Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7408-ENG-36.

LA-UR--84-3137

DE85 000652

**TITLE: THE EFFECT OF ELEVATED-TEMPERATURE NEUTRON IRRADIATION
ON FRACTURE TOUGHNESS OF CERAMICS**

**AUTHOR(S): F. W. Clinard, Jr, G. F. Hurley, R. A. Youngman,
and L. W. Hobbs**

**SUBMITTED TO: First International Conference on Fusion Reactor Materials
Tokyo, Japan December 3-6, 1984
(Proceedings to be published)**

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes.

The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

msw

**The Effect of Elevated-Temperature Neutron Irradiation on
Fracture Toughness of Ceramics**

F. W. Clinard, Jr, G. F. Hurley, R. A. Youngman, and L. W. Hobbs

Materials Science and Technology Division

Los Alamos National Laboratory

Los Alamos, NM 87545, USA

Abstract

Single-crystal forms of MgAl_2O_4 , $\text{Y}_3\text{Al}_5\text{O}_{12}$, and Al_2O_3 were irradiated in the EBR-II fast fission reactor to a fluence of 0.3×10^{26} n/m^2 at 1015 K, and to $1-2 \times 10^{26}$ n/m^2 at 925 and 1100 K. Fracture toughness was subsequently measured at room temperature by an indentation technique, and radiation-induced defect aggregates were characterized by transmission electron microscopy. A slight increase in toughness of MgAl_2O_4 was observed, and attributed to interaction of cracks with strain fields around dislocation loops. No significant change was noted for $\text{Y}_3\text{Al}_5\text{O}_{12}$, despite the presence of a high concentration of unresolved defect clusters. Fracture toughness of Al_2O_3 was markedly increased, with the enhancement apparently attributable in large part to impedance of crack propagation by interaction with the irradiation-induced void lattice.

The Effect of Elevated-Temperature Neutron Irradiation on Fracture Toughness of Ceramics⁺

F. W. Clinard, Jr, G. F. Hurley, R. A. Youngman,* and L. W. Hobbs**

Materials Science and Technology Division

Los Alamos National Laboratory

Los Alamos, NM 87545, USA

Introduction

Ceramics are specified for a variety of applications in fusion reactors, involving their use as electrical insulators, microwave windows and structural materials (1). A prime requirement in all critical applications is retention of adequate strength after neutron irradiation, to resist structural, thermal or swelling stresses.

Fracture strength of ceramics is given by the relationship

$$\sigma_f = A \sqrt{E\gamma} / \sqrt{c}, \quad (1)$$

where E = elastic modulus, γ = fracture energy, c = critical flaw size to cause fracture, and A is a geometrical factor. It may be seen that strength is proportional to $\sqrt{E\gamma}$, the fracture toughness, at constant flaw size. In the work described here, single-crystal samples of MgAl_2O_4 , $\text{Y}_3\text{Al}_5\text{O}_{12}$, and Al_2O_3 were irradiated in a fast fission reactor at elevated temperatures to fluences up to the equivalent of a year at the first wall of a $2\text{MW}/\text{m}^2$ fusion

⁺Work performed under the auspices of the U.S. Department of Energy.

*now with Standard Oil Company, Cleveland, OH 44115.

**consultant, Massachusetts Institute of Technology, Cambridge, MA 02139.

reactor. Fracture toughness was subsequently measured by an indentation technique, and compared with that determined for annealed but unirradiated samples.

Results reported here are of technological as well as scientific interest, since all three test materials are candidates for fusion reactor applications. Although most ceramic components in use today have been fabricated in the polycrystalline form, with the advent of economical methods for growth of single crystals in engineering shapes (2) it is appropriate to consider the use of single-crystal components for fusion devices where improved performance can be anticipated.

Experimental Procedure

The three test materials were obtained from Linde Division, Union Carbide Corporation. Major impurities in wt ppm were, for MgAl_2O_4 : 100 Si, 20 Fe, 8 B, 5 Ca; for $\text{Y}_3\text{Al}_5\text{O}_{12}$: 10 Si, 10 Fe, 5 Ca; and for Al_2O_3 : 60 Fe, 50 Nb, 40 Mo. Coupons of dimensions 10x10x0.5 mm were sealed in an inert gas atmosphere and placed in the EBR-II fast fission reactor. Irradiation test conditions included a fluence of $0.3 \times 10^{26} \text{ n/m}^2$ ($E > 0.1 \text{ MeV}$) at 1015 K, and $1-2 \times 10^{26} \text{ n/m}^2$ at 925 and 1100 K. Control samples were annealed at these temperatures for times corresponding to reactor full power exposures.

Irradiated and control samples were subsequently impressed at room temperature with a Vickers diamond indenter, using loads of from 50 to 1000 g. Cubic MgAl_2O_4 and $\text{Y}_3\text{Al}_5\text{O}_{12}$ were indented in the {111} plane of the samples, while hexagonal Al_2O_3 was indented in the {0001} plane with the

diagonals of the indents along $\langle 10\bar{1}0 \rangle$ and $\langle 11\bar{2}0 \rangle$ directions. Hardness (resistance to plastic flow) was obtained from dimensions of the indent, while fracture toughness (resistance to crack propagation) was determined by applying fracture mechanics techniques to interpretation of crack patterns around the indentations (3). The latter parameter showed the larger changes with irradiation, and is of more significance in an engineering sense than is hardness. In this paper we restrict our discussion to fracture toughness, with a brief description of changes in hardness given in an Appendix. A more detailed account of the results of this work is given elsewhere (4-7).

Radiation-induced defect aggregates were evaluated by transmission electron microscopy. Portions of irradiated and control samples were mechanically thinned to 100 μm thickness before ion beam thinning to electron transparency. Foils were coated with carbon to preclude charging effects, and examined at 200 kV.

The greatest changes in fracture toughness were observed in Al_2O_3 . In order to determine the interaction between induced microcracks and the damage microstructure, TEM examination was carried out on samples after indentation. A grid of two hundred indents of 200g load was placed in a 1x1 mm area; spacing was adequate to avoid interaction of the crack patterns, but sufficiently dense to ensure finding indents in the thinned foil. Most of the thinning was carried out from the unindented side, but enough material was removed from the other surface to allow examination of the foil in the region below the bottom of the indents.

Results and Discussion

1 → MgAl₂O₄. Results of fracture toughness measurements for MgAl₂O₄ are
2 → shown in Figs. 1 and 2. Irradiation-induced changes after exposure to a
fluence of 3×10^{25} n/m² at 1015 K were not significant. However, slight
increases were observed at both 925 and 1100 K after a dose of 2×10^{26}
n/m².

TEM examination of the lower-fluence material showed little or no evidence of defect aggregation (8). Thermal diffusivity measurements reported elsewhere (4,9) revealed that this parameter was little changed in single-crystal spinel by any of the irradiation doses employed here, indicating a low concentration of sub-microscopic defects (e.g., point defects). Thus it may be concluded that the vast preponderance of displacements recombine rather than aggregate or remain isolated.

The slight increases in fracture toughness noted at the higher doses (Fig. 2) were accompanied by formation of faulted Frank interstitial loops, primarily $1/4\langle 110 \rangle\{110\}$ in character but with the added presence of $1/6\langle 111 \rangle\{111\}$ loops in the 925 K material. Configuration of the 925 K loops
3 → (usually clustered as rosettes) is shown in Fig. 3: those formed at 1100 K were similar, but of larger dimensions (8).

A number of possible mechanisms have been proposed to explain toughening of ceramics with heterogeneous microstructures. These include crack bowing (10-12); crack deflection (13,14); and internal stresses associated with coherency of second-phase particles, mismatches in thermal expansion coefficient, or differences in elastic properties of these phases (15-17). Another mechanism of possible relevance to the results for MgAl₂O₄ is

jogging of an atomically-sharp crack front resulting from intersection with point defects, submicroscopic defect clusters, or dislocation loops (typically with thicknesses on the order of atomic dimensions).

The mechanism which appears most appropriate to explain the observed toughening is that involving coherency strain fields. The Frank loops seen here are bounded by edge dislocations, the strain fields of which contain both hydrostatic and shear components. These strains decrease inversely with distance from the loop, but extend throughout the crystal unless modified by those from neighboring loops. A propagating crack would likely be deflected by the strain fields, resulting in crack impedance and consequent toughening. Thus irradiation-induced toughening of spinel bears some resemblance to strengthening of $\text{MgO} \cdot 3.5\text{Al}_2\text{O}_3$ by coherent precipitates as reported by Bansal and Heuer (15), and to toughening of glass by thermal contraction mismatch with a dispersed phase observed by Nadeau and Dickson (17). In the latter work, data were cited showing strength increases of up to 1.8 times that of the pure glass when the particles were small and had an optimal thermal expansion mismatch. The same size particles with no mismatch gave a ratio of 1.4, implying that the difference is the result of crack deflection due to internal stresses. The inhibiting influence of the stress fields was a function of the extent of the mismatch, and the size and spacing of the particles.

Intersection of the crack front with fine-scale defects might also contribute to toughening, but since the concentration of such defects appears to be small, this mechanism would not seem to be a major contributor. At any rate, results for $\text{Y}_3\text{Al}_5\text{O}_{12}$ discussed in the next Section suggest that this mechanism cannot make a large contribution even under more favorable

conditions.

4 → $\text{Y}_3\text{Al}_5\text{O}_{12}$. Results for this ceramic (Figs. 4 and 5) indicate no significant
5 → change in toughness. TEM examination showed no resolvable defect aggregates
after the 1015 K and 925 K irradiations, and only a sparse population of
6 → faulted {110} dislocation loops after 1100 K exposure (Fig. 6). The
concentration of these loops was perhaps 1 % of that observed in spinel.
However, a high concentration of unresolved mottled damage was evident,
suggesting that a significant fraction of the total displacement damage was
retained in the lattice. This was confirmed by thermal diffusivity
measurements, which showed reductions of 60% after irradiation at 1015 and 1100
K and of 80 % after exposure at 925 K (4). If a high concentration of
fine-scale defects were capable of impeding a propagating crack, significant
toughening should have been observed in this material. Absence of such an
effect may imply that these cracks are not atomically sharp, at least in
 $\text{Y}_3\text{Al}_5\text{O}_{12}$.

Al_2O_3 . The effect of neutron irradiation under the conditions employed
1 → here was to cause major increases in fracture toughness. As shown in Figs. 7
8 → and 8, the effect was most marked after a fluence of $1.8 \times 10^{26} \text{ n/m}^2$ at 1100
K, with lesser increases after irradiation at lower doses and/or the other test
temperatures of 1015 and 925 K. Transmission electron microscopy showed that in
all cases the damage microstructure was characterized by a high concentration
of small voids aligned in the c-axis direction, accompanied by unfaulted
interstitial dislocation loops and tangles resulting from these loops (18).
Void parameters for the low-dose irradiation at 1015 K and highest-dose

irradiations at 925 and 1100 K are given in Table 1. TEM examination of cracks emanating from indentations revealed that these cracks interacted strongly with

Fig. 9 → the voids. Figures 9 and 10 compare crack morphology before and after

Fig. 10 → irradiation, and demonstrate that while in the former case cracks are featureless and straight on a fine scale, in the irradiated material cracks undergo void intersection, changes of direction, jogging, and branching. The appearance of jogging and branching was restricted to the 1100 K material, where voids were largest.

The toughening observed in Al_2O_3 may be attributable to the combined effect of a number of mechanisms operating simultaneously. Coherency stresses from the presence of dislocation loops may be a contributing factor, but the evidence of Fig. 10 suggests that crack-void interactions dominate. Neither the crack bowing nor crack deflection models seem appropriate here, as effective toughening in both cases implies a resistance to penetration which voids do not possess. Faber and Evans (13) calculated the lower bound to crack deflection toughening by considering only surface area increase, but the enhancements obtained are not sufficient to explain the present results.

In studies of crack/pore interactions in glass, Green et al. (19) found attraction of the crack to the pore (a result of the tensile strain field around the pore); crack impedance, segmentation and change of shape; and secondary fracture. These phenomena bear some similarity to the microstructural effects observed here, and together with the crack blunting which should result from crack/void intersection could account for the toughening of Al_2O_3 .

We have applied a simple analytical technique based on the model of Evans (11) to correlate observed changes in fracture toughness with changes in void

parameters. This model, which was developed for use with composites and assumes
11 → that cracks bow out between impenetrable obstacles, is illustrated in Fig. 11.
The parameter $2C$ was taken to be the mean free path between void intersections
along random straight lines in the $\{0001\}$ plane. Results of the calculations
12 → are compared with measured changes in fracture toughness, in Table 1. Agreement
is surprisingly good, suggesting that the relevance of the model to void
toughening lies more in its treatment of the geometrical arrangement of voids
than in their physical properties. However, the model underestimates the
increase in toughness after 1100 K irradiation, where the void size is much
greater than for the other two cases; this deviation may be related to the fact
that crack jogging and branching were observed only after the high-temperature,
high-dose irradiation.

Conclusions

Fracture toughness of single-crystal Al_2O_3 is significantly increased
by neutron irradiation to high doses at elevated temperatures, in large part as
a result of impedance of crack propagation by interaction with voids. A lesser
enhancement was noted for $MgAl_2O_4$, and is attributed to interaction of
cracks with strain fields around interstitial dislocation loops. No
significant change of toughness was seen in $Y_3Al_5O_{12}$ despite the presence
of a high concentration of unresolved damage aggregates. These results
indicate that spinel in either single-crystal or polycrystalline form can
exhibit an enhancement of mechanical properties after irradiation in the
environment of a fusion reactor. Al_2O_3 , which as a non-cubic

polycrystalline material is susceptible to microcracking from anisotropic swelling, can show major strengthening if used in single-crystal form.

References

1. F. W. Clinard, Jr. and G. F. Hurley, J. Nucl. Mater. 103 & 104 (1981) 705.
2. G. F. Hurley, A. D. Meuse, A. D. Morrison and S. M. Smith, Bull. Am. Ceram. Soc. 54 (1975) 510.
3. A. G. Evans and E. A. Charles, J. Am. Ceram. Soc. 59 (1976) 371.
4. G. F. Hurley and F. W. Clinard, Jr., in: Special Purpose Materials Annual Progress Report (DOE/ET-0095, 1979) pp. 45 and 59.
5. G. F. Hurley and F. W. Clinard, Jr., in: Special Purpose Materials Annual Progress Report (DOE/ER-0048/1, 1980) p. 51.
6. G. F. Hurley and R. A. Youngman, Third Annual Progress Report on Special Purpose Materials for Magnetically-Confined Fusion Reactors (DOE/ER-0013, 1981) p. 51.
7. G. F. Hurley and R. A. Youngman, Fourth Annual Progress Report on Special Purpose Materials for Magnetically-Confined Fusion Reactors (DOE/ER-0113/1, 1982) p. 45.
8. L. W. Hobbs and F. W. Clinard, Jr., J. de Physique 41 (1980) C6-232.
9. F. W. Clinard, Jr., J. Materials for Energy Systems, in press (1984).
10. F. F. Lange, J. Am. Ceram. Soc. 54 (1971) 614.
11. A. G. Evans, Phil. Mag. 26 (1972) 1327.
12. D. J. Green, J. Am. Ceram. Soc. 66 (1983) C-4.
13. K. T. Faber and A. G. Evans, Acta Metall. 31 (1983) 565.

14. K. T. Faber and A. G. Evans, Acta Metall. 31 (1983) 577.
15. G. K. Bansal and A. H. Heuer, Phil. Mag. 29 (1974) 709.
16. D. J. Green and P. S. Nicholson, Fracture Mechanics of Ceramics Vol. 4 (Plenum, New York, 1976) p. 945.
17. J. S. Nadeau and J. I. Dickson, J. Am. Ceram. Soc. 63 (1980) 517.
18. F. W. Clinard, Jr., G. F. Hurley and L. W. Hobbs, J. Nucl. Mater. 108 & 109 (1982) 655.
19. D. J. Green, P. S. Nicholson and J. D. Embury, J. Mater. Science 12 (1977) 987.

Appendix

Changes in hardness of the materials studied here can be summarized as follows:

MgAl₂O₄. Hardness was essentially unchanged after irradiation at 1015, 925, or 1100 K.

Y₃Al₅O₁₂. No significant change was observed after any of the irradiation treatments.

Al₂O₃. Hardness was increased ~10 % after 1015 K irradiation. Irradiation to either 0.9×10^{26} or 1.8×10^{26} n/m² at 925 K resulted in an increase of ~15%, while the same doses at 1100 K increased hardness ~10%.

Figure Captions

Fig. 1. Fracture toughness of MgAl_2O_4 as a function of diagonal dimension of indentation, as annealed and after irradiation at 1015 K. The slight dependence of toughness on indentation size may be related to surface effects.

Fig. 2. Fracture toughness of MgAl_2O_4 as a function of diagonal dimension of indentation, as annealed and after irradiation to $2 \times 10^{26} \text{ n/m}^2$. Results from material annealed at 925 and 1100 K are not differentiated.

Fig. 3. Bright-field TEM image of faulted dislocation loops in MgAl_2O_4 after irradiation to $2 \times 10^{26} \text{ n/m}^2$ at 925 K. A detailed description of the loops is given in ref. 8.

Fig. 4. Fracture toughness of $\text{Y}_3\text{Al}_5\text{O}_{12}$ as a function of diagonal dimension of indentation, as annealed and after irradiation at 1015 K.

Fig. 5. Fracture toughness of $\text{Y}_3\text{Al}_5\text{O}_{12}$ as a function of diagonal dimension of indentation, as annealed and after irradiation to $2 \times 10^{26} \text{ n/m}^2$. Results from annealed and irradiated material are not differentiated as to temperature.

Fig. 6. Bright-field TEM image of dislocation loops and unresolved damage in $\text{Y}_3\text{Al}_5\text{O}_{12}$ after irradiation to $2 \times 10^{26} \text{ n/m}^2$ at 1100 K.

Fig. 7. Fracture toughness of Al_2O_3 as a function of diagonal dimension of indentation, as annealed and after irradiation at 1015 K.

Fig. 8. Fracture toughness of Al_2O_3 as a function of diagonal dimension of indentation, as annealed and after irradiation to 0.9 and $1.8 \times 10^{26} \text{ n/m}^2$. Results from material annealed at 925 and 1100 K are not differentiated.

Fig. 9. Bright-field TEM image of an indentation-induced crack in unirradiated Al_2O_3 .

Fig. 10. Bright-field TEM image of an indentation-induced crack in Al_2O_3 after irradiation to $2 \times 10^{26} \text{ n/m}^2$ at 1100 K.

Fig. 11. Model for interaction of cracks with irradiation-induced voids.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

DIRECTION OF CRACK PROPAGATION

Figure 11

Table 1. Comparison of observed toughening ($K_{c\text{irrad}}/K_{c\text{contr}}$) and predicted toughening ($\sigma_{\text{irrad}}/\sigma_{\text{contr}}$) in Al_2O_3 . Other symbols are defined in Fig. 11. Measured values of fracture toughness were taken in the range of indentation sizes deemed to give the most reliable results.

Irrad. T (K)	$K_{c\text{irrad}}/K_{c\text{contr}}$	$2r_0$ (nm)	$2C$ (nm)	$2C/2r_0$	$\sigma_{\text{irrad}}/\sigma_{\text{contr}}$
925	1.2	3.6	18	5.0	1.2
1015	1.3	5.2	24	4.6	1.3
1100	2.1	9.0	21	2.3	1.7