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Thermal Shock Resistance of Ceramic Materials in Melt Immersion Tests*

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Abstract

A melt immersion test is applied to determine the relative resistance of ceramic materials to thermal shock failure under high heat flux conditions. The testing method is demonstrated mainly for Al_2O_3 pellets, while AlN is included to represent elevated thermal shock resistance. In order to quantify the resistance to crack formation, the critical temperature difference ΔT_c between sample and metal melt is determined from the failure probability distribution of a set of pellets.

In quenching tests correspondence of ΔT_c with the thermal shock parameter $R = \sigma(1-\mu)/E\alpha$ was found, if the initial surface temperature of the sample was correctly estimated. This assessment was the main concern of the evaluation work.

ΔT_c resulting from heating tests was correlated with the maximum tensile stress in the sample by modeling calculations. The stress limits determined show that the ultimate bending strength could serve as a rough approximation for the materials tested.

Zur Untersuchung der relativen Widerstandsfähigkeit verschiedener keramischer Materialien auf Versagen durch Thermoschock bei hohem Wärmeeinfluß wurde ein Schmelzeintauchtest angewendet. Die Testmethode wird hauptsächlich mit Al_2O_3-Pellets durchgeführt, während AlN verwendet wird, um ein Material mit einem besseren Thermoschockwiderstand darzustellen. Um die Widerstandsfähigkeit gegen Rißbildung in Zahlen auszudrücken, wurde die kritische Temperaturdifferenz ΔT_c zwischen Probe und Schmelze über die Versagenswahrscheinlichkeit bestimmt (jeweils mit einer Reihe von jeder Pelletsorte).

Bei den Abschreckversuchen stimmte ΔT_c mit dem Thermoschockparameter $R = \sigma(1-\mu)/E\alpha$ überein, wenn die Ausgangstemperatur der Probenoberflächen richtig errechnet worden war. Diese Abschätzung war ein Hauptziel dieser Untersuchungen.

Die aus den Aufheiztests resultierende kritische Temperaturdifferenz wurde zu Modellrechnungen mit den maximalen Zugspannungen in Beziehung gesetzt. Die bestimmten Spannungsgrenzen zeigen, daß die maximale Biegefestigkeit der getesteten Materialien als erste ungefähre Näherung dienen kann.

Un test d'immersion dans une masse fondue est utilisé pour déterminer la résistance relative de matériaux céramiques au choc thermique sous flux de chaleur élevé. La technique d'essai est présentée principalement sur des pastilles d'Al_2O_3, tandis qu'AlN est utilisé pour montrer l'effet d'une résistance élevée au choc thermique. Pour quantifier la résistance à la formation de fissures, la différence critique de température ΔT_c entre l'échantillon et le métal fondu est déterminée à partir de la distribution des probabilités de fracture d'un lot de pastilles.

Dans les essais de trempe, on a trouvé une correspondance entre ΔT_c et le paramètre de choc thermique $R = \sigma(1-\mu)/E\alpha$, si la température initiale à la surface de l'échantillon était estimée correctement. Cette estimation était l'un des objectifs principaux du travail d'évaluation.

Par des calculs de modélisation, on a corrélé la ΔT_c résultant des essais par chauffage à la tension maximale dans l'échantillon. Les tensions limites déterminées montrent que la valeur de résistance en flexion peut servir d'approximation grossière pour les matériaux examinés.

* This article is based on investigations by H. Scholz, conducted as part of a Master's degree programme in mechanical engineering at the University of Karlsruhe.

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1 Introduction

Resistance to thermal crack formation is often the decisive demand on ceramic materials applied in mechanical engineering. This is especially true for nuclear fusion reactors in view of the high heat fluxes to ceramic protection plates on fusion plasma-facing components (~ 100–$1000\,W/cm^2$), and also of the high radiofrequency energy fluxes in auxiliary systems for plasma heating (5–$70\,kW/cm^2$). Ceramic windows or supports in the RF transmission lines are subject to considerable heating by dielectric loss, increasing with frequency. High heat power density $\geq 100\,W/cm^3$ (up to about $500\,W/cm^3$) can occur, particularly at the maximum frequency for plasma heating at about 100 GHz.

For both applications in fusion reactors, the crucial heat load occurs under stationary operating conditions. Since it is a rather complex and expensive task to install, to perform and to analyse steady-state heat flux tests at high heat loads, an urgent interest developed concerning a more easily feasible testing method. It should be appropriate to compare the resistance to thermal crack failure of different ceramic materials, and particularly of neutron-irradiated samples with unirradiated ones. The latter is the main concern, because fusion reactor operation involves considerable neutron effects on the ceramic components mentioned whereby, moreover, the test equipment has to be operated under hot cell space and handling conditions.

The testing method chosen is the immersion of cylindrical pellets into a metal melt, which was assumed to guarantee a short-term heat flux density of sufficient magnitude compared to the operating conditions, together with suitably high thermal stresses. Moreover, the sample shape and arrangement facilitates the analysis of temperature and stress distribution. Concerning the applicability of the experimental results, the testing procedure has the merit to cover not only crack initiation but also crack propagation in the thermal stress gradient, which is a decisive advantage compared to the determination of a 'thermal shock parameter' merely calculated from ultimate bending strength.

Of course, the testing method is also suitable, within certain limits, to simulate actual thermal shock events under technical operating conditions. But the present task is the comparison and ranking of ceramic materials with respect to their susceptibility to thermal crack failure. The 'critical' difference between the initial temperatures of the metal melt and the specimen of a certain shape, at which crack formation starts appearing, is determined to characterize the relative thermal shock resistance of different materials. In order to cover a rather large range of the thermal shock resistance of ceramic insulator materials, both Al_2O_3 and AlN samples were included in the demonstration of the melt immersion test method. It is exemplified by the measurements on Al_2O_3, while the less close results on AlN are not presented in detail but are only used for evaluation and comparison, also with modeling calculations.

2 Experimental procedure

The specimens used in the thermal shock tests were cylindrical pellets of Al_2O_3 of moderate strength and of isostatically hot-pressed AlN (diameter: 10 mm, height: 10 mm). The important material data are listed in Table 1. Quenching from a higher temperature as well as rapid heating was performed by immersion in a liquid metal bath. The experimental procedure is fully described in Ref. 1. Liquid tin has been generally used up to 900°C, while a eutectic silver–copper melt was tried for higher temperatures. In order to guarantee an exclusively radial heat flux, axial heat exchange from the pellet specimens was prevented by insulator pellets above and below. The thermal shock was produced by dipping pellet specimens into the metal melt, both at controlled temperature. A metal melt has been chosen as a heat transfer medium, because it allows both quenching and heating tests and guarantees a strong thermal shock. The severity of a thermal shock generally can be characterized by the heat transfer coefficient between sample and heat transfer medium. In the melt immersion tests the heat transfer coefficients range from 1 to $5\,W/cm^2\,K$ and, in contrast to water quenching, are nearly constant over a wide range of test conditions, because no boiling occurs. According as to whether crack

Table 1. Material properties

	Al_2O_3	AlN	Melts[a]	
			Sn	Ag–Cu
Density (% TD)	98·3	98·9		
Thermal conductivity (at RT) (W/cm K)	0·34	0·76	0·334	1·33
Coefficient of linear thermal expansion (20–1 000°C) (1/K)	$8{\cdot}2 \times 10^{-6}$	$5{\cdot}5 \times 10^{-6}$		
Ultimate bending strength (MPa)	205 ± 25	273 ± 35		
Weibull modulus	8·5	8·3		
Young's modulus (GPa)	378	322		
Poisson's ratio	0·25	0·23		

[a] At all bath temperatures.

Table 2. Critical conditions and parameters for thermal shock resistance

Material		ΔT_c (*K*)	*R* (*K*)	*R** (*K*)	$\sigma_{t,max}$ (*MPa*)
Al_2O_3	Heating from 23°C in Sn	485			183
	Quenching to 260°C in Sn	105	50	102	186
AlN	Heating from 60°C in Ag–Cu	1 300			462
	Quenching to 250°C in Sn	318	119	311	313

formation is observed or not, the temperature difference in the subsequent test is reduced or increased. In heating tests the specimens started from room temperature in each case, while in quenching tests the melt temperature was kept at a constant level and the specimen temperature was varied by preheating in a small tube furnace above the melt crucible. After each test the cracks that could have formed in the specimen were traced with a fluorescent penetrating liquid. In heating tests crack formation could also be indicated acoustically.

3 Results

The test results are shown in Table 2. The critical temperature differences given correspond to the first appearance of cracks in each case. The distribution of individual critical temperature differences in Al_2O_3 specimen sets is demonstrated in Figs 1 and 2. Critical temperature differences are considerably higher in heating tests than in quenching tests. This is due to the fact that the compressive strength of ceramic materials is much greater than their tensile strength. On quenching maximum tensile stresses occur at the specimen surface. Additionally, surface flaws could favour crack initiation and further reduce the critical temperature difference. On the other hand, rapid heating will produce compressive stresses at the specimen surface, while crack initiation by tensile stresses is shifted to the centre of the specimen. Therefore this test procedure is better

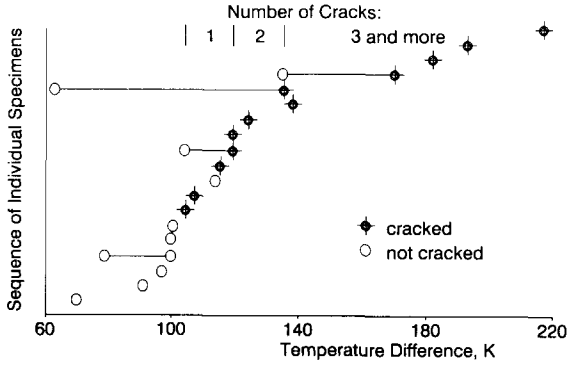

Fig. 1. Distribution of the failure resistance of Al_2O_3 pellets in quenching tests, represented by a critical temperature difference.

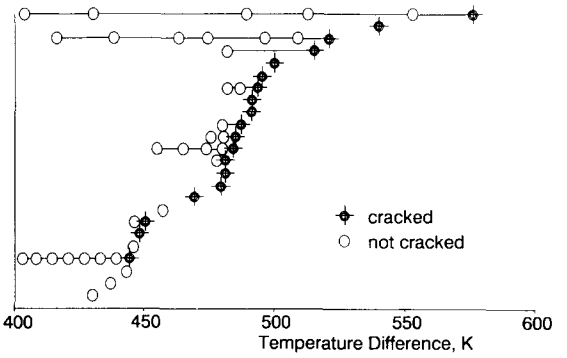

Fig. 2. Distribution of the failure resistance of Al_2O_3 pellets in heating tests, represented by a critical temperature difference.

suited to characterize and to compare material quality as such.

In the heating tests, a crack developed radially from the specimen centre at the critical temperature difference in Al_2O_3, often with branching near the surface. Fragmentation occurred with increasing temperature difference. Preceding subcritical thermal shocks neither changed the crack appearance nor the critical temperature difference. On the other hand, subcritical thermal shocks on AlN specimens resulted in reduced critical temperature differences and large fragmentation in subsequent tests.

In the quenching tests on Al_2O_3 specimens a radial microcrack developed from the surface at the critical temperature difference. With increasing temperature difference more and more cracks appeared, also in branching configuration (Fig. 3). In AlN

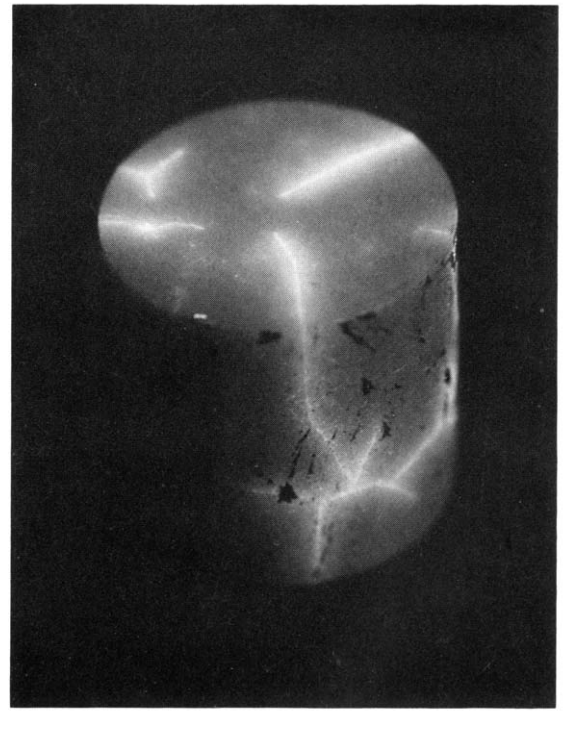

Fig. 3. Crack pattern in a quenched Al_2O_3 pellet observed shortly above the critical temperature difference.

specimens the developing crack was considerably longer. Table 2 shows that the thermal shock resistance of AlN is much better than that of Al_2O_3, as expected from the property data in Table 1, particularly concerning thermal conductivity.

4 Evaluation

4.1 Quenching tests

Thermal stress resistance parameters are now usually used to give an estimation of the behaviour of ceramic materials under thermal load, based on the knowledge of relevant physical and mechanical properties. A comprehensive review of various parameters was given by Hasselman.[2] Well-known parameter definitions are:

$$R = \frac{\sigma(1-\mu)}{E\alpha} \tag{1}$$

and

$$R' = \frac{\sigma(1-\mu)}{E\alpha} k \tag{2}$$

where σ = tensile strength
μ = Poisson's ratio
E = Young's modulus
α = thermal expansion coefficient
k = thermal conductivity.

R is in direct correlation with the critical temperature difference on quenching; its dimension is K. On the other hand, R' relates to the case of steady-state heat flux, which is far from the present test conditions.

R was derived from Hooke's law with regard to thermal expansion. This parameter gives the maximum tolerable temperature difference for bodies of simple shape with two-dimensional stress. Immediate application to quenching processes would require that the heat transfer is infinite. In the second column of Table 2, R is given to be compared with the critical temperature differences measured on quenching. R is low by a factor 2 (Al_2O_3) or 2·7 (AlN). Similar results were reported by Lewis[3] mainly for ceramics free of glass phase.

This is due to the fact that only the actual temperature difference within the specimen is effective in developing thermal stresses. Therefore a modified thermal stress parameter is recommended to be used for static media, where heat transfer exclusively depends on heat conduction. The modification proceeds from an expression for the contact temperature, which is well known in heat exchanger design:[4]

$$T_K = \frac{T_{mat} + \frac{\sqrt{\lambda\rho c_{med}}}{\sqrt{\lambda\rho c_{mat}}} T_{med}}{1 + \frac{\sqrt{\lambda\rho c_{med}}}{\sqrt{\lambda\rho c_{mat}}}} \tag{3}$$

T_K gives the surface temperature of the material dipped into the liquid medium, which is instantly adjusted by transient heat conduction, provided there is good wettability without boiling. From $\Delta T_c = T_{mat,e} - T_{med}$, and $R = T_{mat,c} - T_{K,c}$ one can derive:

$$\Delta T_c = R\left(1 + \frac{\sqrt{\lambda\rho c_{mat}}}{\sqrt{\lambda\rho c_{med}}}\right) = R^* \tag{4}$$

R^* is the modified parameter which gives a direct estimation of ΔT_c. This is technically applicable information which allows one to avoid extensive approximation for the heat transfer coefficient. It can be applied also to curved (such as cylindrical or spherical) surfaces, if the penetration depth of the temperature change is much smaller than the radius of curvature. R^* data calculated for the present materials are given in the third column of Table 2. Indeed satisfactory agreement can be stated with the measured ΔT_c data.

4.2 Heating tests

In the evaluation of the heating tests a computer program was applied to approximate the thermal and mechanical state in an infinitely long cylinder of a homogeneous, perfectly elastic material. The code is written in the programming language APL and uses temperature dependent material properties. First of all the temperature distribution in a cylinder with radial heat flux is calculated by solving the differential equation

$$\frac{1}{r}\frac{d}{dr}\left\{\lambda(T)\frac{dT}{dr}\right\} = c_p(T)\rho(T)\frac{dT}{dt} \tag{5}$$

where r = radius
λ = thermal conductivity
T = temperature
c_p = specific heat
ρ = density.

The numerical solution is calculated by the finite difference method. The system pellet-bath is subdivided into annular rings of equal width, usually 50 rings are used, 10 in the pellet and 40 in the bath. Boundary conditions are the surface temperature of the system, which is kept constant at the initial

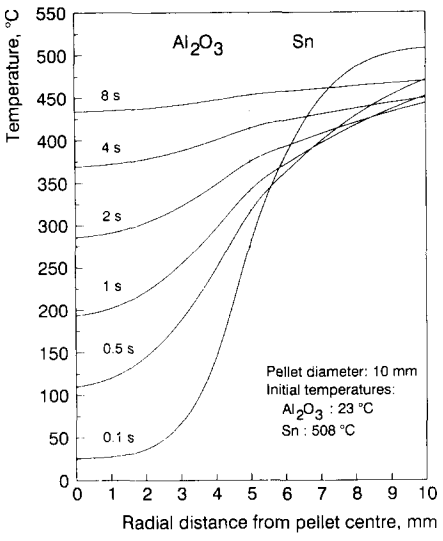

Fig. 4. Temperature in an Al_2O_3 pellet and in the tin bath at different times as a function of the radius.

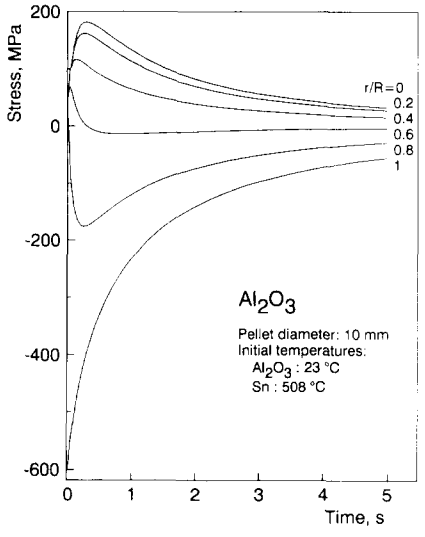

Fig. 5. Tangential stresses in an Al_2O_3 pellet at different distances from the centre as a function of time.

temperature of the bath, and the fact that the temperature gradient in the centre of the pellet is zero. Computation starts with the contact temperature T_K defined in Section 4.1. It is assumed that the surface of the pellet takes this temperature immediately. As an example Fig. 4 shows the distribution of temperature in an Al_2O_3 pellet and in the tin bath at different times.

The thermal stresses (plane strain) in the circular cylinder of infinite length are given by[5]

$$\sigma_\theta(r) = \alpha E/(1-\mu)\{f(R) + f(r) - T(r)\} \quad (6a)$$

$$\sigma_r(r) = \alpha E/(1-\mu)\{f(R) + f(r)\} \quad (6b)$$

$$\sigma_z(r) = \alpha E/(1-\mu)\{2f(R) - T(r)\} \quad (6c)$$

with

$$f(r) = \frac{1}{r^2}\int_0^r T(r) r \, dr \quad (7)$$

where σ_θ = tangential stress
σ_r = radial stress
σ_z = axial stress
α = thermal expansion coefficient
μ = Poisson ratio
E = Young's modulus
R = radius of the cylinder.

The temperature dependence of the material properties is completely taken into account by including the prefactor $\alpha E/(1-\mu)$ in the integrals f of eqns 6(a)–(6c). Figure 5 illustrates the time dependence of the tangential stresses in an Al_2O_3 pellet at different distances from the centre. The maximum tensile stress produced in a cylinder by heating occurs in the centre.

The maximum tensile stresses $\sigma_{t,max}$ which develop at the pellet centre line in heating tests were calculated and are given in the last column of Table 2, together with those at the pellet surface in quenching tests. Comparison with Table 1 shows that $\sigma_{t,max}$ of Al_2O_3 comes into the lower part of the scattering range of the ultimate bending strength. This is in accordance with corresponding results on other oxide ceramic materials.

On the other hand, $\sigma_{t,max}$ calculated for AlN is rather high in the heating test which may be due to the uncertain thermal conductivity used for the Ag–Cu melt. $\sigma_{t,max}$ in quenching is much less sensitive in this respect, but could be influenced by the increased density and, probably, improved strength in a surface layer of this sample material.

The experiments indicated that the ultimate bending strength is a suitable strength criterion for thermal shock resistance. A conservative stress limit seemed to be about 80% of the mean ultimate bending strength. But in respect of the volume dependence of strength, this specification is thought to be of minor importance.

5 Conclusions

An approximation for the initial surface temperature of solid materials on quenching rendered the thermal shock parameter R technically applicable. A modified version R^* allows one to estimate the maximum tolerable temperature difference between material and cooling medium. In the case of boiling or lacking wettability the estimation remains conservative. On the other hand, considerable convection, if occurring, has to be included in the determination of R^*.

The recommended approximation for the initial surface temperature is also useful to facilitate the calculation of critical conditions on the heating of ceramic components.

Both for quenching and heating of pellet specimens of the materials tested, the ultimate bending strength was found to present an appropriate stress limit in respect of crack formation.

Acknowledgement

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