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Subcritical crack growth in high-grade alumina for container applications

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ABSTRACT

The subcritical crack growth of a high-grade, 99.9 % alumina ceramic in a concentrated salt solution at 70 C° was investigated by lifetime measurements in static bending tests. For crack growth velocities lower than 10^{-8} m/s, the crack growth velocity - stress intensity factor relationship was found to be a power law with an exponent in the order of 50. The material had an inert bending strength close to 400 MPa. These values of the material parameters qualify the alumina investigated for high-level waste container applications from the point of view of subcritical crack growth.

ZUSAMMENFASSUNG

Mittels Lebensdauerbestimmungen in statischen Biegeversuchen wurde das unterkritische Risswachstum einer 99.9-prozentigen Aluminiumoxydkeramik hoher Qualität in einer konzentrierten Salzlösung bei 70 C° untersucht. Für Risswachstumsgeschwindigkeiten kleiner als 10^{-8} m/s lässt sich die Beziehung zwischen Risswachstumsgeschwindigkeit und Spannungsintensitätsfaktor als Potenzfunktion mit einem Exponent der Grössenordnung 50 ausdrücken. Die inerte Biegefestigkeit des Materials beträgt rund 400 MPa. Was das unterkritische Risswachstum betrifft, qualifizieren diese Werte der Materialparameter das untersuchte Aluminiumoxyd als Material zur Herstellung von Behältern für hochradioaktive Abfälle.

RESUME

La propagation sous-critique de fissures dans une céramique d'alumine à 99.9 %, de qualité élevée, a été analysée dans une solution saline concentrée, à 70 C°, en mesurant la durée de vie au cours d'essais statiques de flexion. Pour des vitesses de propagation des fissures de moins de 10^{-8} m/s, la relation entre la vitesse de propagation des fissures et le facteur d'intensité de tension s'est avéré être une loi de puissance avec en exposant de l'ordre de 50. Le matériau a une résistance inerte à la flexion d'environ 400 MPa. En ce qui concerne la propagation sous-critique de fissures, ces valeurs des paramètres caractéristiques qualifient l'alumine analysée en vue de la fabrication de conteneurs pour déchets hautement radioactifs.

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1. Introduction

Al_2O_3 is being considered as a candidate material for containers to be used for the ultimate storage of high level waste. This material has important advantages in terms of its corrosion behaviour. But it is disadvantageous that alumina exhibits subcritical crack growth under tensile loads, especially in a water environment. Due to defects introduced in fabrication and processing failure by crack extension has to be expected to occur as long as tensile stresses cannot be totally excluded. In a first investigation [1] the qualification of a hot isostatically pressed alumina [2,3] was judged by determination of crack growth in synthetic water representative for groundwaters in the granitic bedrock of Northern Switzerland. Strong crack growth was found as the main reresult and for this reason the material investigated could not be recommended for containers. In this report the same analysis made for an alternative alumina material will be described.

2. Basic relations

The failure of ceramic components is often caused by subcritical crack propagation. In the range of linear-elastic fracture mechanics crack growth is governed only by the stress intensity factor K which describes the stresses near a crack tip

$$\frac{da}{dt} = v(K) \qquad (1)$$

If σ denotes the stress and a the depth of a crack in a structure, K is defined by

$$K = \sigma Y \sqrt{a} \qquad (2)$$

where Y is a geometric correction factor dependent on the shape of the crack and the component. To allow correct lifetime predictions the relation (1) has to be known, especially for extremely low crack growth rates. A simple method to determine such curves based on lifetime measurements was proposed in [1,4].
The lifetime t_f is given by rewriting eq.(1)

$$t_f = \int_{a_i}^{a_c} \frac{da}{v} \qquad (3)$$

where a_i is the initial crack size and a_c its critical value at the moment of failure.

From eqs.(2) and (3) one obtains

$$t_f = \frac{2}{(\sigma Y)^2} \int_{K_{Ii}}^{K_{Ic}} \frac{1}{v} K_I \, dK_I \tag{4}$$

where K_{Ii} denotes the initial stress-intensity factor

$$K_{Ii} = \sigma \sqrt{a_i} \, Y_i \tag{5}$$

Y_i is the geometric function of the initial crack, and K_{Ic} is the fracture toughness of the material considered. To evaluate lifetime measurements under different stresses it is suitable to consider the quantity

$$t_f \sigma^2 Y^2 = 2 \int_{K_{Ii}}^{K_{Ic}} \frac{1}{v} K_I \, dK_I \tag{6}$$

So the quantity $t_f \sigma^2 Y^2$ is only a function of the initial stress intensity factor K_{Ii} as can be deduced from the right-hand side of eq.(6).

Differentiation of eq.(6) with respect to the initial stress intensity factor K_i results in

$$v(K_{Ii}) = - \frac{2}{t_f \sigma_c^2} (K_{Ic}/Y_i)^2 \frac{d[\log(K_{Ii}/K_{Ic})]}{d[\log(t_f \sigma^2 Y_i^2)]} \tag{7}$$

where $$\sigma_c = K_{Ic}/(Y\sqrt{a_i}) \tag{8}$$

is the strength in the absence of subcritical crack growth, i.e. the so-called "inert strength".

For a power law $v \propto K^n$ one would obtain

$$d[\log(K_{Ii})]/d[\log(\sigma^2 t_f Y^2{}_i)] = -1/(n-2) \tag{9}$$

3. Description of the procedure

The procedure of evaluation v(K) is relatively simple. In a first series of tests N samples are tested in dynamic bending tests at high stress rates in an inert environment to give the distribution of so-called "inert strength". The N values of strength are arranged in an increasing order. In a second series, also involving N specimens, the lifetimes are measured. The results are also arranged in increasing order. The ν-th value of lifetime $t_{f,\nu}$ is associated with the ν-th value of inert bending strength $\sigma_{c,\nu}$. The latter is transformed into $K_{i,\nu}/K_c$ using the relation

$$K_i/K_c = \sigma/\sigma_c \tag{10}$$

Because lifetimes and associated strength data are known, the v-K_I-curve can be plotted using eqs.(7,8).

4. Experimental investigations

4.1 Dynamic bending strength

For the experiments three batches of bending bars 4.4 x 4.4x50 mm, made of a commercial 99.9% Al_2O_3 (Metoxit AG, Thayngen, Switzerland), were provided by NAGRA.

The main contaminations of the raw material were determined to be:

MgO	400 ppm (grain refiner)
SiO_2	30 ppm
Fe_2O_3	15 ppm
CaO	5 ppm
Na_2Co	40 ppm
K_2O	< 1 ppm
Cr_2O_3	"
TiO_2	"

The raw material had an average grain size (d_{50}) of 0.5µ and a BET of 7 m^2/g. It was used as spray-dried.

The material was isopressed on an automatic isopress in the shape of cylindrical rods and sintered at a temperature of 1550°C. After sintering the rods were inspected for cracks by the Zyglo crack penetration method. Bars of 4.4 x 4.4 x 50 mm were cut out of the rods and the surface was diamond ground.

Two batches (I+II) of 50 and 30 pieces respectively, were prepared. The ceramic microstructure can be described as follows

Density 3.90 g/cm^3 , Average grain size 4 μ

Before testing the alumina specimens were annealed for 4 hours in a vacuum of 10^{-5} bar at 1150°C.
To determine the inert bending strength σ_c for each batch 15 specimens were subjected to dynamic 4-point bending tests in air with a 20mm inner span and a 40mm outer span. The tests were performed at a very high loading rate of 3000N/s - using a transient recorder - to avoid any subcritical crack propagation.
The distribution of ceramic strength values can often be described by a Weibull distribution. For the bending strength in an inert medium, σ_c, the cumulative frequency F , i.e. the probability that the actual strength of a randomly chosen sample lies below σ_c, is given by

$$F = 1 - \exp[-(\sigma_c/\sigma_0)^m] \qquad (11)$$

where m and σ_0 are the Weibull parameters. Plotting the measured data in the form

$$\ln \ln 1/(1-F) = m \ln(\sigma_c/\sigma_0) = m \ln \sigma_c - m \ln \sigma_0$$

results in a straight line with the slope m.
In fig.1 the Weibull representation of the inert strength data is shown. Application of the maximum-likelihood method yielded the parameters for the two batches mentioned before

I : $m= 9.7$ $\sigma_0= 434$ MPa
II : $m= 7.5$ $\sigma_0= 392$ MPa

A third batch was lapped in order to assess the effect of this treatment on the inert strength. No improvement was observed.
The fracture toughness was determined to be $K_{Ic} = 3.9\ \mathrm{MPa}\sqrt{m}$ for all three batches.

4.2 Lifetime measurements

A salt solution (based on NAGRA AN/84-61) was specified as the test environment. The concentrations in mg/l were

NaCl	8297
KCl	86
$MgCl_2$ $6H_2O$	22
$SrCl_2$ $6H_2O$	64
NaF	8
$NaHCO_3$	84
$CaCl_2$ $2H_2O$	3191
Na_2SO_4	2307

identical to those used in [1].

In this environment static bending tests were performed at $70^{\circ}C$. The obtained lifetimes t_f are plotted in fig.2 in a Weibull representation.

4.3 Results

The geometric function for small surface cracks with an assumed semicircular shape is

$$Y = 1.04 \frac{2}{\sqrt{\pi}} \qquad (12)$$

In accordance with eq.(6) the lifetime quantity $t_f\sigma^2$ was calculated from these measurements and plotted in fig.3 . The mean curve is given by the dash-dotted curve.

From these data the subcritical crack growth rate was computed applying eq.(7). Figure 4 shows the result as a v-K_I-curve. It can be seen that for low crack growth rates the v-K_I-curve can be expressed by a simple power law

$$v = A\, K_I^{\,n}$$

with $A = 2.2 \cdot 10^{-31}\ MPa^{-48} m^{-23} s^{-1}$ and $n = 48$.

The corresponding straight line is plotted in addition to the data points of fig. 4 . For the Al_2O_3 investigated this power law describes the crack growth rate up to $K_I/K_{Ic}=0.75$.

With this power law eq.(4) can be easily integrated. Because most of the time is spent within this power law range the choice of the upper integration limit does not affect the lifetime:

$$K_{Ii}^{n-2} << K_{Ic}^{n-2}$$

The integration then leads to

$$t_f = B\sigma_c^{n-2}\ \sigma^{-n} \qquad (13)$$

with $$B = 2/[AY^2(n-2)K_{Ic}^{n-2}] \qquad (14)$$

Using the values of A and n obtained above one obtains $B=0.028\ MPa^2h$. Equation (13) is appropriate to allow lifetime predictions. Since the crack sizes in ceramics are statistically distributed, this prediction will yield a corresponding lifetime distribution.
Substituting t_f for σ_c in eq.(11) using eq.(13) results in a Weibull-distribution for the time-to-failure with the cumulative frequency

$$F = 1 - \exp[-(t_f/t_0)^{m^*}] \qquad (15)$$

with

$$m^* = \frac{m}{n-2} \qquad (16)$$

and

$$t_0 = B\sigma_0^{n-2}\sigma^{-n} \qquad (17)$$

The lifetime distribution is finally given by

$$t_f = B\sigma_0^{n-2}\exp[\ \frac{n-2}{m}\ \ln\ \ln\frac{1}{1-F}\ ]\ \sigma^{-n} \qquad (18)$$

In fig.5 this equation is plotted for the experimentally obtained values of B, n and mean values of $m=9$ and $\sigma_0=420MPa$.
The dashed lines represent the material investigated in [1].
For containers suitable for ultimate storage an admissible failure probability of $F = 0.001$ is assumed. Two points are of interest:

a) A minimum lifetime of 1000 years may be required. Which are the admissible tensile stresses?

Insertion of $F = 10^{-3}$, $n=48$, $m=9$, $B=0.028\ MPa^2h$, $\sigma_0=420MPa$ and $t_f = 8.76\cdot10^6h$ and use of eq.(18) gives

$$\sigma_{max} = 105\ MPa$$

b) In the container residual stresses σ_{res} caused in fabrication are supposed. The expected lifetimes are

1. $t_f = 1.1$ years for $\sigma_{res} = 120$ MPa
2. $t_f = 6950$ years for $\sigma_{res} = 100$ MPa
3. $t_f = 3 \cdot 10^8$ years for $\sigma_{res} = 80$ MPa

These examples are entered in fig.5

The lifetime predictions mentioned in this chapter are valid for parts of approximately the same size as the specimens tested. It is known that the strength and the life time will decrease if the volumina and surface of a real design is larger than that of measured bending specimens. Consequently, the tolerable stresses and attainable lifetimes diminish. The distribution of stresses in the container wall has to be known to allow exact calculations.

5. Comparison with alumina investigated in [1]

To give a comparison with the hot isostatically pressed alumina investigated in [1] both v-K_I-curves are plotted in fig.6 . Since the fracfracture toughness data are identical ($\approx 4 \mathrm{MPa}\sqrt{\mathrm{m}}$), both curves can be directly compared in this representation.
It is obvious that the alumina investigated in this report is characterized by significantly lower subcritical crack growth and a higher exponent n which is very important for lifetime predictions.
The superiority becomes also clear from fig.5 where the lifetimes predicted for the hot pressed material (dashed lines) are distinctly lower. The experimental evidence so far is insufficient to explain this effect, which might in principle be due to differences in grains size, composition, sintering procedure etc..
From the point of view of subcritical crack growth the material investigated seems to be qualified for long-term applications as a container material.

6. Summary

The subcritical crack growth of a 99.9% alumina in a highly concentrated salt solution at 70°C was investigated by lifetime measurements in static bending tests. The most important results are:

- The Weibull parameters of the inert bending strength distributions for the two surface qualities investigated were found to be $m = 7.5\text{-}9.7$, $\sigma_0 = 392\text{-}434$ MPa
- The obtained $v\text{-}K_I$-curve can be expressed for $v<10^{-8}$ m/s by a power law with an exponent $n \simeq 50$. This high value implies a good crack growth resistance.
- The high purity alumina investigated is superior to the hot isostatically pressed alumina previously [1-3] investigated

7. References

[1] T. Fett, K. Keller, D. Munz, "Determination of crack growth parameters of alumina in 4-point bending tests", NAGRA Technical Report 85-51, Sept. 1985.

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8. Figures

Fig.1 Weibull-plot of the inert bending strength σ_c for the two batches.

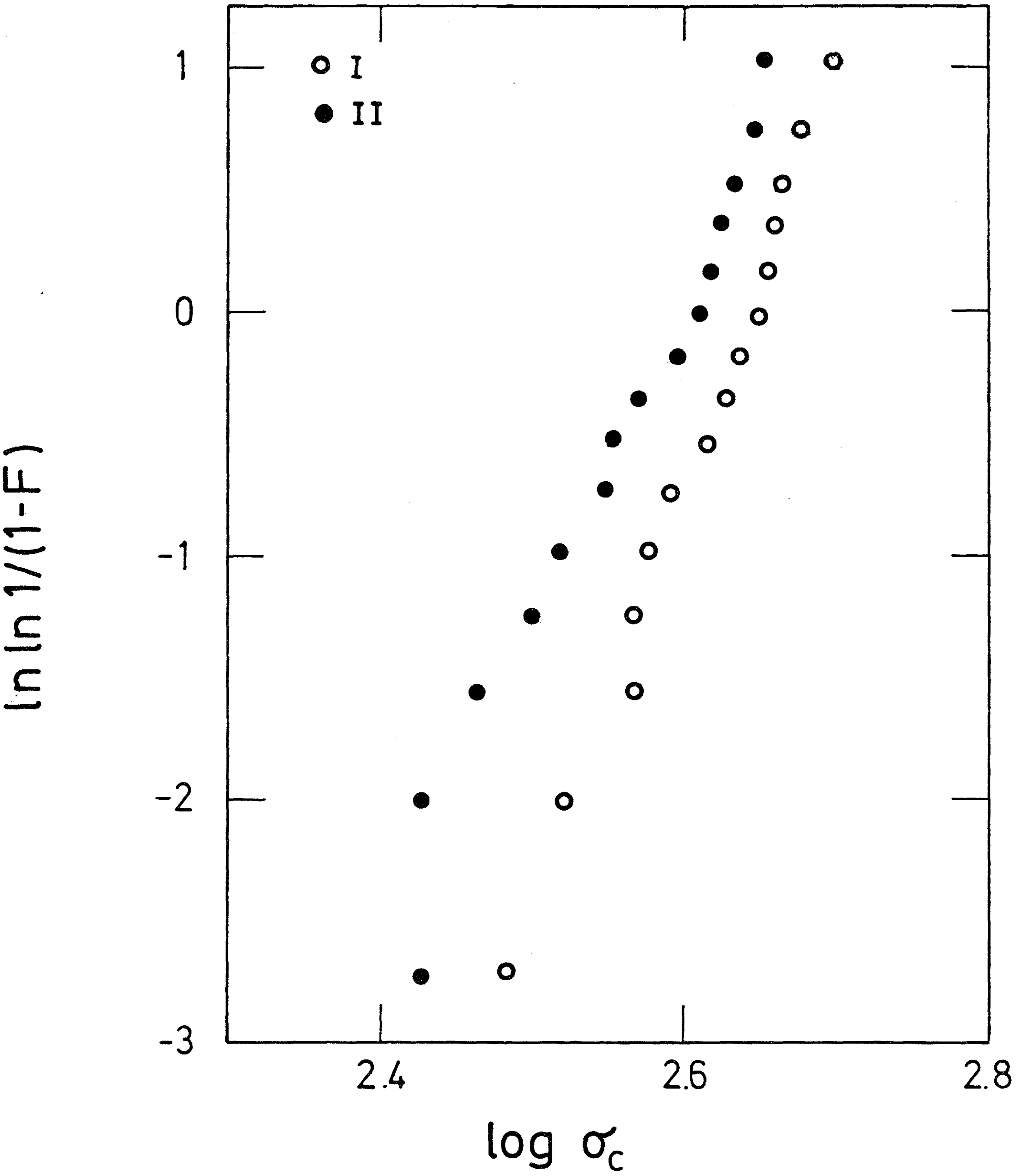
Fig.2 Lifetimes in 4-point bending tests in salt solution at 70°C.

Fig.3 Lifetime quantity $t_f\sigma^2$ as a function of "normalized stress" σ/σ_c.

Fig.4 v-K_I-curve obtained from eq.(7).

Fig.5 Nomograph for lifetime predictions.
(dashed lines: material investigated in [1])

Fig.6 Comparison of subcritical crack growth behaviour of the competing aluminas.



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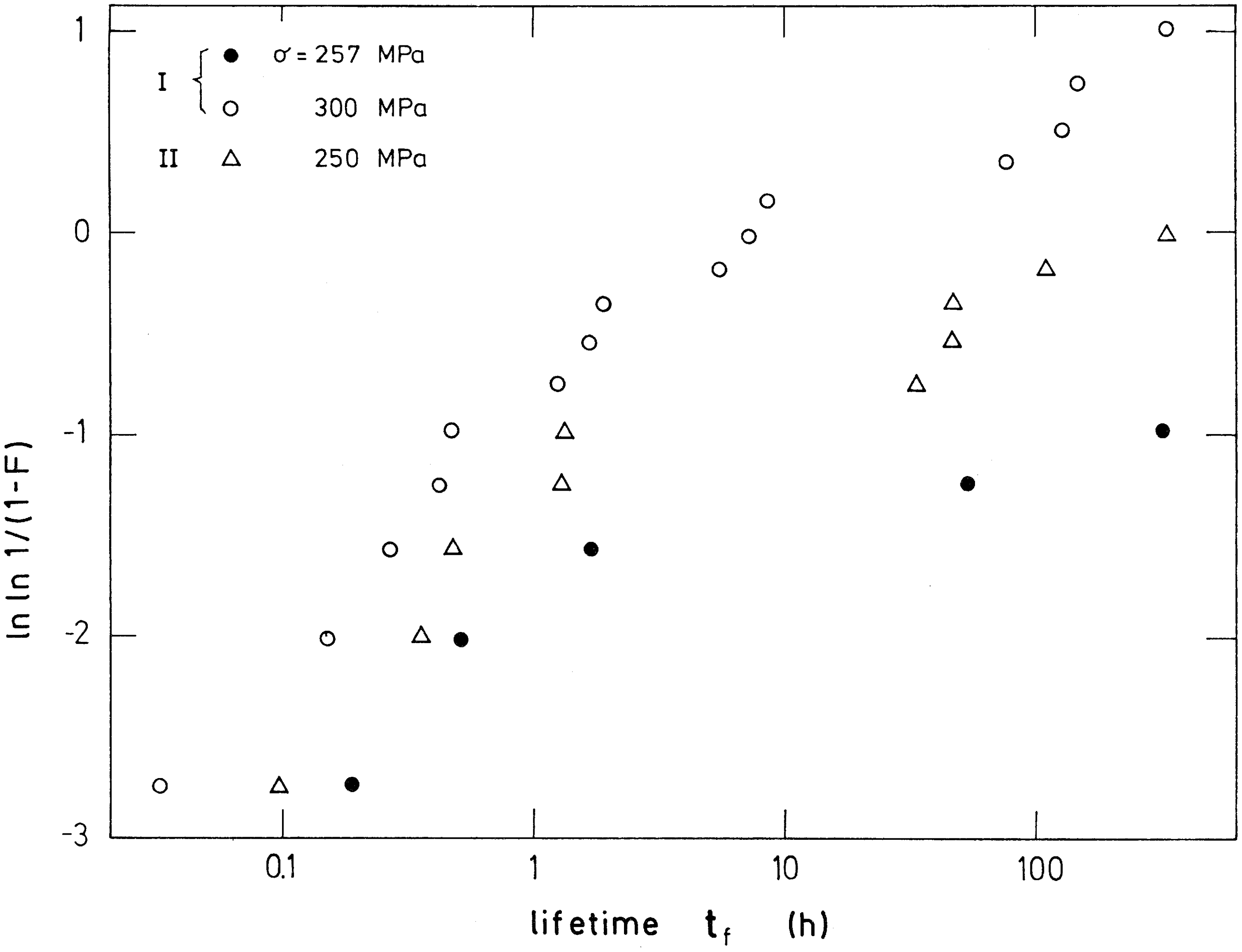

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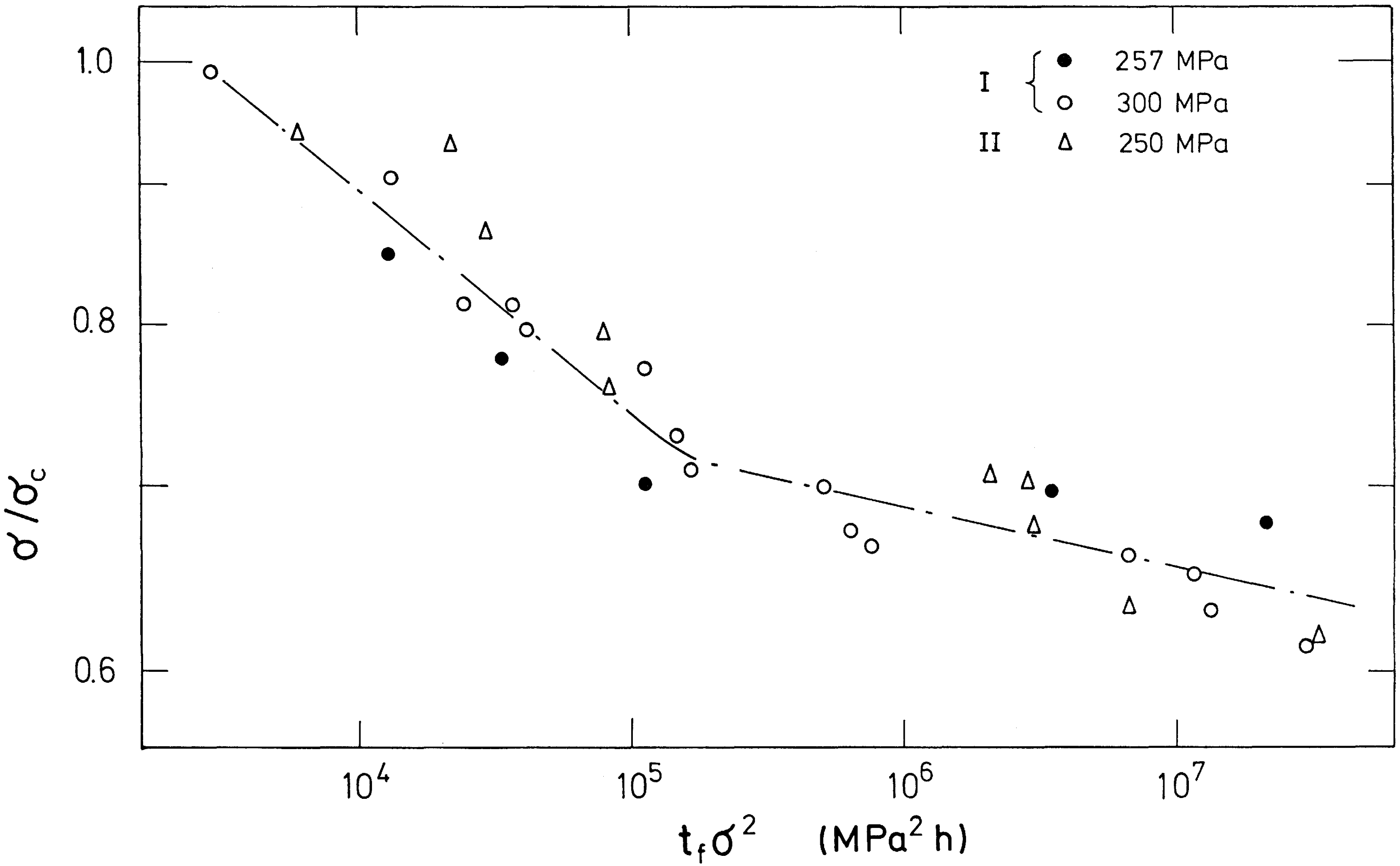

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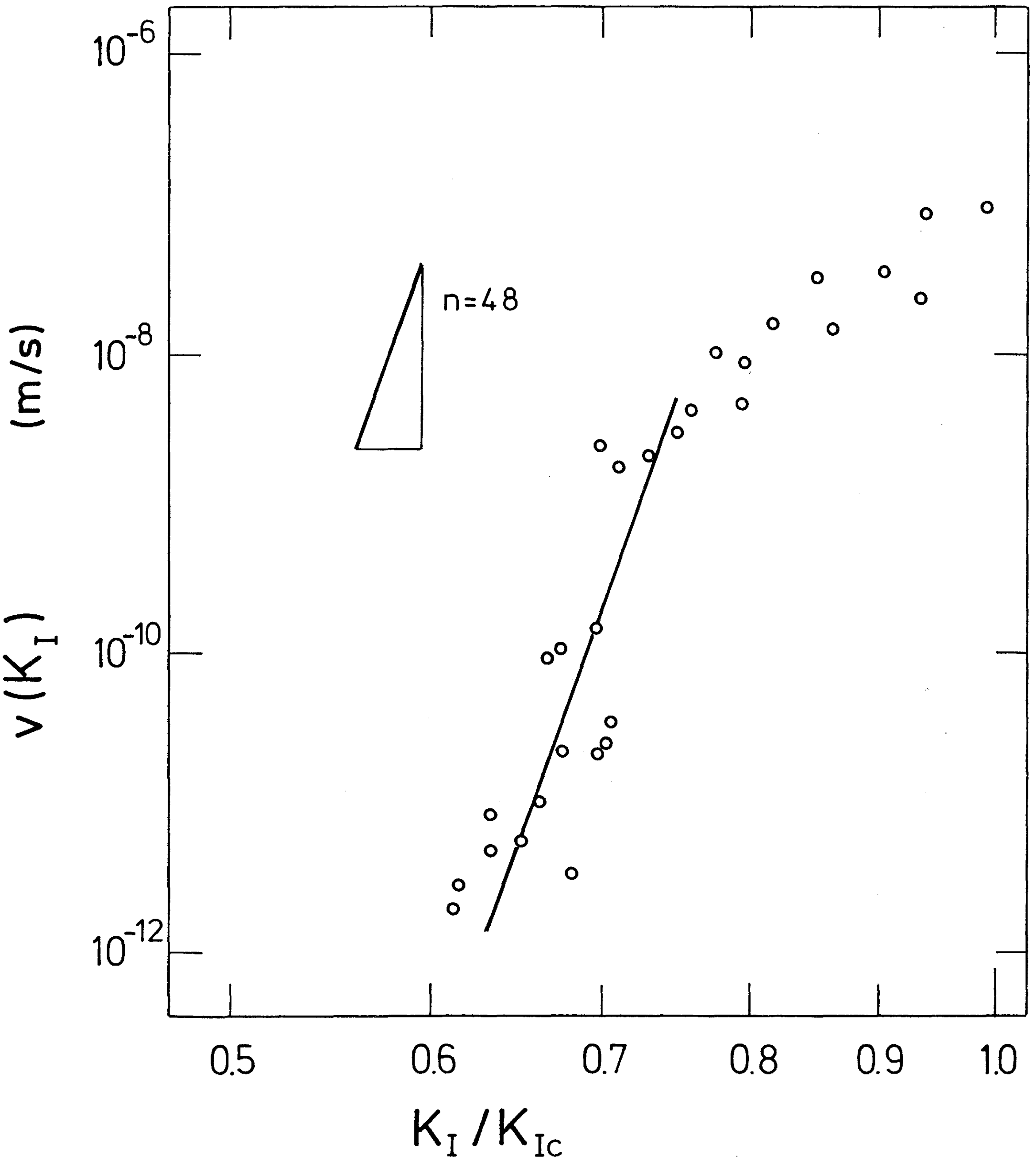
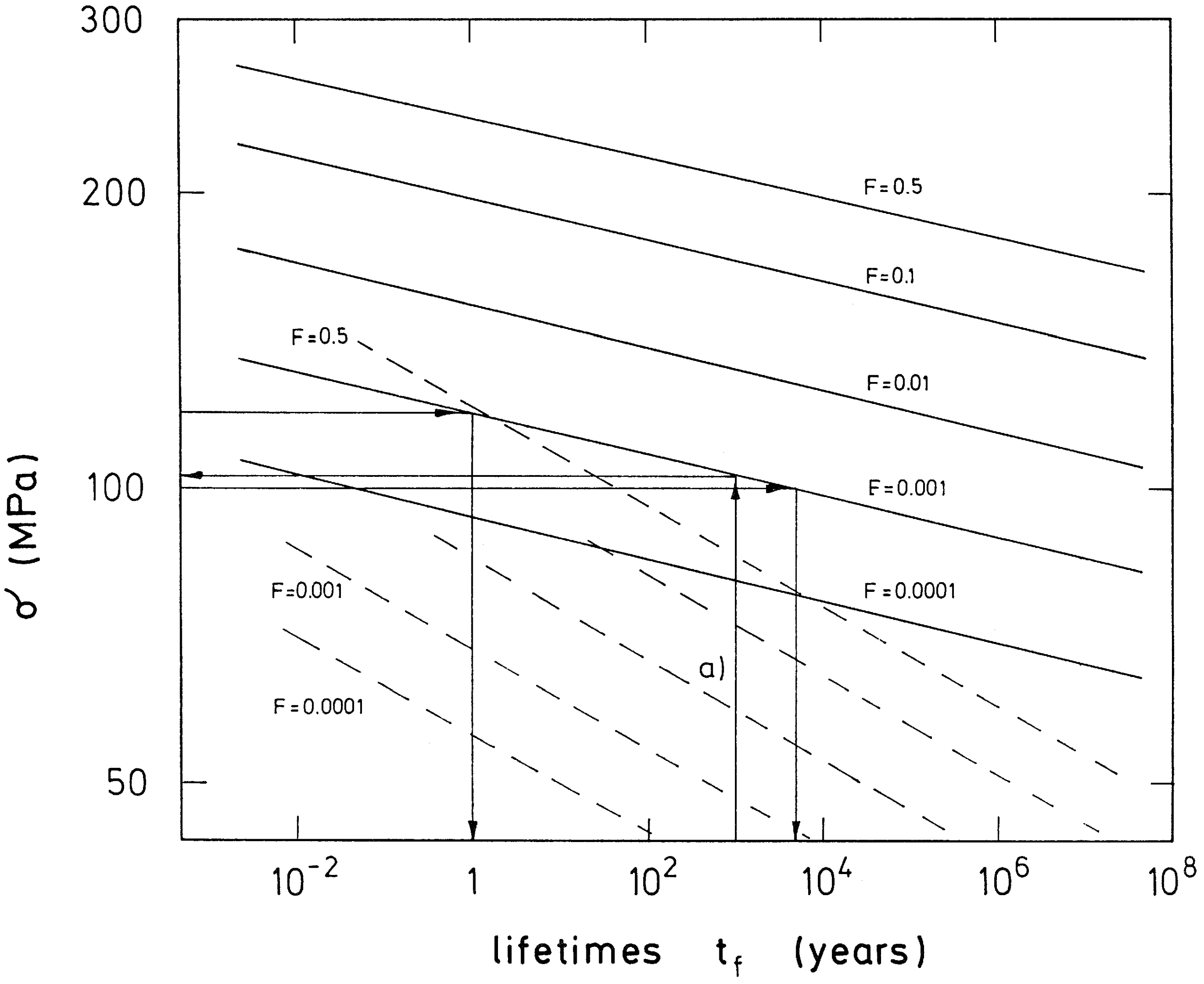


Fig.4 v-K_I-curve obtained from eq.(7).



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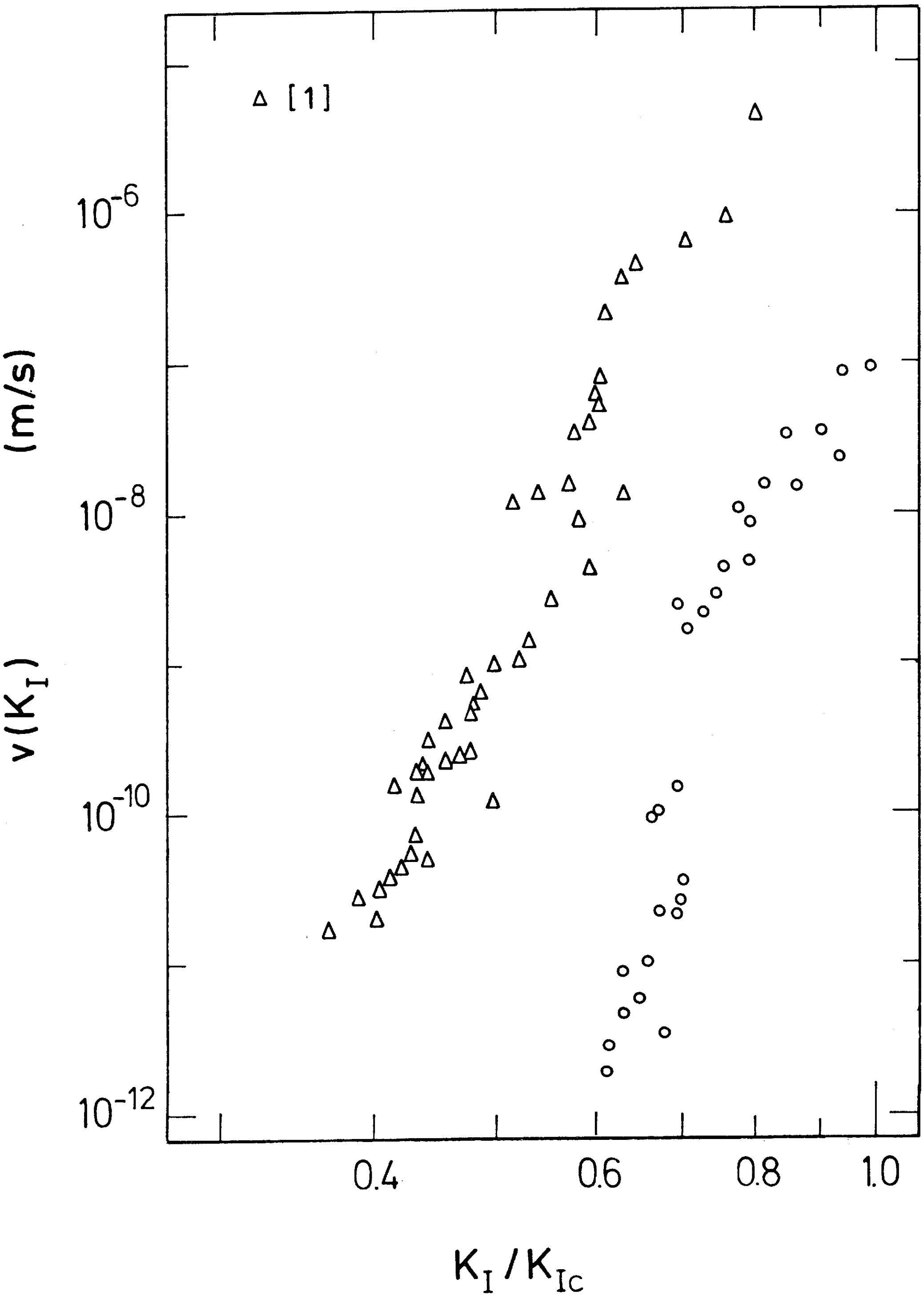

Fig.6 Intercomparison of subcritical crack growth behaviour of the competing aluminas.