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Nondestructive Evaluation Method on Mechanical Property Change of Graphite Components in the HTGR by Ultrasonic Wave Propagation with Grain/Pore Microstructure

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ABSTRACT

Oxidation damage is one of the crucial factors to degrade mechanical properties of graphite components in the HTGRs. The oxidation increases the porosity of graphite and, hence, results in the degradation. In order to evaluate the oxidation damage at neutron irradiated conditions, a new analytical method by ultrasonic wave propagation characteristics was developed. Irradiation effects, a dimensional change and a pinning of dislocations in crystals, on the propagation characteristics in graphite are taken into consideration in the method. It was shown that an equivalent velocity of the wave in graphite is increased by the irradiation, and that a signal height of a propagated waveform is increased by the irradiation, and it decreases with increasing porosity caused by the oxidation. The Young's modulus for an ideal graphite polycrystals without pore was evaluated by considering the wave velocity in them in order to evaluate the change of the apparent modulus at simultaneous irradiated and oxidized conditions as an application of the developed method. It was also shown that the oxidation-induced change of the modulus is appropriately evaluated by the method, suggesting that it is possible to evaluate the change for the irradiated conditions. It can be said from this study that the developed method is promising to evaluate the oxidation damage on graphite components in the HTGRs by nondestructive way.

KEY WORDS: ultrasonic wave, propagation, UT, signal, graphite, pore, microstructure, Young's modulus, irradiation, oxidation, nondestructive evaluation, HTTR, HTGRs, in-service inspection

INTRODUCTION

Graphite materials are used for core internal structural components of the high-temperature gas-cooled reactors (HTGRs) because of their excellent heat resistibility. They will be gradually damaged by neutron irradiation and oxidation due to small amount of impurities in the primary coolant gas. Even in normal reactor operation, the oxidation damage is one of the crucial factors to degrade mechanical properties of graphite. The oxidation increases porosity of graphite and, hence, results in the degradation. It is necessary for the graphite components in the HTGRs to confirm their structural integrity by an in-service inspection (ISI). For the High Temperature Engineering Test Reactor (HTTR) [1] in the Japan Atomic Energy Research Institute (JAERI), the ISI is to be carried out by surveillance test specimens and a TV camera. Mechanical properties of graphite are inspected by the specimens loaded in the core and surface conditions of the components are also checked visually by the TV camera which resists radioactivity [2].

Ultrasonic testing (UT) is a mature non-destructive inspection technique and is applicable to porous ceramic materials including graphite [3-8]. Inner porous conditions in the porous body can be estimated by an analysis of the UT signals [9], since wave propagation characteristics in them highly depend on their inner porous conditions. In our previous study [8], it was shown that a wave propagation analysis model [9] for the porous materials taking account of wave-pore interaction process is applicable to graphite in order to estimate inner porous condition change caused by the oxidation. It was also shown that the Young's modulus of oxidized graphite can be appropriately evaluated by the propagation model, provided that change of a pore shape is taking into account [8]. We, then, proposed an advanced inspection method for the graphite components in the HTGRs to evaluate the change of the porous conditions from the ultrasonic wave propagation characteristics. The method has a potential to evaluate the oxidation damage on mechanical properties of the graphite components from the UT signals. This non-destructive approach is thought to be a promising technique in the future to support a reliability of obtained data in the surveillance test and/or the TV camera inspection in the ISI.

However, the Young's modulus of graphite, evaluated from an apparent sound velocity of the ultrasonic wave, is increased by the neutron irradiation [10]. It suggests that the propagation characteristics in graphite are affected by the irradiation in addition to the porous conditions. For the graphite components in the HTGRs, irradiation effects on the propagation characteristics should be, therefore, considered to estimate the oxidation-induced mechanical property change from the UT signals. In the present study, the irradiation effects on the propagation characteristics are analytically evaluated by considering the change of the modulus and the porosity. Design database of IG-110 graphite for the HTTR [11] is used in the evaluation. As an application of this study, the oxidation induced Young's modulus change at the neutron irradiated conditions is estimated in this method.

ANALYSIS

Wave Propagation Analysis

The wave propagation characteristics in the porous body had been analyzed by a propagation model [9] taking account of wave-pore interaction process. In the model, it is assumed that spherical pores with a radius r are located homogeneously in the body. If a wave comes into collision with a pore, it will go forward creeping through the pore edge with some probability as shown in Fig.1 [9]. The creeping wave has a time delay in comparison with a direct wave without collision. Ultrasonic waves are propagated into the body through interactions with a great number of pores. The propagation characteristics are analyzed by a statistical method with cumulating of the time delay and the collision probability. The propagated waveform is expressed by the Gaussian function with a height of H as follows [9]:

$$H = 1/(t_w\sqrt{2\pi}) \quad , \tag{1}$$

$$t_w = [3\phi rL\{1-\pi(4\pi/3\phi)^{-2/3}\}]^{1/2} \times (\pi\beta/\alpha - 2)/(4V_i) \quad , \tag{2}$$

$$\alpha = V_c/V_p \ , \tag{3}$$

$$\beta = 4L_p/(2\pi r) \quad , \tag{4}$$

where L is a propagation length, V_i a sound velocity in an ideal polycrystals without pore, ϕ a porosity, V_c and V_p respectively velocities of the creeping and direct waves and $4L_p$ a perimeter of the pore. As a result of this propagation analysis, the Young's modulus E of the porous body is evaluated as a normalized value by that of the ideal polycrystals E_i [9].

$$E/E_i = (1-\phi)/\{1+3\phi(\pi\beta/\alpha-2)/8\}^2, \tag{5}$$

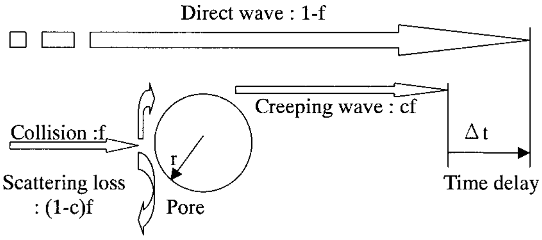

Fig. 1 Ultrasonic wave propagation model in porous body [9].
Probability f is for wave-pore collision and c is for wave creeping around pore.

The present authors had shown the above propagation model is applicable to the IG-110 graphite [12], a fine-grained isotropic graphite. Typical mechanical properties of the IG-110 are listed in Table 1 [12]. Its porosity 0.212 was calculated from its apparent volume density 1.78×10^3 (kg/m^3) and the theoretical density of graphite 2.26×10^3 (kg/m^3). A value of $\beta/\alpha = 1.41$ in Eqs.(1)-(5) is available with assuming a spherical pore shape at un-oxidized condition of the IG-110. For an analysis for the oxidized graphite, the value should be varied according as the oxidized conditions in order to take the pore shape change into account [8].

Table 1 Typical mechanical properties of IG-110 graphite [12].

Density (kg/m^3)	1.78×10^3
Porosity	0.212
Mean grain size (ƒ Êm)	20
Young's modulus (GPa)	9.8
Tensile strength (MPa)	24.5

Irradiation Effects on Young's Modulus

Since the dynamic Young's modulus of graphite is increased by the neutron irradiation, it can be used to investigate the irradiation effects on the propagation characteristics. For the IG-110 at irradiated above 400 ^{0}C, for example, it shows a great increase at relatively low neutron fluence levels and then continues to slightly increase with increasing the fluence. The initial great increase is explained by an irradiation induced pinning of dislocations in graphite crystals [10]. It leads to increase of the modulus of the crystals which results in the increase of the apparent modulus. The irradiation also causes bulk shrinkage of graphite which affects on the modulus [10]. The shrinkage causes a decrease of the porosity of graphite, and then leads to increase in the modulus. The change of the modulus is described as : E'/E_0, where E_0 and E' are respectively the modulus before and after the irradiation. The irradiation effects on the change are expressed as follows [13].

$$E'/E_0 = f_1(Dimension) \times f_2(Pinning) \tag{6}$$

where f_1 and f_2 are terms of the effects for the irradiation-induced dimensional change and for the pinning of the dislocation, respectively. Since they will affect the wave propagation characteristics in the irradiated graphite, they are respectively taken into consideration to study the irradiation effects on the propagation characteristics.

Young's Modulus of Ideal Graphite Polycrystals for Oxidation Damage Evaluation

If the oxidation occurs at the irradiation conditions, Eq.(6) is rewritten using an additional term of the oxidation damage f_3 as follows [13].

$$E'/E_0 = f_1(Dimension) \times f_2(Pinning) \times f_3(Oxidation)\ . \tag{7}$$

If the irradiation effects of f_1 and f_2 at an arbitral neutron fluence level are determined, the change of the modulus of the irradiated graphite can be evaluated by considering the oxidation term f_3. Although the oxidation damage can be evaluated by the wave propagation analysis by Eq.(5), the ideal value E_i is for the un-irradiated conditions. If the ideal value for an arbitral neutron fluence level is determined, the change of the modulus can be evaluated by the propagation analysis.

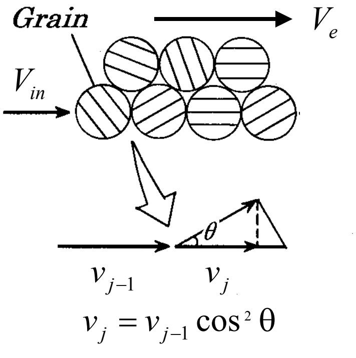

Fig.2 Wave propagation model in ideal graphite polycrystals.

We proposed an analytical model, shown in Fig.2, to determine the ideal value E_i for graphite polycrystals at the un-irradiated conditions. In the model, the polycrystals are considered to be an aggregate of identical crystals of which equivalent Young's modulus is E_e. The modulus for the polycrystals is decided with considering the wave propagation manner in them. It is assumed when a wave comes into the crystal with a velocity v_{j-1}, it is deflected mainly to the direction of the basal plane of the crystal which has an angle θ to the initial propagation direction as shown in the figure. The velocity of the deflected wave is described as $v_{j-1}\cos\theta$. The magnitude of vector component of it to the initial direction is shown as v_j :

$$v_j = v_{j-1}\cos^2\theta\ . \tag{8}$$

The each grain has an arbitral angle θ , the equivalent velocity V_e for the polycrystals is then shown as:

$$V_e = k_1 V_{in} \int_{-\pi/2}^{\pi/2} \cos^2 \theta d\theta \quad , \tag{9}$$

where V_{in} is an initial velocity in the polycrystals and k_1 is a constant. Since the Young's modulus is proportional to the square of the velocity of the wave, the modulus for the ideal polycrystals is obtained by:

$$E_i = k_2 E_e (V_{in} / V_e)^2 \quad , \tag{10}$$

where k_2 is a constant. We had obtained the E_i value as 17.5 GPa for the ideal graphite polycrystals at the un-irradiated conditions. It is roughly comparable to the value of 18.1 GPa estimated empirically [14]. The value for the irradiated conditions is to be decided based on this value with considering the pinning components.

RESULTS AND DISCUSSION

Irradiation Effects on Wave Propagation

For the IG-110, design databases [11] on mechanical properties at irradiated conditions for the HTTR was established through many experiments. Figure 3 shows the design data for the irradiation-induced dimensional change and an analytical result on the equivalent velocity estimated from the dimensional change. The change of the velocity is expressed as a ratio V'/V_0, where V_0 and V' are velocities before and after the dimensional change. The design data at irradiation temperatures of 800, 1000 and 1200 ^{0}C were chosen for this analysis to study irradiation temperature dependency. To evaluate the velocity change, the dimension term f_1 of the Young's modulus change in Eq.(6), which is considered to be proportional to the square of the velocity and a density, was analyzed from the dimensional change by Eq.(5). It should be noted here that the pinning component was not included in the present analysis. It can be seen from this figure that the change of the velocity almost linearly increases with increasing the neutron fluence. At the irradiation temperature of 1200 ^{0}C, the largest dimensional change caused the maximum decrease of the porosity and it resulted in the maximum increase of the velocity. By applying the wave propagation analysis to the dimensional change, the dimension term of the modulus change was estimated, and then the change of the equivalent velocity caused by the dimensional change was evaluated.

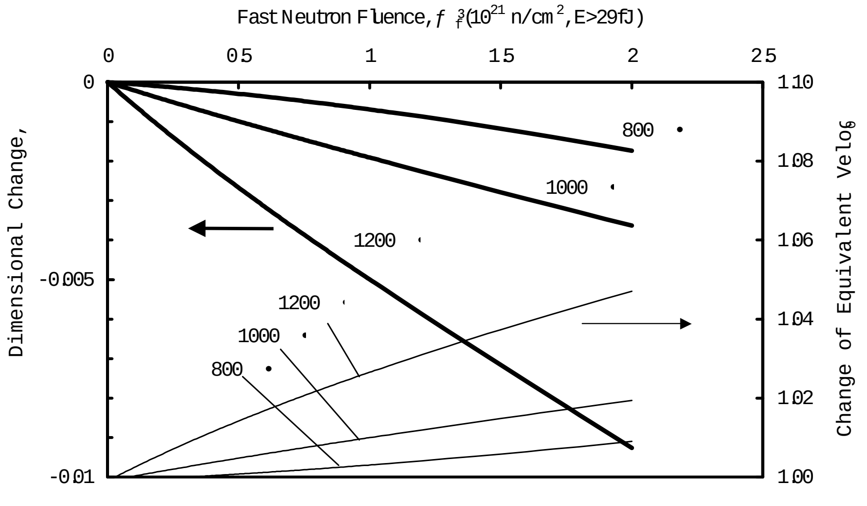

Fig.3 Analytical result of change of equivalent velocity caused by dimensional change.

The pinning term f_2 in Eq.(6) was evaluated by the database [11] on the irradiation-induced Young's modulus change and by the analytical result of the dimension term f_1. By the pinning effect, the equivalent velocity is thought to be changed. For the ideal polycrystals without pore, the change of the velocity is described as V_i'/V_i, where V_i and V_i' are velocities for before and after the irradiation. Figure 4 shows the change of the Young's modulus and analytical result of the change of the equivalent velocity as a function of neutron fluence. The design database at 800, 1000 and 1200 ^{0}C irradiation was also used. The change of the velocity rapidly increases at low neutron fluence level, and then is almost saturated within the analyzed fluence level. The change decreases with increasing irradiation temperature. It suggests an annealing effect on the pinning. The change of the velocity by the pinning is quite larger than that by the dimensional change. It suggests that the contribution of the pinning term f_2 on the change of the Young's modulus is quite larger than that of the dimensional term f_1.

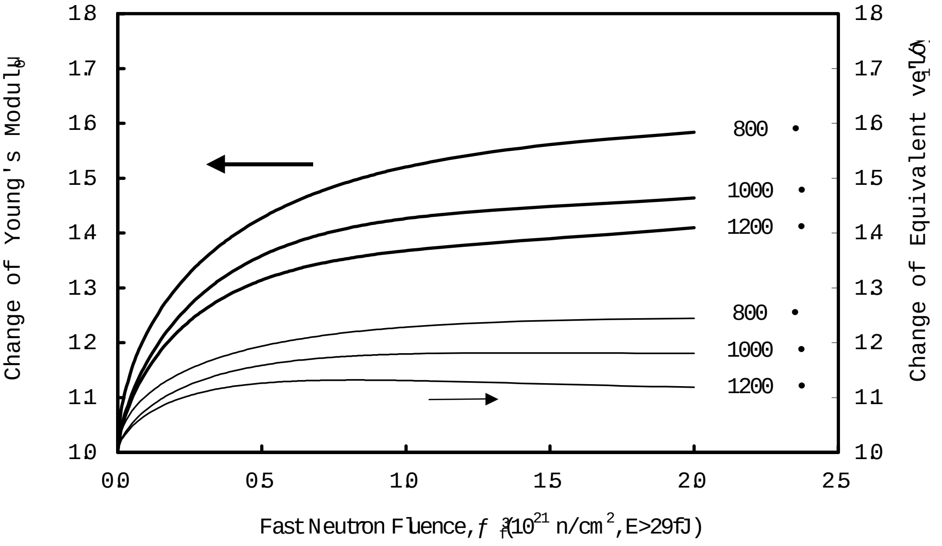

Fig.4 Analytical result of change of equivalent velocity caused by pinning of dislocations.

The irradiation causes the dimensional change and the pinning of the dislocation. The former effect on the wave propagation was evaluated by considering the porosity change in Eqs.(1)-(5). The latter effect was evaluated by using the velocity V_i' and the Young's modulus E_i' for the ideal polycrystals at the irradiated conditions in the equations.

Oxidation Effects on Wave Propagation

The oxidation causes a change of the porous conditions in graphite. It leads a change of the wave propagation characteristics, and then result in a change of the propagated waveform. If the oxidation occurs at the irradiated conditions, the porous condition change should be evaluated in addition to the irradiation effects on the wave propagation. The maximum signal height H' for the simultaneous oxidized and irradiated conditions was evaluated by Eqs.(1)-(4) considering the porous condition change and the irradiation effects. The change of the height is described as a gain G as:

$$G = 20Log_{10}(H'/H_0), \tag{11}$$

where H_0 is a height at un-irradiated and un-oxidized conditions. Figure 5 shows an analytical result of the gain as a function of porosity at a fast neutron fluence 1.0x10^{21} n/cm^2. At the porosity of 0.212 corresponding to the nominal value of the IG-110, if there is no pinning effect, the gain will result in G=0. The increase of the gain at this porosity is caused by the pinning. The lower temperature irradiation caused the larger gain. The gain decreases with increasing the porosity suggesting that the signal height decreases with increasing the oxidation damage.

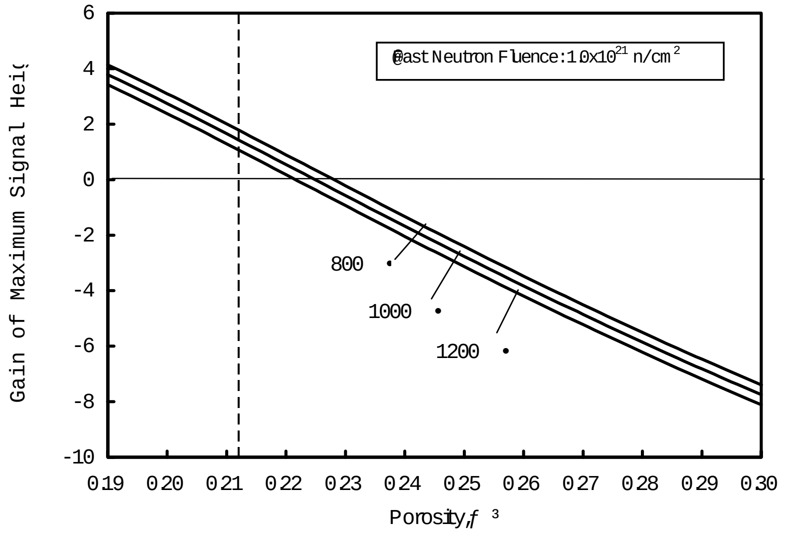

Fig.5 Gain of maximum signal height of ultrasonic wave as a function of porosity at fast neutron fluence of 1.0x10 ^{21} n/cm^2. Nominal porosity of IG-110 is 0.212.

Oxidation Damage Evaluation

As an application of the developed method for the analysis on the propagation characteristics, we tried to evaluate the change of the Young's modulus at simultaneous oxidized and irradiated conditions. The change at a fast neutron fluence of 1.0x10^{21} n/cm^2 was evaluated by Eq.(5) with the ideal modulus Ei=17.5 GPa. The irradiation effects and the oxidation damage were taken into consideration as the same way for the signal height analysis. For the each irradiation temperature, the modulus was estimated from E_i' and the change of the equivalent velocity caused by the pinning shown in Fig.4. The modulus E_i' was respectively evaluated as 26.4, 24.3 and 22.4 GPa for the irradiation temperature of 800, 1000 and 1200 ^{0}C. Figure 6 shows an analytical result of the Young's modulus of the IG-110 as a function of porosity. Experimental results [15] on the modulus of the IG-110 for the un-irradiated conditions are also plotted. In the experiment, the porosity was changed by forced oxidation, i.e. Burn-off. For a plot of the experimental data, since the data were given as a ratio of before and after the Burn-off, we estimated the value for after the Burn-off using the nominal value of 9.8 GPa for the as-obtained IG-110. It can be seen from this figure that the modulus was increased by the neutron irradiation, and that it decreases with increasing the porosity. The analytical curves are composed of two regions. At the lower porosity region, the pore shape was treated as a sphere, because it was shown that pore shape of the as-obtained IG-110 of which porosity is 0.212 can be regarded as a sphere in the propagation analysis [8]. On the other hand, at the higher porosity region, the pore shape change by oxidation was considered by changing β/α value in Eqs.(1)-(5). The relationship between the β/α and the porosity for the IG-110 at the oxidized conditions [8] was obtained by modifying the relationship for alumina [16]. The analytical result for the un-irradiated conditions is in quite good agreement with the experimental data, suggesting that the oxidation-induced Young's modulus change of irradiated graphite will be evaluated by this method.

By the developed method in this work, the porosity of irradiated graphite will be estimated by measuring the ultrasonic wave propagation characteristics. If oxidation damage is considerable magnitude, it is possible to estimate oxidation-induced mechanical property change through measuring the propagation characteristics. This new approach is a promising to evaluate the oxidation damage on graphite components in the HTGRs by nondestructive way and to support the reliability of the test results of the ISI. Although, in the present work, the design database for the IG-110 on the irradiation-induced dimensional change and Young's modulus change were used to establish the new evaluation method, they should be measured directly by in-core graphite materials for a real application. It is because that the design database was established by many experiments which contain some deviations and that they are strongly affected by in-core environment. They can be measured by the surveillance specimens in the core, for example. At the present stage of this study, the wave propagation analysis is applicable only for the uniform condition of pore-size and pore distribution in the porous body, an advanced model which can treat the un-uniformity of them are necessary for a more precise analysis in the future.

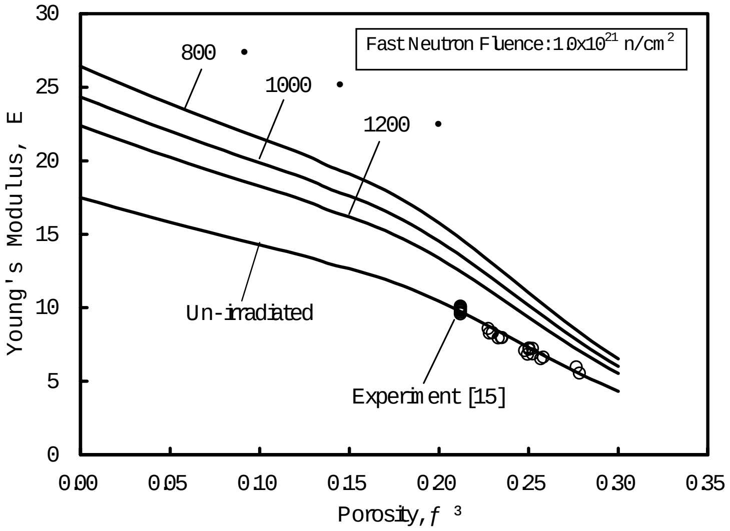

Fig.6 Analytical results of Young's modulus change as a function of porosity at fast neutron fluence of 1.0x10^{21} n/cm^2.

CONCLUSIONS

In order to evaluate the oxidation damage on the graphite components in the HTGRs at the neutron irradiated conditions, a new analytical method by the ultrasonic wave propagation characteristics was developed. The irradiation effects, the dimensional change and the pinning of the dislocations, on the wave propagation characteristics in graphite are taken into consideration in the method. It was shown that the equivalent velocity of the wave is increased by the irradiation, and that the signal height of the propagated waveform is increased by the irradiation and it decreases with increasing the porosity caused by the oxidation. For an application of the developed method, the ideal Young's modulus for the ideal graphite polycrystals was evaluated by considering the wave velocity in them in order to evaluate the change of the apparent modulus at the simultaneous irradiated and oxidized conditions. It was also shown that the oxidation-induced change of the modulus is appropriately evaluated by the method, suggesting that it is possible to evaluate the change for the irradiated conditions. It is concluded from this study that the developed method is promising to evaluate the oxidation damage on graphite components in the HTGRs by nondestructive way. Further investigation is necessary to develop an advanced model to treat un-uniformity of the porous conditions.

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NOMENCLATURE

E = Young's modulus
f_1 = dimension component
f_2 = pinning component
f_3 = oxidation component
G = gain
H = signal height
k_1, k_2 = constant
L = wave propagation length
$4L_p$ = perimeter of pore

r = pore radius
V, v_j, v_{j-1} = wave velocity
V_c, V_p = wave velocity of the creeping and incident waves
V_e = equivalent wave velocity in ideal polycrystals
V_{in} = initial wave velocity in ideal polycrystals
α = V_c/V_p
β = $4\ L_p/(2\bullet r)$
ϕ = porosity
θ = angle of basal plane to initial wave direction
Subscripts
0 = value for un-irradiated and/or un-oxidized conditions
i = value for ideal polycrystals without pore
Superscripts
’ = value for irradiated and/or oxidized conditions

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