HIGH AND MEDIUM FREQUENCY NON DESTRUCTIVE TESTING OF THE THERMAL SHOCK RESISTANCE OF CERAMICS

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ABSTRACT

The increasing use of ceramics as components for high temperature applications calls for an improvement of their thermomechanical properties ,chiefly the thermal shock resistance. Both non destructive and destructive tests are developed . Three ceramic materials ($A\ell_2O_3$, Si_2N_2O, $A\ell N$) are tested, three geometries of samples (long cylinders, short cylinders and discs) are used and the cumulative effects of shocks are studied. Results are discussed and some reservations are made about the practical applicability of the theories.

KEYWORDS

Thermal shocks - Non destructive tests - Discs - Alumina - Aluminium nitride - Silicon oxynitride.

INTRODUCTION

In spite of their typical brittleness, ceramic materials have always been prefered to metals for high temperature applications because of their good refractoriness. Such components are submitted to severe thermomechanical constraints uncompatible with their bad thermal and mechanical resistance. This behaviour results from the fact that no plasticity can relax the stresses and a only single mechanism can dissipate the elastic energy stored during the shock : the creation of new surfaces by nucleation and propagation of cracks (Gupta, 1973). A great deal of papers has been devoted to thermal shocks and two theoretical approaches are possible : the thermoelastic analysis (Kingery, 1955) which leads to the estimation of the critical temperature at which the cracks nucleate, and the energetic analysis (Hasselman, 1969) which gives both the previous critical temperature and the degree of damage. So, two concepts about thermal shock resistance are introduced (Nakayama, 1966) : the fracture resistance and the damage resistance to thermal shocks ,the relative importance of which depends both on the material and on the use of each component (Davidge, 1967). However, these analyses are based on simplified assumptions and involve parameters numerically badly known. Thus, an accurate characterization of the thermal shock resistance of materials, always requires experiments.

EXPERIMENTAL DATA

Thermal Shock Resistance

Several experimental methods for studying the thermal shock resistance are known: static (destructive) methods such as measurement of spalling (A.S.T.M., 1958), measurement of loss of weight (Larson, 1974), measurement of loss of strength (Gupta, 1972) ; dynamic (non destructive) methods, which follow the changes of frequency and attenuation values of travelling (Boisson, 1974), or standing waves (Ainsworth, 1968). The acoustic emission technique permits original measurements of the time to failure under thermal stresses and of the value of the heat transfer coefficient (Evans, 1975). Moreover, the dye penetrant method enables us to observe the extent of cracking (Gupta, 1973). The measurement of the strength after thermal shock is the most classical method described in the literature. In the case of thermal shocks by cooling (quench), a great number of materials show a typical discontinuity in the σ_f versus ΔT plots at a critical value of ΔT as shown in Fig. 1. The main advantage of this method is that it informs both on the fracture resistance (critical temperature ΔT_c) and on the damage resistance (ratio σ'_f/σ_f) ; thus it may be considered as a reference method. Its major disadvantage is to be a destructive test which does not allow the study of cumulative shocks and calls for numerous samples,even if experimenter takes care to avoid redondant data (Seaton, 1973). Another disadvantage is that it links the thermal shock resistance to a mechanical parameter (σ_f), which is scattered in brittle solids (Davidge, 1979). Due to this scatter, it leads sometimes to experimental values of delicate fitting (Ziegler, 1979).

Experimental Methods, Materials and Samples

The present work has developed non destructive techniques both for use few numerous samples and for study the occasional effects of cumulative shocks. Besides, it has been attempted to develop experimental methods, the applications of which to the study of thermal shock resistance have not been mentioned in the literature. In this frame, two dynamic methods have been developed : (1) the first in the kHz range uses longitudinal vibrations of bars (Glandus, 1974) and flexural vibrations of discs (Ryll Nardzewski, 1975), (2) the second in the MHz range uses ultra sonic waves according to the Pulse Echo Overlap Technique (Gault, 1977). Destructive tests for the measurement of the strength after thermal shock by 3 point bending for bars and by biaxial flexure for discs (Wachtman, 1972) have also been performed. The extent of cracking has been followed by a decorative technique.

The originality of the present work resides in the following points : (1) For M.F. techniques, the literature describes flexural vibrations of bars only. In the present work the use of longitudinal vibrations has been prefered for such a geometry because it does not involve a thermoelastic contribution in measurements of the damping capacity. Moreover flexural vibrations of discs (Glandus, 1979 a) have been used, and the interest of such a geometry for ceramic materials has been underlined elsewhere (Glandus, 1980 a). (2) The H.F techniques have not been (or very rarely) used by other workers for the study of the thermal shock resistance. (3) For the classical measurement of the strength after shock, the literature reports only 3 point and 4 point bending tests, but never biaxial flexure tests. (4) The three techniques (static, dynamic M.F., dynamic H.F.) have been developed simultaneously and cumulative shocks have been performed and followed by non destructive tests.

The methods previously described have been applied to three dense ceramics : polycrystalline α alumina of commercial quality (DEGUSSIT Aℓ 23), aluminium nitride (AℓN) hot pressed without additive by the E.R.A.-C.N.R.S. N° 539 Laboratory

(Lecompte, 1979), silicon oxynitride (Si_2N_2O) hot pressed with 5 % MgO (Wt) as additive by the E.R.A.-C.N.R.S. N° 539 Laboratory (Mary, 1974).

Three geometries have been used : long cylinders (Ø 6 × ℓ 80), short cylinders (Ø 20 × 20), discs (Ø 30 × 3) and the experimental techniques used are listed in the Table 1.

TABLE 1 Experimental Tests Versus Shape of Samples (C = cumulative tests - N.C. = non cumulative tests).

Tests / Shape	Destructive		Non Destructive	
	3 point bending	Biaxial flexure	M.F.	H.F.
Long Cylinders	N.C.	/	C 20 kHz	/
Discs	/	N.C.	C and N.C. 10-40 kHz	/
Short Cylinders	/	/	/	C 6-10 MHz

Thermal Shocks and Measurements

The samples were first annealed for 1 hour in a furnace previously heated to the temperature T, then quenched in a bath of water hold at room temperature ($\Delta T \simeq T - 20°C$). They were agitated during a few minutes in this bath and dried for 15 mn at 110°C. The details of the testing methods used in the present work have been described in previous papers (Glandus, 1979 b), (Glandus, 1980 b). For destructive tests, the strength (3 point bending for bars, biaxial flexure for discs) have been measured after the thermal shock cycle. For the M.F. dynamic method the relative frequency ratio ($\Delta N/N_o$), the frequency ratio for the two first overtones (N_2/N_1) and the damping (Q^{-1}) - or the ratio ($\Delta Q^{-1}/Q^{-1}$) - have been measured after thermal shocks for cumulative tests and the same values have been measured before and after thermal shocks for non cumulative tests. For the H.F. dynamic method the velocity of longitudinal and shear waves (V_L and V_S) and the acoustic attenuation (α) have been measured after thermal shock.

RESULTS AND DISCUSSION

The results obtained for silicon oxynitride have been published elsewhere (Glandus, 1979 b), (Glandus, 1980 c), as well as those for aluminium nitride (Glandus, 1980 b), and for alumina (Glandus, 1980 d). Fig. 2 to 5 are given as examples.

It may be noted that the Fig. 4 has not the typical aspect of the Fig. 1, but it is very close to the one obtained by Ainsworth (1968) under the same experimental conditions but for another commercial polycristalline alumina.

The critical quenching temperature(ΔT_c), are listed in Table 2.

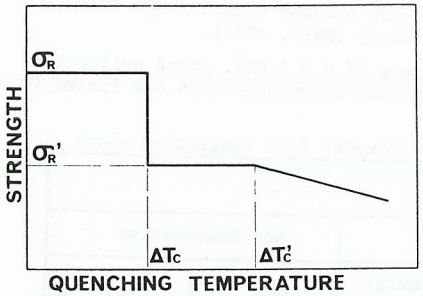

Fig. 1 Typical variations of strength vs ΔT.

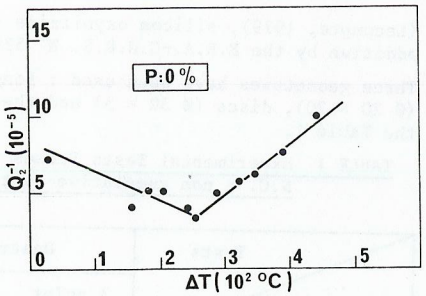

Fig. 2 Damping vs ΔT. Disc AℓN. Cumulative shocks.

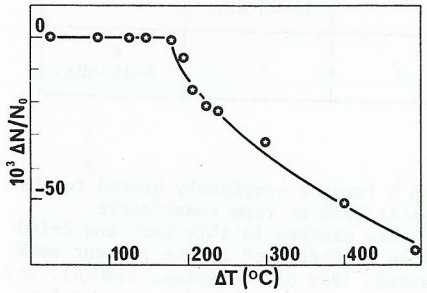

Fig. 3 Relative frequency ratio vs ΔT. Long cylinder $A\ell_2O_3$. Cumulative shocks.

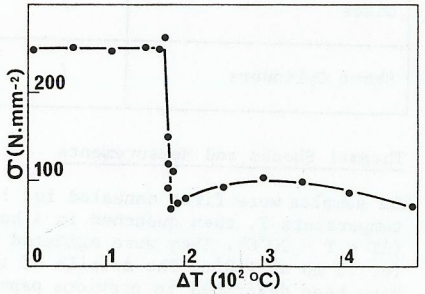

Fig. 4 3 point bending strength vs ΔT. Long cylinders $A\ell_2O_3$. Non cumulative shocks.

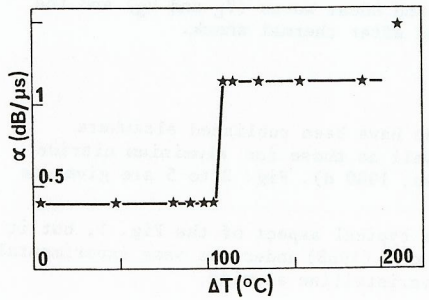

Fig. 5 Acoustic attenuation vs ΔT. Short cylinder $A\ell_2O_3$. Cumulative shocks.

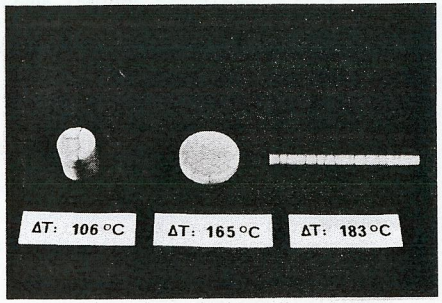

Fig. 6 Dye penetrant observations. $A\ell_2O_3$ samples.

TABLE 2 Critical Quenching Temperature versus Sample Shape, Materials and Experimental tests.

Shape	Methods		ΔT_c (°C)		
			$A\ell_2O_3$	Si_2N_2O	Aℓn
Long Cylinders Ø 6 X 80	N.C.	3 point bending	175	/	/
	C	M.F.	180	/	/
Discs Ø 30 X 4,5	N.C.	Biaxial flexure	170	390	/
	C	M.F.	165	395	/
	C	M.F.	180	380	155
Short Cylinders Ø 20 X 20	C	H.F.	105	240	140

Whatever the test, the ΔT_c values for a given material and a given geometry are in good agreement. The very good agreement between the values relative to destructive and non destructive tests (discs tested in biaxial flexure and in vibration of plates) indicates the validity of the non destructive tests here developed. The good agreement between values for cumulative and non cumulative tests shows that, in the present experimental conditions, the cumulative effects of shocks (Thermal fatigue) are negligible. For all the materials, it may be noted a size effect : the more voluminous the sample (cylinders), the less the critical quenching temperature.

The qualitative observations by dye penetrant technique are in good agreement with the quantitative results as illustrated in Fig. 6

In the case of medium frequency measurements, contrary to some bibliographical reports (Nakayama, 1966), (Ainsworth, 1968) ,the variations of the resonant frequencies are not due to variations of the elastic moduli (E and G) of the material, but to a loss of continuity of the cracked sample : (1) Dynamic measurements at high frequency have proved (Glandus, 1979 b) that the elastic moduli are hardly sensitive to the thermal shock severity. This fact has been checked for several materials (Boisson,1974),and the results obtained by using two ultra sonic methods (Gault, 1977) - Pulse Echo Overlap and Phase Comparison - allow the same conclusion. (2) Elastic constant are intrinsic properties of materials, directly linked to the interatomic potentials and it seems logic that they do not depend on macroscopic flaws. It is thus possible to understand the difficulties encountered by other authors (Hasselman, 1970 a) in their attempt to characterize the E = f (ΔT)function.

At a macroscopic scale, the variations of the damping capacity in medium frequency measurement are chiefly due to solid friction at the crack boundaries and the increase of the acoustic attenuation in high frequency measurement comes from acoustic scattering on the same crack boundaries. Effects at a microscopic scale such as an increase of the number of mobile defects due to the quench, are also possible.

One can now try to link the experimental values of ΔT_c to the theoretical analysis of the thermal shock resistance.

Thermoelastic theory.
In the case of a water quench the outside of the sample is in a tensile state and

the inside in a compressive state due to the expansion gradients induced by the inhomogeneous cooling of the sample. The stresses so induced first increase and reach a maximum value (Glenny, 1958) after a more or less long time, then they decrease as far as the interior temperature tends to be equal to the exterior temperature of the sample. The cracks nucleate if the maximum value of these thermal stresses exceeds in a single point the tensile strength of the material. A first thermal shock resistance parameter R (R' for the lower rates of heat transfert) is defined (Hasselman, 1970 b) :

$$R = \frac{\sigma_T f(\nu)}{E\alpha}$$

σ_T is the tensile strength, E the Young modulus and α the thermal expansion coefficient ; $f(\nu)$ is a function of the Poisson ratio and of the sample shape, $f(\nu) = (1-\nu)$ for many experimental cases .

The critical quenching temperature is derived from this parameter by : $\Delta T_c = R/A$. A is a non dimensional parameter ($0 \lesssim A \lesssim 1$) the numerical value of which is more close to 1 as the Biot's modulus (β) of the experiment is more close to infinity; $\beta = ah/k$, a is a "mean dimension" of the sample, h the heat transfert coefficient and k the thermal conductivity of the material. This analysis enables us to justify the previous remark about voluminous samples whose critical quenching temperatures are lower than those of small samples.

Practical applications of this theory are subjected to some reservations : (4)The strength of brittle materials can exhibit very important statistical variations (Davidge, 1979) and this aspect is not taken into account in the thermoelastic theory. (2) The analytic expression of R involves the tensile strength σ_T. This parameter is practically never measured and the bending strength σ_b is only known. In the frame of a statistical approach of the strength of brittle materials one must derive the proper value of σ_T from the experimental value of σ_b by using the Weibull's analysis (Davidge, 1979) - for example $\sigma_T \simeq \sigma_b/2$ in 3 point bending test. It is this calculated value of σ_T which must be used in the derivation of R. (3) The exact calculation of R requires a correct choice of the numerical values of the terms A, E, σ_T, α. The thermal shock being a transiert phenomenom, one must find a mean temperature T ($0 \lesssim T \lesssim T_c$) which is representative of the whole phenomenom. Kingery (1955) suggests to take $T = 0.8\ T_c$ and Ainsworth (1968) has given an example of application of this hypothesis. However, such a computation calls for the knowledge of $A = f(T)$, $E = f(T)$, $\alpha = f(T)$ and $\sigma_T = f(T)$.

These remarks may explain why the examples of a good agreement between measured and calculated values of ΔT_c are very scarce in the literature and why calculations postulate sometimes very different hypothesis (Davidge, 1967), (Hasselman, 1970 a).

Table 3 lists the values of R and ΔT_c for the three materials of the present study and for a single geometry of a disc.

TABLE 3 R and ΔT_c Values for Discs of Different Materials

Mat. \ R and ΔT_c	R(°C)	Calculated ΔT_c (°C)	Measured ΔT_c (°C)
$A\ell_2O_3$	30	60	170
$A\ell N$	70	140	255
Si_2N_3O	170	340	390

It may be noted that the agreement between measured and calculated ΔT_c values is never perfect and it appears that the lower the R parameter, the worse the agreement.

Energetic theory

This analysis does not suppose the nucleation of cracks when ΔT reaches ΔT_c, but assumes that preexisting cracks such as Griffith's microcracks can propagate when the critical ΔT_c value is reached. For this critical value of the temperature difference, the preexisting defects may propagate kinetically from an initial length ℓ_o to a final length ℓ_f which minimizes the total energy of the cracked system ($\partial U/\partial \ell = 0$).

The propagation occurs only for $\partial U/\partial \ell < 0$ and the calculated critical temperature difference is :

$$\Delta T_c = \left[\frac{\Pi G\ (1-2\nu)^2}{2E_o\alpha^2(1-\nu^2)}\right]^{1/2} \left[1 + \frac{16\ (1-\nu^2)\ N\ell^3}{9\ (1-2\nu)}\right] \left[\ell\right]^{-1/2}$$

ℓ_o, α and ν have been previously defined, E_o is the Young modulus of the uncracked material, G its shear modulus and N the density of cracks in the bulk.

The possibilities of practical applications of this pertinent analysis are few. Indeed, besides the questions rised during the discussion of the thermoelastic theory, it requires the knowledge of the density and the morphology of preexisting defects. However, it is a useful approach because it leads to a good explanation of the classical $\sigma_f = f(\Delta T)$ curve of the destructive tests, and also, because it involves the R'''' thermal shock resistance parameter expressed in terms of the final crack length ℓ_f. This parameter is a very important one and (Nakayama, 1966) has pointed out its good correlation with the industrial "number of cycles" tests.

$$R'''' = \frac{\gamma E}{\sigma_T^2\ (1-\nu)} \quad \text{or} \quad R'''' = \frac{K_{1c}^2}{2\sigma_T^2\ (1-\nu^2)}$$

γ is the surface energy for the initiation of fracture and K_{1c} the critical value of the stress intensity factor in mode I. For the materials tested in the present work the calculated values of the R'''' parameter are listed in Table 4.

No material exhibits a perfect resistance to thermal shocks, because a high resistance to thermal stresses and a high damage resistance are uncompatible. Indeed, a good resistance to thermal shock fracture requires high values for σ_T, low values for E and α, and good damage resistance requires low values for σ_T, high values for E and γ. There is a dilemna here, which has been discussed by other authors (Ainsworth, 1968), (Hasselman, 1973) and the solution of which is different for each given case. For the materials selected for this study, the R and R'''' thermal shock parameters are listed in Table 4.

TABLE 4 R and R'''' Thermal Shocks Resistance Parameter for Three Materials

R_i \ Mat.	$A\ell_2O_3$	$A\ell N$	Si_2N_2O
R (°C)	30	70	170
R'''' (μm)	550	65	450

It appears that silicon oxynitride exhibits the best compromise between the contrary demands previously underlined, that polycrystalline alumina is well resistant to damage but not to fracture and that aluminium nitride does not seem to be well resistant to thermal shocks.

CONCLUSION

The theoretical analyses of the thermal shock resistance of brittle materials lead to formulae of a rather poor applicability, and examples of good agreement between calculated and measured values of the critical quenching temperature is scarce. It seems that a more profitable approach is to calculate the value of thermal shock parameters as R, R' , R"" which appears as quality factors and allow comparisons between different materials. For an accurate characterization of the thermal shock resistance of a given component, experiments are absolutely necessary, and the dynamic methods developed in the present work allow the use of few samples only. Moreover, they are non destructive, thus they are well suited to the study of cumulative shock effects and they overcome the constraint of "homogeneous batch" by their ability of measurement before and after the thermal shock. At last, in the case of high damage resistant materials, these methods appear of better accuracy than the classical destructive test.

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