Werkstoffanwendungen im Maschinenbau

Band 20

Stanley van Kempen

Characterization and Numerical Simulation of Constrained Sintering in Al_2O_3 and $MgAl_2O_4$ based Ceramic Laminated Composites

Institut für Werkstoffanwendungen im Maschinenbau

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Institut für Anwendungstechnik Pulvermetallurgie und Keramik an der RWTH Aachen e.V.

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Characterization and Numerical Simulation of Constrained Sintering in Al_2O_3 and $MgAl_2O_4$ based Ceramic Laminated Composites

Charakterisierung und Numerische Simulation des Co-Sinterns Al_2O_3 und $MgAl_2O_4$ basierten Keramische Mehrlagige Kompositwerkstoffe

Von der Fakultät für Maschinenwesen der Rheinisch-Westfälischen Technischen Hochschule Aachen zur Erlangung des akademischen Grades eines Doktors der Ingenieurwissenschaften genehmigte Dissertation

vorgelegt von

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Tag der mündlichen Prüfung: 27.04.2021

Werkstoffanwendungen im Maschinenbau
hrsg. von Prof. Dr.-Ing. Christoph Broeckmann

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Shaker Verlag
Düren 2021

Bibliographic information published by the Deutsche Nationalbibliothek
The Deutsche Nationalbibliothek lists this publication in the Deutsche Nationalbibliografie; detailed bibliographic data are available in the Internet at http://dnb.d-nb.de.

Zugl.: D 82 (Diss. RWTH Aachen University, 2021)

Printed in Germany.

ISBN 978-3-8440-8094-0
ISSN 2195-2981

Shaker Verlag GmbH • Am Langen Graben 15a • 52353 Düren
Phone: 0049/2421/99011-0 • Telefax: 0049/2421/99011-9
Internet: www.shaker.de • e-mail: info@shaker.de

Summary

Ceramics are versatile materials that are positively known for their thermal and chemical stability, hardness, and wear and corrosion resistance. In structural applications, however, they are mostly known for being unreliable due to a low fracture toughness. Ceramic laminated composites (CLCs) have been developed to improve these aspects of structural ceramic materials. Using co-sintering, tape-cast layers of different materials are combined to obtain highly reliable composites that can be tailored to their prospective application. These layers exhibit anisotropic shrinkage during sintering, due to alignment of non-spherical particles during tape-casting. Thermal expansion mismatch and sintering shrinkage differentials between the individual layers leads to residual stresses during firing. In turn, these may result in warping, delamination, or fracture during laminate manufacturing. In this work, a Shinagawa-based viscous sintering model is modified to accommodate anisotropic sintering as a means to predict deformation and residual stresses in co-sintered CLCs with Al_2O_3 and $MgAl_2O_4$ layers using numerical simulation. Different particle size distributions were used to produce tapes with axial shrinkages ranging between 6.9 - 20.4% and anisotropic shrinkage ranging between 0.1 - 3.7%. Input parameters for the model are determined using viscosimetry and optical dilatometry experiments conducted on individual layers and laminated samples. Furthermore, the particle orientation and orientation degree are characterized using SEM imaging and considered in the model. Validation of the model is based on curvature experiments on bilayer materials after sintering and *in-situ* on co-sintered bilayers using optical dilatometry. Furthermore, residual stress measurements are conducted on symmetric trilayers materials with varied materials, layer thickness, and process parameters using synchrotron radiation. The proposed model can accurately predict anisotropic shrinkage in materials with average grain sizes up to 1 μm. It loses prediction accuracy when the average particle size exceeds 5 μm which is caused by several numerically derived parameters not being valid for larger particle sizes. The deformation of bilayer laminates after sintering corresponds reasonably well with experiments, as well as the general behavior of the model in response to different processing parameters, such as sintering temperature, heating rate, and sintering time. Simulation of the residual stress distribution corresponds reasonably well with experiments. The stress magnitude is shown to be lower than that in experiments despite qualitative trends being predicted well. The interaction between layers in bi- and trilayer laminates is simulated well qualitatively for a large amount of layup permutations. This enables the user to evaluate the effects of differently stacked CLCs with relative ease. The necessary amount of experimentally determined parameters and the amount of numerically derived parameters were reduced to a minimum, which simplifies practical implementation of the model. For practical applications of ceramic laminated composites, the findings of the experiments show that it is highly plausible that the residual stresses in CLCs can be controlled by strategically selecting the involved materials based on their sintering shrinkage

behavior. Residual stresses could be varied approximately between -200 – +200 MPa. The joining method (adhesive, adhesive with ceramic filler, or thermal compression) used for individual layers can be used to further increase or mitigate the residual stresses. This will expectedly have a significant influence in the development of damage tolerant CLCs, as the residual stress state can be employed to control damage mechanisms in complex laminates.

Kurzfassung

Keramische Werkstoffe sind positiv bekannt für ihren thermischen und chemischen Stabilität, ihre Härte, sowie für ihre Verschleiß- und Korrosionsbeständigkeit. In Anwendungsfälle worin sie strukturell beansprucht werden gelten sie aufgrund ihren niedrigen Bruchzähigkeit allerdings als nicht-zuverlässig. Laminierte keramischen Kompositwerkstoffe bieten Auskunft, um diesen negativen Aspekt zu verbessern. Das sog. Co-sinterverfahren ermöglicht der Herstellung robuster Kompositwerkstoffe. Diese ermöglichen eine anwendungsorientierte Optimierung da sie aus diversen foliengegossenen Schichten unterschiedlicher Werkstoffe aufgebaut werden können. Während dem Sintern, schwinden die einzelnen Schichten auf anisotroper Weise aufgrund einer Ausrichtung der asphärischen Partikeln während dem Foliengieß-Prozess. Differenzen im Sinterschwindung und thermischer Ausdehnungskoeffizienten führen zu Eigenspannungen während dem Sintern. Die Eigenspannungen, führen zu Wölbung, Delamination oder Risse im Laminat. In dieser Arbeit wurde ein viskös Sintermodell, basierend auf ein sog. Shinagawa Modell, entwickelt. Das Modell erlaubt der Vorhersage des Verformungs- und Eigenspannungsverhaltens co-gesinterten keramischen Laminate aus Al2O3 und MgAl2O4 mit anisotropes Sinterschwindungsverhalten, mittels numerischer Simulation. Unterschiedlichen Partikelgrößenverteilungen wurden verwendet um Folien mit axialer Schwindung im Bereich 6.9 - 20.4% und anisotroper Schwindung im Bereich 0.1 - 3.7% zu erzeugen. Eingangsparametern für das Modell wurden ermittelt anhand Viskosimetrie und optische Dilatometrie-Experimente auf Einzelschichten und Laminate. Auch wurden die Orientierung und Orientierungsgrad der Partikel mithilfe REM Analyse ermittelt. Der Validierung des Models ist mit Krümmungsexperimente im optischen Dilatometer auf 2-Schicht-Laminate nach und während dem Sintern erfolgt. Außerdem, wurden Eigenspannungsmessungen an symmetrischen 3-Schicht-Laminate mit unterschiedlichen Werkstoffen, Schichtdicken und Herstellungsprozessparameter durchgeführt mithilfe von Synchrotron-Experimente. Das entwickelte Simulationsmodell ermöglicht eine akkurate Vorhersage des anisotropen Sinterns in Werkstoffe mit mittleren Korngrößen bis 1 μm. Der Vorhersagegenauigkeit wird geringer, sobald der mittlere Korngröße des simulierten Werkstoffs größer als 5 μm ist. Dies ist zurückzuführen auf verschiedenen numerisch abgeleiteten Parametern, die nur für ein gewissen Bereich der Korngrößen gültig sind. Die Verformung von 2-Schicht-Laminate nach dem Sintern, sowie das Verhalten des Modells in Abhängigkeit der gewählten Prozessparametern (wie z.B. Sintertemperatur, Heizrate und Zeit) entsprechen die Experimente. Dies gilt auch für die Verteilung der Eigenspannungen in gesinterten Laminaten. Der Größenordnung der Eigenspannungen ist, obwohl qualitativ genau simuliert, niedriger als in die Experimente. Der Interaktion zwischen einzelne Schichten in 2- und 3-Schicht-Laminate wurde qualitativ gut simuliert. Dies ermöglicht der Evaluierung der Herstellbarkeit unterschiedlicher Laminataufbauten. Der Anzahl notwendigen Parameters, und damit der Anzahl der notwendigen Experimente, für das Modell wurde minimiert was einen praktischen

Einsatz des Modells vereinfacht. Die Experimente haben die Wahrscheinlichkeit gezeigt, dass die Eigenspannungen der getestete keramische Kompositwerkstoffe kontrolliert werden können im Bereich 200 – +200 MPa. Weiterhin kann einen geeigneten Verbindungsmethode der Schichten zur Verstärkung/Schwächung der Eigenspannungen führen. Der Erwartung ist, dass dies ein signifikanter Einfluss auf der Entwicklung keramischer Laminatwerkstoffe hat, da die Eigenspannungen der Kontrolle über die Schädigungsmechanismen in komplexen Laminaten ermöglichen.

Acknowledgements

First and foremost, I would like to extend my gratitude towards Professor Christoph Broeckmann for his supervision and for giving me the opportunity to conduct my research at the IAPK and IWM institutes. Furthermore, I want to thank Prof. Olivier Guillon for being part of my exam committee.

Special thanks goes out to Alexander Bezold, for his guidance and for the insightful discussions we've had. He has motivated me to push my own boundaries and improve my capabilities throughout the course of this thesis, in projects, as well as in academics in general. I want to thank all the colleagues from the IWM and IAPK for their support and the input they provided for the research and the projects that we have worked on. In particular, I would like to thank Robert Mager, Vanessa Derichs, Wolfgang Kayser, Sebastian Riehm, Karl Burkamp, Pierre Uhl, Sammy Abraham, and Christian Schlieker for their unrelenting support with the development and manufacturing of experimental setups, the production of samples, and the characterization of all of the materials that we have worked with over the years. Furthermore, I would like to thank Felix Weber, Anh Ngoc Giang, Sree Sistla, Yuanbin Deng, and the other PhunRaum colleagues for their helpful discussions and input regarding everything relating to modeling and simulation, as well as for keeping the caffeine levels at an optimum at all times. There are many colleagues that I have not mentioned explicitly but please know that my thanks also goes out to you. I will always treasure your support and the good times we had after working hours.

I want to recognize the IAPK's project partners for their helpful discussions and for a pleasant and fruitful cooperation over the past years. My gratitude goes out to Professor Andreas Roosen, Professor Nahum Travitzky, Ruth Hammerbacher, Hannes Lorenz, Benjamin Dermeik, and Dr. Daniel Jakobsen of the University of Erlangen-Nuremberg for providing samples and experimental results used in this thesis. I also would like to thank Friederike Lange, Jochen Zwick, and their colleagues of Siemens A.G. for providing a great project, for all the support they have offered, and for their timing of project meetings.

I would like to express my thanks to my family and friends, whom I love dearly and have not seen nearly enough during the time I was working on this thesis. I especially want to thank my wife for all her patience and support over the past years.

Finally, I want to thank the HZB, in particular Dr. Manuela Klaus and Prof. Christoph Genzel for their support and for the allocation of the synchrotron radiation beam time. Furthermore, I acknowledge the financial support by the Federal Ministry for Economic Affairs and Energy of Germany in the project "Development of novel combustion techniques for the Clean Energy Center for environmentally friendly energy generation" (project numbers 03ET7011G, 03ET7011R, and 03ET7073J).

Aachen, April 2021 Stanley van Kempen

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Nomenclature

Symbols

α	Thermal expansion coefficient
$\Delta\alpha$	Thermal expansion coefficient differential
β	Slope of average grain growth
β_s	Sintering stress correction factor
γ	Surface tension
γ_{gb}	Grain boundary tension
δ	Deflection
$\dot{\delta}$	Deflection rate
δ_{ij}	Kronecker delta
ϵ_{cr}	Creep strain
ϵ_f	Free sintering shrinkage
ϵ_i^f	Free sintering shrinkage in i-direction
$\dot{\epsilon}_i^f$	Free sintering shrinkage rate in i-direction
ϵ_i	Strain in i-direction
ϵ_{ij}	Strain tensor
ϵ_{ij}^{vp}	Viscoplastic strain tensor
$\dot{\epsilon}_{ii}$	Trace of the strain rate tensor
$\epsilon_{s,N}$	Sintering shrinkage of the N-th layer
$\Delta\epsilon^{\alpha}$	Thermal shrinkage differential
$\Delta\epsilon^{\sigma}$	Sintering shrinkage differential
ζ_i	Equation specific adjustable parameter where i designates different adjustable parameters within the same function
η	Shear viscosity
η_0	Shear viscosity of the fully dense isotropic body
η_c	Viscosity constant
η_p	Shear viscosity of the porous isotropic body
η_{PSD}	Particle size dependent shear viscosity
η_T	Viscosity constant for exponential shear viscosity function
θ	Porosity
θ_d	Diffraction angle
κ	Curvature of laminate
$\dot{\kappa}$	Curvature rate of laminate
κ_{neck}	Neck curvature
λ	Thermal conductivity
μ	Mean
ν	Poisson's ratio
ν_0	Viscous Poisson's ratio of a fully dense isotropic body
ν_p	Viscous Poisson's ratio of a porous isotropic body
ν_{ij}^p	Viscous Poisson's ratio tensor of a porous body
ξ	Anisotropy factor
ξ_s	Stress induced anisotropy factor
ρ	Density

$\dot{\rho}$	Densification rate
ρ_r	Relative density
$\Delta\rho_r$	Change in relative density
Σ	Sintering potential
σ	Standard deviation
σ_b	Grain boundary stress
σ_i	Stress in the i-direction
σ'_{ij}	Deviatoric stress tensor
σ_m	Average (hydrostatic) stress
σ_r	Radial stress
σ_{res}	Residual stress
σ_s	Sintering stress
τ	Angle between flow velocity and rotational axis of spheroid
$\dot{\tau}$	Shear rate
τ_{ij}	Shear stress tensor
ϕ	Orientation angle
ϕ_f	Angle between major axis and shear direction
φ	Normalized shear viscosity
φ_a	Incidence angle
χ	Dihedral angle
χ_a	Inclination angle
ψ	Normalized uniaxial viscosity
Ω	Atomic volume
Ω_v	Vacancy volume
A	Surface contact area
$A_{2D,i}$	Grain area
A_{ii}	Anisotropy tensor
AR	Aspect ratio
a_i	Semi-axis i of an ellipsoid
B	Creep strain hardening coefficient
b	Sample width
C	Dimensionless constant
C_E	Correction factor for dynamic Young's modulus
c	Circumference of particle contact area
c_p	Specific heat
D_0	Frequency factor
D_b	Grain boundary diffusion coefficient
d	Grain diameter
d_0	Initial grain diameter
d_{eq}	Equivalent grain diameter
d_p	Particle diameter
E	Young's modulus
E_A	Young's modulus of Al_2O_3
E_S	Young's modulus of $MgAl_2O_4$
E_p	Uniaxial viscosity of the isotropic porous body
E_i^p	Uniaxial viscosity in the i-direction of a porous body

F	Force
F_s	Sintering force
F_v	Shape factor of mean spheroid
f_b	Bimodal probability density distribution
f_f	Flexural frequency
f_l	Log-normal probability density distribution
f_n	Normal probability density distribution
f_p	Particle frequency
G_0	Shear viscosity modulus of a fully dense isotropic body
G_p	Shear viscosity modulus of a porous isotropic body
$\overline{G_p}$	Effective shear viscosity modulus of a porous isotropic body
g	Gravitational constant
h	Thickness/height
h_{gb}	Effective grain boundary width
K	Grain orientation degree
K_0	Bulk viscosity modulus of a fully dense isotropic body
K_p	Bulk viscosity modulus of a porous isotropic body
$\overline{K_p}$	Effective bulk viscosity modulus of a porous isotropic body
k	Boltzmann's constant
L	Length
l	Laminate length
M	Proportionality constant for grain boundary mobility
M_{gb}	Grain boundary mobility
$M_{gb,0}$	Grain boundary mobility coefficient
m	Mass
N	Number of layers
n	Grain growth exponent
P	Laplacian pressure
Δp	Gas pressure in closed pores
Q	Activation energy for viscous flow
Q_d	Activation energy for grain boundary diffusion
Q_T	Activation energy for exponential viscous flow function
R	Universal gas constant
R_i	Average particle size group radius
R_C	Radius of curvature
$\dot{r}$	Grain growth rate
r_b	Reference particle radius
r_i	Mean particle radius of i th particle
r_c	Instantaneous critical radius with zero growth
r_{cr}	Critical grain radius
r_{eq}	Equivalent particle radius
r_p	Particle radius
r_0	Initial grain radius
r_{pore}	Pore radius
$r_{pore,0}$	Initial pore radius
S	Solubility of particle with infinite radius

T	Temperature
T_{sint}	Sintering temperature
T_{start}	Sintering start temperature
t	Time
t_{green}	Layer thickness in green state
t_i	Thickness of the ith layer
t_{sint}	Layer thickness after sintering
u	Exponential constant for viscous moduli
v	Numerically derived viscosity parameter
W	Exponential constant for sintering stress
w	Numerically derived viscosity exponent
z	Axial position

Abbreviations

AR	Aspect ratio
CLC	Ceramic laminated composite
CS	Co-sintering
EBSD	Electron back scatter diffraction
FE	Finite element
FEM	Finite element method
LTCC	Low temperature co-fired ceramic
SEM	Scanning electron microscope
TC	Thermal compression