Key-Hole Notches in Isostatic Graphite: A Review of Some Recent Data

Filippo Berto, A. Gagani, Relly Victoria V. Petrescu and Florian Ion T. Petrescu

Department of Engineering Design and Materials, NTNU, Trondheim, Norway
ARoTMM-IIToMM, Bucharest Polytechnic University, Bucharest, (CE) Romania

Abstract: Fracture of the breakable isostatic graphite is concerned using the experimental and theoretically, the plates containing nicks holes key subject to varying degrees of mixed task. The main purpose of this work is a double one. In the first place, in order to offer a new set of experimental results on fracture of samples of graphite scored, with different values of the load mixed and radii notch and which may be helpful for researchers, as it enlarges the very limited data available; and secondly, to provide a criterion fracture of the polycrystalline graphite under the conditions mentioned above. The main purpose of this work is to offer a new set of experimental results (70 new data) on the fracture of the samples of the loose graphite of key holes, the different values of mixed loading, the tilt angle and radii notch and which may be of help as widens the very limited data available. By using the value of the average density of the stem of energy in a well defined, a criterion of fracture of the polycrystalline graphite under the conditions referred to above, it is proposed to predict the static resistance of the samples taken into account. The third part of the work deals with the analysis of the direction of initiation of fracture and spread of the crack in the early. The average value of the stem density of energy in a well defined is used to predict the static resistance of the samples taken into account. Good agreement is found between the experimental data for the tasks critical failure and the theoretical predictions based on average constant strain density of energy on the volume of the material.

Keywords: Graphite, Isostatic Graphite, Fracture, Polycrystalline Graphite, Notched Components, Nuclear Energy Application

Introduction

Several researchers have studied in the resistance of the fracture of the previous polycrystalline graphite be under the module I (opening cracks) or in mixed mode I/II (cracks opening-slip) load conditions.

For example, Awaji and Sato (1978) were among the first researchers who used the cracker brazilian disc (BCD) specimen in order to study the mixed mode I/II fracture toughness of two materials polycrystalline graphite test medium.

The CD is a test specimen of a circular form in which a crack the center is generated for measuring fracture stiffness of fragile materials.

Yamauchi et al. (2000; 2001), use also made of specimens of the disc type (e.g., CBD specimen and kinks () circular specimen semi SCB subjected to three points of bending the charging) and investigated the mixed mode fracture I/II tenacity graphite.

Other specimens of the test have been used for the Exploration of the behavior of materials fracture polycrystalline graphite. For example, a single notch on the edge of bending specimen (Li et al., 1999) and the three points of sandwich bending specimen (Shi et al., 2008) are two other types of specimens used in the past for testing fracture on graphite.

In another attempt, Etter et al. (2004) have investigated I fracture KIC tenacity of an isotropic graphite polycrystalline porous, in addition to the composite graphite/aluminum through one edge of the bushing specimen beam. The most extensive applications of the fibers of graphite in composite materials such as the composite graphite epoxy), were determined also some researchers to study the behavior
of the breaking strength of these materials under the module I simply mix mode I/II loading conditions (Yum and You, 2001; Sukjoo and Bhavani, 2007; Wosu et al., 2005; Jurf and Pipes, 1982).

Lomakin et al. (1975) made use of an energy release rate criterion for analyzing the fracture initiation in cracked graphite specimens under pure mode I loading.

There is also a set of criteria for the fracture in the specialized literature to predict debut in mix mode fracture of the fragile I/II, in various materials of engineering, such as graphite.

Maximum voltage Tangential (MTS), the criterion (V.R.: They are described and a little hunting, 1963), the density of the minimal energy stem (SED) criterion (A little hunting, 1974) and the rate of release of the maximum energy or the criterion G (Hussain et al., 1974) have been more frequently used by the researchers.

Using a criterion of MTS modified, Ayatollahi and Aliha (2008) have submitted the best estimates for the Debut fracture mixed mode into two classes of graphite polycrystalline containing cracks sharp.

Are generated cracks in graphite, mainly due to the faults of manufacture or because of the coalescence micro-pore structural or other defects, which are inherently incorporated in the graphite.

Whereas the cracks are regarded as unpleasant entities in most materials of engineering, however, the splines U or in the shape of a V are sometimes it is desirable in the design and manufacture of products manufactured from graphite.

Forms of graphite heating elements of the graphite and mandrel graphite are just a few examples for industrial components which contain U or slashes in the form of V.

An overview of the literature shows that in spite of extensive studies on how I and fracture of the mixed with specimens of the graphite the cracker, very few works were busy with the fracture of the fragile components of graphite scored-V.

Follows Ayatollahi and Torabi (2010) has recently made a series of tests by the fracture on three specimens of the test scored-V different, made of a material polycrystalline graphite. They have also proposed to a criterion environment stress and estimated the results of their experimental, with an accuracy very good. However, the results submitted by Ayatollahi and Torabi (2010) is confined to the pure mode loading conditions I.

There are various practical conditions, in the case where the slots in the components in graphite are subject to a combination of stretch and deformation to the shear bolt (or mix mode I/II of charge).

In a recent, the presence of the authors have investigated in mix mode fracture of the fragile graphite polycrystalline both experimental and theoretical level (Ayatollahi et al., 2011).

First of a series of experiments by the fracture have been carried out on samples of Brazilian disc cut centrally from the graphite to determine the tasks by the fracture in different combinations of how I and charge mode II.

Then, a theory based on the criterion of SED (Lazzarin and Zambardi, 2001; Lazzarin and Berto, 2005; Berto and Lazzarin, 2009; Berto et al., 2007; Gómez et al., 2007) was used to estimate the tasks of fracture of the experimentally obtained.

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Fracture Experiments

Details of the material in the graphite, the specimen testing and experiments by the fracture are given in this section.

Material

The tests by the fracture have been carried out on a commercial graphite polycrystalline isostatic.

The size of the average cereals has been measured using the SEM technical and density was determined by the method buoyancy.

The properties of the basic material of the graphite does tested are listed in Table 1: The size of the average cereals yield is 2 um, porosity of 7%, density in bulk 1850 kg/m³, the average value of resistance to the drawbar of 28.5 MPa, the module Young of 8,05 GPa and shear mode of 2,439 GPa. Resistance to compression is equal to 110 MPa, while the bending strength is 49 MPa.

| Table 1. Mechanical properties |
|-------------------------------|
| Material property             | Value  |
| Elastic Modulus E [MPa]       | 8050   |
| Shear Modulus G [MPa]         | 3354   |
| Poisson's Ratio ν              | 0.2    |
| Ultimate Torsion Strength [MPa]| 30     |
| Ultimate Compression Strength [MPa]| 110  |
| Ultimate Tensile Strength [MPa]| 46    |
| Fracture toughness [MPa m¹/²]| 1.06   |
| Hardness [Shore]              | 58     |
| Density [Kg/dm³]              | 1.85   |
| Porosity [%]                  | 7      |
| Resistivity [µΩm·m]           | 11     |
| Thermal Conductivity [W/(m·K)]| 11     |
All tests have been carried out under a load on a testing device servocontrolled MTS axial (±100 kN/±110 Nm, ±75 mm/±55°). The load has been measured by a cell MTS with error ±0.5% at full scale.

Geometry of the Specimens

As shown in Fig. 1, the sample used in the present investigation is a card that contains a central flaw in the keyhole with a radius of the notch ρ and the distance between the center of the holes. The specimen is subject to a request to the drawbar. By changing the angular β, different combinations of how I and II (or tension and the deflection of the shear bolt) may be produced for the key holes. When the load is applied along the bisectoarei notch (i.e., β = 0), the key holes are subject to deformation mode pure I. By increasing the angle β from zero the charge status is changed from pure mode I to the mixed mode II.

For all specimens graphite tested, the Width (W), the depth of the notch (A) and the thickness of the t was 50 mm, 10 and 10 mm respectively.

Five values of the radius notch ρ = 0, 25, 0, 5, 1, 2, 4 mm have been taken into account for the manufacture of samples of the test to ensure that the effects of the radius of the peak notch on the fracture Mix Mode of the copies of the graphite are studied. In order to obtain another mix mode, were taken into consideration four values of the angular β (β = 0, β = 30°, β = 45° and β = 60°).

In order to prepare the samples for testing of graphite, the first series of plates of 10 mm in thickness were cut off from a block of graphite. Then, specimens have been manufactured with precision, using a jet of water the debited 2-D CNC. Before carrying out the experiments, the cut surfaces of the copies of the graphite have been polished by grooves using fine sand paper to remove any possible concentrations of local stress due to the roughness of the surface.

A total number of 70 Mix Mode I/tests by the fracture II have been carried out for the different parameters of the geometry notch. In respect of each form of the geometry and the angle of loading, three tests by the fracture separate, have been carried out using a voltage of compression-multipurpose machine of testing in conditions of travel control, with a speed of 0.05 mm/min. curves relating to the task of travel recorded during the tests by the fracture were all linear and fractured specimens from a date. Therefore, the use of a criterion of rupture fragile, on the basis of the Linear Fracture Mechanics springs (LEFM) is permitted. The average values of the loadings among rupture (F) recorded by the test are given in Table of Fig. 2 for each specimen.

Fig. 1. Geometry of the specimens (all dimensions in mm)
A review of Table of the Fig. 2 shows that the fracture of the loading increases when the loading conditions change from pure mode I ($\beta = 0$) in relation to the mixed mode I II. In addition, the task of fracture of the increase in order to increase the peak-ray notch, irrespective of how the Joint Committee.

**SED Criterion**

In order to estimate the task of fracture in components of graphite scored, engineers have need of a criterion of suitable fracture developed on the basis of the mechanical behavior of the material around the top notch. In this section, a criterion on the basis of the stem of energy density is described by which the tasks by the fracture obtained from the trials may be estimated very well.

The criterion of average energy density stem (SED), as shown in Refs. (Lazzarin and Zambardi, 2001; Lazzarin and Berto, 2005; Berto and Lazzarin, 2009; Berto et al., 2007; Gómez et al., 2007) argues that there has been a up friable when the average value of the stem density of energy in a certain volume of control, shall be equal to a critical value $W_c$.

This critical value varies from one material to the material, but does not depend on the geometry of the notch and clarity. The volume of control shall be considered to be dependent on the resistance to the tensile and the resistance to tearing $K_{tc}$ where materials are fragile or almost fragile subjected to static loads.

In conditions of deformation plane critical length, $R_c$, can be evaluated in accordance with the following expression (Berto and Lazzarin, 2009):

$$R_c = \frac{(1+\nu) \cdot (5 - 8\nu)}{4\pi} \left(\frac{K_{tc}}{\sigma_t}\right)^2$$  \hspace{1cm} (1)

In the case in which it is $K_{tc}$ tear-resistant (fracture toughness), $\nu$ the Poisson’s ratio and $\sigma_t$ the ultimate tensile stress at tensile a specimen of the plain which shall not be subject to a linear behavior pad. This critical value may be determined by the resistance to tearing $\sigma_t$ under expression of Beltrami.

$$W_c = \frac{\sigma_t^2}{2E}$$  \hspace{1cm} (2)

In parallel, the definition of the volume of control by means of checking the $R_c$ needs to know and the resistance to tearing $K_{tc}$, Poisson ratio $\nu$, see Equation 1. The task of critical that is durable by a notched
component can be estimated by the imposition of $W$ equal to the critical value $W_c$, which is seen as a constant here in the way I, II and the way in flat conditions-mode joint committee. This assumption has been checked on wide for a number of different materials fragile and quasi-fragile components (Berto and Lazzarin, 2009; Berto et al., 2007; Gómez et al., 2007).

As we have mentioned earlier, material properties of graphite used in this investigation are: $\sigma_t = 46$ MPa, $K_{IC} = 1.0$ MPa.m$^{0.5}$, Poisson’s ratio $\nu = 0.2$. As a result, the SED critical for graphite to be tested is $W_c = 0.13$ MJ/m$^3$ and the radius of the volume of the control is $R_c \cong 0.17$ mm considering realistic conditions of deformation of the surface.

Dealing with the blunt nicks under tasks in the mix mode, the matter becomes more complex than in the manner of charging, mainly because the voltage maximum tensile strength is outside of the line bisector notch and its position varies depending on how I and mode of stress II distributions. The maximum voltage that appears along the edge of the circular hole has been calculated using the numeric code FE code ANSYS 12.0°. For all geometrical have been created two models.

The first model has been directed mainly to the determination of the point at which were localized maximum voltage of the master and the SED maximum; the second model has been more refined, with a precise definition of the volume control in the case in which should be on average energy density stem.

All the analyzes have been carried out through the use of the elements of the eight nodes in the hypothesis of conditions the stem plane.

The table of Fig. 2 summarizes the contours of the experimental results, numeric and theoretical for copies of graphite tested with four different angles tilt $\beta = (0^\circ, 30^\circ, 45^\circ, 60^\circ)$ investigated in research.

In particular, this table summarizes the theoretical level ($F_{th}$) and the load average experimental to failure ($<F>$) for each angle of loading $\beta$ and notch $\rho$ radius. The table also provides the maximum value of the principal stress ($\sigma_{max}$) and the SED value, obtained from FE models of specimens of graphite by the application of the average value of the stress critical. It is interesting to note that the maximum principal stress along the notch edge is much greater (approximately twice) than the ultimate tensile stress of the material which can justify the approach based on the average value of SED over a volume control.

As it can be seen, the agreement between the experimental results obtained for specimens of graphite and the theoretical predictions based on a constant value of the energy from the local deformation is satisfactory, with a maximum gradient equal relative by 12%.

### Crack Initiation Angles

The approach angles of initiation of the cracks, they have been experimentally measured through the use of a microscope and a software called Las dedicate Leica Application Suite.

The average value of the measured angles are compared in the table of Fig. 3 with those obtained by numerical analysis for the identification of the point along the edge of the in the case of SED and then the maximum voltage reaches critical.

![Fig. 3. Experimentally observed angles compared with numerical values](image-url)
Applications

Graphite has many applications. A new one is in nuclear reactors (Petrescu and Calautit, 2016a; 2016b; Petrescu et al., 2016).

Spectral reactors require a neutron moderator. Graphite has qualities that make it particularly suitable for this application: It is a moderator that is both low neutron absorber, refractory and not very vulnerable to corrosion, inexpensive and well known to industrialists because of its other applications.

Graphite has been used since the beginning of nuclear power and it will remain an essential material for future gas-cooled reactors. Its importance as a nuclear material is still worthy of research. Its implementation and its reactor behavior make it a very special material.

There are many forms of carbon (vitreous carbon, coke, anthracite, pyrocarbon, carbon black, carbon nanotubes, fullerenes), but only two are allotropic: Diamond and graphite.

If it is sufficiently pure, graphite is a good neutron moderator because it slows down the neutrons without absorbing them (the effective cross-section of 12C is small and its cross-section of elastic scattering is strong). It possesses interesting mechanical properties at high temperature, is relatively easy to machine and activates little under irradiation.

A nuclear graphite must have good mechanical properties, hence a high density; It must have good dimensional stability under irradiation, therefore good isotropy; Finally, it must capture the neutrons as little as possible and constitute a waste after irradiation with the lowest activity possible, thus containing very small quantities of absorbing or activatable impurities.

Polycrystalline graphites are used as structural materials in gas-cooled nuclear reactors with a thermal neutron spectrum. They are manufactured from petroleum coke or coal tar pitch and a binder. The calcined coke is ground, sieved and then the grains obtained are mixed in proportions suitable for obtaining a good density and favoring the departure of the volatiles from the binder. The mixture of coke is generally kneaded at 165°C, with a coal tar pitch, shaped by spinning, or by compression either unidirectional or isostatic and then fired at between 800°C and 1200°C, to coke the binder. Thereafter, the product may undergo one or more impregnations, generally with petroleum pitch, in order to increase its density and its mechanical properties. Finally, it is graphitized between 2500°C and 3000°C, to obtain the hexagonal crystalline structure. This graphitization takes place in the presence of purifying agents (NaF, MgF₂, Cl₂,...), which make it possible to obtain a graphite of nuclear quality with a low content of impurities.

The nature of the coke used and the technique of shaping retained are very important because they determine the isotropy of the graphite obtained and therefore the evolution of its macroscopic properties under irradiation. At equivalent grain size, petroleumokes are generally more anisotropic than coal pitch cokes. However, the smaller the coke grain size the more the graphite obtained is isotropic. Anisotropic or quasi-isotropic graphites are produced by spinning or by uniaxial compression, while isotropic graphites are manufactured by isostatic compression.

The irradiation of graphite by fast neutrons causes the displacement of carbon atoms out of their equilibrium position, creating defects (interstitials and gaps). This phenomenon results in an accumulation of energy, called "Wigner energy". The stored energy can reach 2000 J.g⁻¹, a considerable value whose release by return of the equilibrium atoms would raise the graphite temperature from ambient to 1200°C. This accumulation of energy has constituted a potential fire hazard for air-cooled reactors operating at low temperatures. The lack of prevention can lead to an accident, such as that in the experimental reactor at Windscale, England in 1957.

When the irradiation temperature is less than 120°C, the isolated defects (1 to 4 atoms) are not very mobile and the stored energy accumulates rapidly. These defects can be cured by raising the temperature. The recombination of the defects is accompanied by a release of heat, represented by a differential enthalpy "peak" (dH/dθ), located at about 200°C and which may exceed the specific heat of the non-irradiated graphite. An irradiated graphite is energetically stable if for any temperature the differential enthalpy is lower than the specific heat of the non-irradiated graphite. If the graphite is heated, the energy begins to be released from the threshold temperature θ₁. As soon as the tripping temperature θ₂ is reached, the temperature rises adiabatically, causing a spontaneous release of heat, to a final temperature θ₃.

In order to show a thermal instability of the graphite due to the Wigner energy, the following double condition must be fulfilled:

**Conclusion**

The SED criterion has been extended to the fields cut rounded with the tip, in order to estimate the breaking load of the components of graphite dowels. It has been demonstrated that the proposed method is suitable for the isostatic graphite highlighted in the terms and conditions how to load joint venture, being the experimental results in accordance with the foreseeable results by addressing SED. Of the sound between the results of theoretical and experimental, it may be deduced directly that for graphite isostatic critical energy and the radius of the volume of control are both material properties constant and independent of the charge mode and can be simply evaluated from a way I just test.

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In order to show a thermal instability of the graphite due to the Wigner energy, the following double condition must be fulfilled:
An irradiation temperature of less than 115°C.
A neutron fluence greater than 1,6E\(^{20}\) n.cm\(^{-2}\) \(\Phi\)FG, i.e., 0.11 displacement per carbon atom.

In practice, for graphite irradiated between 30°C and 120°C, most of the energy stored is concentrated in the “peak” located at about 200°C. The importance of this peak decreases when the irradiation temperature increases. When it is above 170°C, the Wigner energy peak at 200°C disappears. This reflects the fact that, at high temperature, the irradiation defects in the graphite do not accumulate, as they recombine as they are formed.

Thus there is no risk of spontaneous release of Wigner energy for graphite irradiated above 300°C.

**Behavior of Graphite under High Temperature Irradiation**

In future high temperature reactors, graphite will be irradiated between 500°C and 1200°C depending on the components considered, i.e., at temperatures well above those to which it is irradiated in the MAGNOX, UNGG and AGR reactors. As mentioned above, Wigner energy will not be a problem for this type of reactor, but other phenomena affecting graphite will have to be taken into account.

**Dimensional Variations**

At the scale of the crystallites (whose sizes according to the crystallographic axis c (Lc) are between 20 nm and 140 nm) assimilable to single crystals, the fast neutron flux generates the displacement of carbon atoms in the interstitial position between the graphene planes and gaps within these same planes. The accumulation of the gaps will lead to a contraction of the crystalline mesh along the axis a and that of the interstitials to a dilatation along the axis c. Under irradiation, the size of the crystallites according to a (La) will therefore decrease while Lc will increase. The increase of the irradiation temperature leading to an increase in the mobility of the defects, the concentration of isolated interstitials and vacancies decreases and leads, with fluence, to increasingly small changes in La and Lc. Obviously the dimensional variations of the polycrystalline graphite are not limited to those of the crystallites and depend essentially on the following parameters:

- The temperature of irradiation: Between 300°C and 700°C, contraction occurs in the two preferential directions of the polycrystalline graphite (parallel and perpendicular to the graphite planes) with greater deformations in the direction parallel to the grains. The deformation rates as a function of the fluence as well as the dimensional variations decrease with the increase of the temperature of irradiation. Beyond 700°C, there is also a contraction in both directions, but then the rates of deformation increase with the irradiation temperature.

The size of the crystallites: The size and the perfection of the crystallites increase with the graphitization temperature. However, the greater the size of the crystallites, the better the dimensional stability of the graphite under irradiation.

The isotropy of graphite: In general, the deformation velocities as a function of the neutron fluence and the dimensional variations are all the smaller as the graphite is isotropic.

**Thermal Conductivity**

During the irradiation, more or less extensive defects are created in the crystallites, leading to a rapid decrease of the mean free path of the phonons. Thus, the evolution of the thermal conductivity of crystallites is mainly related to the concentration of isolated gaps and loops of gaps. The degradation of the thermal conductivity of polycrystalline graphite thus appears at very low neutron fluences (10\(^{18}\) n.cm\(^{-2}\) (E>0.1 MeV)).

At a given temperature and regardless of the grade of graphite, the thermal conductivity decreases monotonically with fluence at a rate that decreases as fluence increases. It reaches a saturation value from 4.10\(^{15}\) n.cm\(^{-2}\) (E>0.1 MeV) for irradiation temperatures between 500°C and 1000°C.

At a given fluence, the lower the thermal conductivity of the graphite under irradiation, the lower the irradiation temperature. Thus, at 1200°C for a fluence of 10\(^{18}\) n.cm\(^{-2}\), the normalized conductivity \(\left(\lambda_1/\lambda_0\right)_{1200°C}\) is close to 1.

**Elasticity Module**

Under irradiation, the Young modulus (E) of the polycrystalline graphite increases very greatly, due to the blocking of the shear deformations by the interstitial defects, which can lead to embrittlement of the material. This increase occurs at low fluences and it is all the more important that the irradiation temperature is low. Indeed, when the irradiation temperature increases, the mobility of the defects increases, the isolated interstitials are organized in clusters and then in portions of new graphene planes. The shear deformations are then less and less thwarted, which has the effect of limiting the increase of Young's modulus.

For quasi-isotropic graphites irradiated above 300°C and 3.10\(^{21}\) n.cm\(^{-2}\) (E>0.1 MeV), the Young modulus values remain constant up to 9.10\(^{21}\) n.cm\(^{-2}\) (E>0.1 MeV). For higher fluences, their E values increase further due to closure of the porosity. Finally, starting from 1.5.10\(^{22}\) n.cm\(^{-2}\), there is a sudden decrease in E, linked to the generation of a new porosity, corresponding to the transition from the contraction phase to that of expansion of the graphite.
Creep Under Irradiation

While the thermal creep of the graphite appears only above 2000°C, the creep under irradiation occurs at 100°C and leads to deformations which may be ten times greater than those obtained outside irradiation. The creep deformations under irradiation $\varepsilon_f$ of the graphites are written in the form:

$$\varepsilon_f = \frac{\sigma}{E_0} \left(1 - e^{-\frac{\sigma}{k\gamma}}\right) + k\sigma \gamma$$

(3)

where, $\sigma$ is the stress, $E_0$ the Young modulus of the graphite and $\gamma$ the neutron fluence.

The primary and secondary creep deformations correspond respectively to the two terms of the equation. They are both proportional to the applied stress $\sigma$:

"Primary" creep which intervenes only at low fluences and during which the rate of deformation of the graphite decreases continuously. During this step, the suppression of the applied stress, while continuing the irradiation, causes cancellation of the deformation $\varepsilon_f$. Thermal annealing leads to identical restoration. There are few measurements of the primary creep constant $b$ and these are fairly dispersed. Nevertheless, it seems that this constant increases with the temperature of irradiation.

"Secondary" creep refers to a stationary state and is characterized by a constant deformation rate as a function of neutron fluence. The deformations generated during this step are permanent and cannot be canceled by suppressing the applied stress. For irradiation temperatures between 500°C and 1400°C, the secondary creep constant $k$ increases with temperature.

Graphite Corrosion

If it exhibits good mechanical behavior at high temperature, graphite is also very sensitive to the presence of oxidizing species in helium. The oxidation of graphite produces gaseous species (CO, CO$_2$, H$_2$, ...) functions of the oxidizing gas and is accompanied by a degradation of the material, which in extreme cases could have an impact on the safety of the installation. It is envisaged to introduce traces of oxidant (water) in the helium gas to control the chemistry of the coolant in the reactors RHT and RTHT. These oxidizing species have in particular the objective of maintaining a protective film on the surface of the metallic materials. Moreover, accidental scenarios in this type of reactor envisage a massive air intake into the primary circuit, for example following the rupture of the "hot-duct", the hot pipe connecting the two tanks in the current drawings. It is therefore very important to know the behavior of graphite in the presence of oxidizing species, whether in normal operation or in an incidental situation (Bonal and Robin, 2005).

Author’s Contributions

All the authors contributed equally to prepare, develop and carry out this manuscript.

Ethics

This article is original and contains unpublished material. The corresponding author confirms that all of the other authors have read and approved the manuscript and no ethical issues involved

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