thickness of the A95Z5 monolith.
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Keywords: Films; Mechanical properties; Microstructure; Strength; Laminates; Al_2O_3/TZP; Electrophoretic deposition

1. Introduction

Electrophoretic deposition (EPD) is a reliable, low cost method for producing laminates thicker than those attainable with chemical deposition techniques, and is suitable for a wide range of compositions. Consequently, EPD is receiving increased attention for the fabrication of laminated and graded (step-graded or continuously graded) materials, either as self-supported materials or as coatings.

Several publications describe the experimental devices and parameters used to obtain laminated materials by EPD for either functional or structural applications.[1–21] Tables 1 and 2 review published papers dealing with fabrication of laminated and graded materials, respectively. In these tables, the formulation of the colloidal suspensions is shown, as well as the composition, geometry and dimensions of the resulting laminates.

Studies on EPD of laminated materials (Table 1) mostly focus on the determination of processing parameters involved in the preparation of alternating layers of Al_2O_3 and tetragonal ZrO_2, while those describing the fabrication of graded materials (Table 2) mainly deal with the improvements reached in structural properties such as toughness and hardness. The testing conditions and the mechanical properties determined on such layered structures are summarized in Table 3. In the most general case, characterization is performed in terms of Vickers indentation hardness and toughness and very little attention has been paid to bulk properties that would be strongly determined by the processing method, such as strength.

In a previous work,[22] the Weibull distribution of strength values, determined by three point bending with 20 mm span and a cross head rate of 0.5 mm/min, for thin (≈170 μm) 10 mm × 25 mm plates made on alumina with 5 vol.% of tetragonal zirconia (A95Z5) was found to be well described by a simple two parameter Weibull distribution, giving an average strength of ≈160 MPa, and a Weibull modulus around 3. In agreement with the low Weibull modulus, indicating

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doi:10.1016/j.jeurceramsoc.2004.10.018

Table 1
Laminated materials obtained by EPD

Reference	Laminated materials	Colloidal suspension formulation		Final material characteristics		
		Media	Additives	Geometry	Laminated thickness	Layers thickness (μm)
Sarkar et al.[1]	Al_2O_3/Y-TZP	Ethanol	HCl (pH < 5)	Planar	1.5 mm (80 layers)	2–1
Sarkar et al.[2]	Al_2O_3/Y-TZP	Ethanol	HCl (pH < 5)	Radial	350 μm (36 layers)	
Bissinger et al.[3]	ZTA/Y-TZP/Laz	Ethanol	HCl (pH < 5)	Planar	2.5 mm (>100 layers)	2–30
Sarkar et al.[4]	Y-TZP/Al_2O_3	Ethanol	HCl (pH < 5)	Planar		13–21
Fisher et al.[5]	Al_2O_3/Y-TZP	H_2O		Planar		8–20
Vandeperre et al.[6]	SiC/C	*n*-Butylamine + 5% acetone + 20% isopropanol		Radial		10–100
Ferrari et al.[7]	Al_2O_3/Y-TZP	H_2O	Dolapix CE64	Planar	10 layers	25–50
Zhitomirsky and Gal-Or[8]	Al_2O_3/Ce-TZP	Isopropanol		Radial	20 layers	1–10

Table 2
Graded materials obtained by EPD

Reference	Graded materials	Colloidal suspension		Material characteristics		
		Media	Additives	Geometry	Laminated thickness	Layers thickness
Sarkar et al.[9]	Al_2O_3/Y-TZP	Ethanol	Acetic acid	Planar	6 mm	
Ding et al.[10]	Ni/Al_2O_3	Ni^{2+} aqueous solution				
	Cu/Al_2O_3	Cu^{2+} aqueous solution				
Merk[11]	Ni/SiC	Ni^{2+} aqueous solution				
Barmak et al.[12]	Al_2O_3/Al/Ni	Ni^{2+} aqueous solution		Planar	110 μm	20–40 μm
Sarkar et al.[13]	Al_2O_3/Ni	Ethanol	pH < 5	Planar		
	Y-TZP/Ni					
	Al_2O_3/$MoSi_2$					
Zhao et al.[14]	Y-TZP/Ce-TZP	*n*-Butylamine	PVA	Planar	1.8 mm	
Zhao et al.[15]	Al_2O_3/Ce-TZP	*n*-Butylamine + acetone	PVA	Radial	3 mm	
Börner and Herbig[16]	58AZ/48AZ	H_2O	Dolapix PC21	Planar	2.3 mm	
Put et al.[17]	WC–6% Co/WC–25% Co	*n*-Butylamine + acetone		Planar	2.3 mm	
Put et al.[18]	Y-TZP/WC	*n*-Butylamine + acetone		Planar	2 mm	
Vleugels et al.[19]	Al_2O_3/Y-TZP/Al_2O_3	*n*-Butylamine + acetone		Planar	5 mm	
Kaya[20]	Al_2O_3/Y-TZP	H_2O	Celor	Radial		Nanostructure

Table 3
Properties of laminates produced by EPD

References	Materials	Thickness	Mechanical test conditions	Mechanical properties
Vanderperre et al.[6]	SiC/C	1–2 mm (125 cm^2)	3 points bending, 0.05 mm/s charge rate, 20 mm span	Crack deflection
Prakash et al.[2]	Al_2O_3/Y-TZP	–	Microindentation Vickers (8 kgf load)	Crack deflection
			4 points bending	95% Y-TZP: 995 MPa, 75 kJ/m^2; 95% Al_2O_3: 488 MPa, 17.8 kJ/m^2
Sarkar et al.[9]	Al_2O_3/Y-TZP	6 mm	Vickers microindentation (3 kgf load)	$H_V = 26 - 15$ GPa; $K_{IC} = 2.5$–10 MPa $m^{1/2}$
Ding et al.[10]	Ni/Al_2O_3, Cu/Al_2O_3	–	Vickers microindentation (0.05 kgf load)	H_V, increases with a Ni matrix
			Pin-on-disc	Increases wear and adhesion
Zhao et al.[14]	Y-TZP/Ce-TZP	2.1 mm (9 cm^2)	Vickers microindentation (5 kgf load)	$H_V = 12.6 - 10.3$ GPa; $K_{IC} = 3.4$–10.2 MPa $m^{1/2}$
Zhao et al.[15]	Al_2O_3/Ce-TZP	1 mm (tubular)	Vickers microindentation (5 kgf load)	$H_V = 14.5 - 10.5$ GPa; $K_{IC} = 2$–10.2 MPa $m^{1/2}$
Put et al.[17]	WC–6% Co/WC–25% Co	1.5 mm (9 cm^2)	Vickers microindentation (0.5 and 10 kgf load)	$H_{V0.5} = 21 - 9$ GPa; $H_{V10} = 19 - 8.5$ GPa
Vleugels et al.[19]	Al_2O_3/Y-TZP/Al_2O_3	5 mm	X-ray diffraction, residual stresses	$\sigma(Al_2O_3) = 92 \pm 5$ MPa
Kaya et al.[20]	Al_2O_3/Y-TZP		Vickers microindentation	$H_V = 19.4$–10.4 GPa; $K_{IC} = 3.1$–7.1 MPa $m^{1/2}$

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