FINITE ELEMENT ANALYSIS OF THE TEMPERATURE DISTRIBUTION INSIDE THE DIAMOND SYNTHESIS CHAMBER WITH DIFFERENT CALCITE RINGS[1]

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Abstract

In this work the objective was to evaluate by finite elements simulation the influence of the shape of calcite rings on the temperature distribution inside the high pressure chamber for the synthesis of diamond. A model of the high pressure and high temperature arrangement for synthesizing diamond crystals was simulated by a computer program based on finite elements. Using this simulation, the temperature distribution inside the high pressure chamber was determined for calcite rings with different shapes. The optimum conditions associated with the shape of the calcite rings, which serve as thermal insulator, was obtained. This permitted to minimize the temperature gradients inside the chamber and make the diamond synthesis more uniform.

Key words: Simulation; Temperature profile; Diamond; Finite element method.

ANÁLISE POR ELEMENTOS FINITOS DA DISTRIBUIÇÃO DE TEMPERATURA NA CÂMARA DE SÍNTESE DE DIAMANTES COM DIFERENTES ANÉIS DE CALCITE

Resumo

O objetivo deste trabalho foi avaliar por simulação com elementos finitos a influência da forma dos anéis de calcite na distribuição de temperatura na câmara de alta pressão para síntese de diamantes. Foi simulado por um programa de computador baseado em elementos finitos um modelo do dispositivo de alta pressão e alta temperatura para síntese de cristais de diamante. Usando esta simulação, foi determinada a distribuição de temperatura na câmara de alta pressão com diferentes formas dos anéis de calcite. Foi obtida a condição ótima associada com a forma dos anéis de calcite, que serve como isolante térmico. Isto permitiu minimizar os gradientes de temperatura na câmara e tornar a síntese de diamantes mais uniforme.

Palavras-chave: Simulação; Perfil de temperatura; Diamante; Elementos finitos.

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1 INTRODUCTION

Diamonds are well known by their extreme hardness and other unique properties such as very high thermal conductivity and very low coefficient of expansion.[1] The hardness, in particular, confers a broad range of industrial application of diamond tools mainly for drilling, cutting and polishing rocks and metals.[2] In general, diamonds used for industrial purpose are synthetically produced mostly by high pressure and high temperature (HPHT) conditions attained inside special press equipments.[3] These diamonds can be synthesized in the form of powder, single crystal or polycrystalline particles. The synthesis process occurs in relatively short times that could last from seconds to a few minutes, depending on the type of diamond and desired size.[4]

Technically, the transformation of graphite into diamond (G→D) takes place within a well-arranged mixture of graphite powder and a metallic alloy, which serves as a catalyst solvent. As the HPHT conditions is reached, for instance 6 GPa and 1900 K, inside a high pressure chamber (HPC) the molten alloy dissolves carbon from the graphite and diamond crystal are then nucleated and grow to sizes, usually, no greater than 0.5 mm[5]

Owing to the severe HPHT conditions and the limited dimensions of the chamber, it is practically impossible to perform detail measurements of the existing temperature and pressure at the precise location where the G→D synthesis is occurring.[6] In the industry, these measurements are indirectly performed during calibration or by association with values of electric current or mechanical power at the moment of synthesis. As a consequence, the knowledge of the pressure and temperature values is very restricted and the variation profile inside the HPC extremely difficult to obtain by experimental methods.[7]

By mathematical modeling and computer simulation it is possible, however, to determine not only the values but also the local distribution of pressure and temperature in the diamond synthesis.[8] Since diamond transformation is based on thermally activated mechanisms, such as carbon diffusion, temperature is considered the most important parameter. Therefore, the first objective of this work was to develop a computational and mathematical model to simulate the temperature distribution in a HPHT diamond synthesis process using the Finite Element Method.[9-11] After validation, the model was used to evaluate the influence of the shape of calcite rings on the temperature distribution inside the high pressure chamber for the synthesis of diamond.

2 THE MODELED SYSTEM

In the present work the modeling was based on a real anvil with central concavity type high pressure device (HPD), schematically shown in Figure 1 in its pressurized state.[12] Due to the cylindrical symmetry only the left side is depicted in Figure 1. The main parts of the system are the following: 1-anvils; 2- reaction mixture (graphite powder and a metallic alloy); 3- calcite container; 4- calcite rings; and 5- graphite discs.

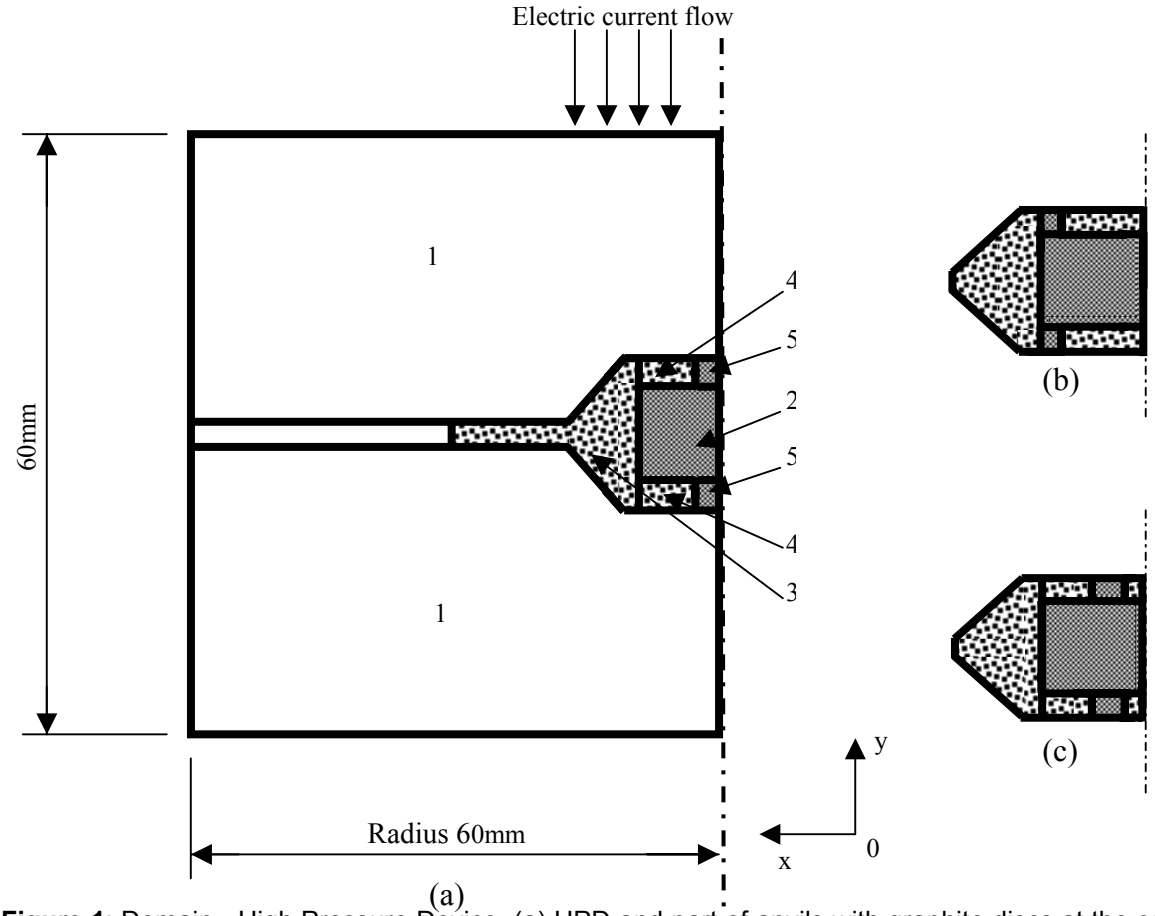


Figure 1: Domain - High Pressure Device. (a) HPD and part of anvils with graphite discs at the center of the calcite rings – as normally applied in industry; (b) graphite discs outside of the calcite rings; and (c) graphite discs at the center of the calcite rings.

Figure 2 presents the top view of the calcite container showing the three variations in the shape and position of the calcite ring analyzed in the present work.

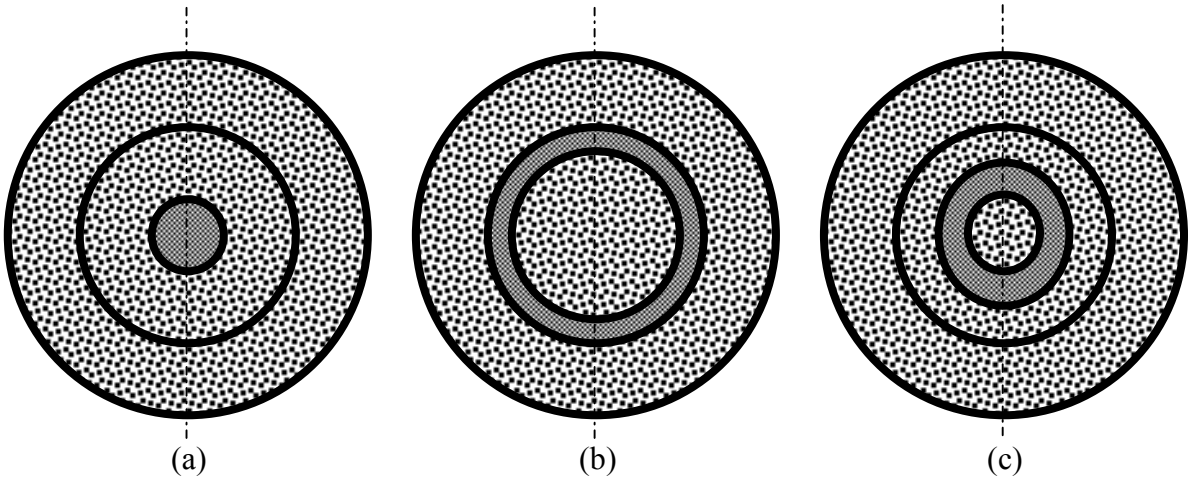


Figure 2: Calcite container with different shapes of calcite rings. (a) graphite discs at the center of the calcite rings – as normally applied in industry; (b) graphite discs outside of the calcite rings; and (c) graphite discs at the center of the calcite rings.

Soon after the HPC is pressurized, the system is submitted to an electric current to generated heat to the process. Inside the HPC the current flows mainly from the anvil to the reaction mixture, respectively parts 1 and 2 in Figure 1 (a), which are

conductors of electricity. As the system is kept pressurized and over heated, with HPHT conditions corresponding to the field of thermodynamic equilibrium for diamond existence, groups of carbon atoms will dissolve into the liquid catalyst alloy. At each one of these groups, a crystallographic rearrangement of atoms takes place and diamond nuclei are formed. Within a convenient interval of time, these diamond nuclei will grow and attain a proper size for industrial application. In addition to the processing time, other factors specially the temperature, influence the final characteristics and properties of the transformed diamonds.[13]

Another point worth mentioning is the reason for a local distribution of temperatures i.e., the establishment of a field of temperatures inside the HPC. The pair of anvils, part 1 in Figure 1 (a), is made of hard metal with both elevated hardness and thermal conductivity. This promotes strong temperature gradients located at the region where synthesis is occurring inside the HPC. That is why a pair of calcite rings, part 4 in Figure 1 (a), is placed on top (and bottom) of the reaction mixture to dissipate heat. However, the calcite rings cannot occupy the whole surface of the graphite since the electric currents has to flow through. Thus, a non-uniform thermal dissipation and related field of temperature will necessarily exist.

Still another phenomenon contributes to a non-uniform temperature distribution and consequently to a field of temperatures inside the HPC during the G→D transformation. Contrary to graphite, diamond has dielectric properties and behaves as an insulator. Therefore, as more diamond is formed inside the reaction mixture region, the flow of electrical current, for the same applied voltage, will decrease. Therefore, a decrease in temperature is also to be expected, which favors the non-uniform distribution of temperatures. In practice, the decrease in current density may be compensated by an increase in voltage but even this procedure tends to enhance the field of temperatures.

By means of the presented model, it is expected that the influence of the different arrangements and shapes of the calcite rings on the temperature distribution could be evaluated both, at the reactive mixture and at the interval part of the anvils next to the reactive mixture.

3 THE MATHEMATICAL MODELING

This was done using the heat equation:

$$c^2\nabla^2 u + q(x,y,z)u = u_t \tag{1}$$

where "u" is the temperature, "q" is a heat source and "c" are material properties.

To the steady state the Eq. 1 turns:[14]

$$\frac{\partial}{\partial x}\left(kx\frac{\partial u}{\partial x}\right) + \frac{\partial}{\partial y}\left(kx\frac{\partial u}{\partial y}\right) = -q(x,y) \tag{2}$$

where k is the thermal conductivity.

$$k = k(x,y)$$

Eq. (2) is associated with the following boundary conditions illustrated in Figure 1:

$$u = u(x, y)$$

$$u(L, y) = \ u(x,0) = \ u(x, h) = 50\,^{0}\text{C};$$

$$\left.\frac{\partial u}{\partial x}\right|_{x=0} = 0 \quad \text{(Symmetry condition);}$$

where h and L are, respectively, the height and the radius of the HPC.

4 THE FINITE ELEMENT SOLUTION

Lets consider the domain $\Omega \subset R^2$ with frontier Γ and the Sovolev space $H^1(\Omega)$. Into the space:

$$H = \{u \in H^1(\Omega); u = 30 in \Gamma\}$$

$$V = \{v \in H^1(\Omega); v = 0 in \Gamma\}$$

the weak formulation of the problem (2) is given by Hughes:[15]

To find $u \in H$ such as

$$\int_\Omega kx \nabla u . \nabla v d\Omega = \int_\Omega qv d\Omega \quad \forall v \in V \ .$$

Lets consider the domain $\Omega \subset R^2$ and lets the Sovolev space be $H^1(\Omega)$. The variational formulation of the problem in Eq. (2) is:

To find $u \in H^1(\Omega)$ such that

$$\int_\Omega kx \nabla u . \nabla v d\Omega = \int_\Omega qv d\Omega \ , \ \forall v \in H^1(\Omega). \tag{3}$$

The partition of Ω into N_e finite elements, which constitute the mesh, is given by

$$\overline{\Omega} = \bigcup_{e=1}^{Ne} \bar{\Omega}^{\,e} \text{ with } \Omega^e \cap \Omega^f = \varnothing \ , \ e \neq f$$

where Ω^e corresponds to the interior of the generic element e, and $\bar{\Omega}^{\,e}$ is the closure. The mesh parameter is given by, $h = \max\ h_e$, $e = 1,2,...N_e$, in which h_e is the diameter of the element e. Then, the space of finite elements of degree k is given by

$$S_h^k(\Omega) = \{v^h \in H^1(\Omega), v_e^h \in P_k(\Omega^e)\}$$

$$V_h^k(\Omega) = \{v^h \in V, v_e^h \in P_k(\Omega^e)\}$$

where u_e^h e v_e^h is the restriction of u^h e v^h to the element and respectively, and $P_k(\Omega^e)$ is the set of polynomial defined in Ω^e with degrees less than or equal to k.

Therefore, the problem in Eq. (3), given by its approximation by finite elements, is:

To find $u^h \in S_h^k(\Omega)$ such that:

$$\int_\Omega kx \nabla u^h . \nabla v^h d\Omega = \int_\Omega q v^h d\Omega \quad , \forall v^h \in S_h^k(\Omega). \tag{4}$$

5 NUMERICAL RESULTS

The problem in Eq. 4 was solved using 7100 triangular elements. For the solution the thermal conductivities (W/(mm.K)) were considered as: 0.1, 2.5×10^{-2} and 1.83×10^{-3}, for the reaction mixture composed of graphite and catalyst alloy, hard metal anvil and calcite, respectively.

Figures 3 and 4 exemplifies the results obtained for one heat source. In Figure 3 one can see that the calcite ring is responsible for a lower temperature gradient inside the reactive mixture. By contrast, a significant temperature gradient is formed at adjacent parts. In other words, the temperature gradient is displaced from the reactive mixture towards the calcite rings, the container and the compression anvils. As a consequence, the highest temperature gradients are located in the external region surrounding the reactive mixture.

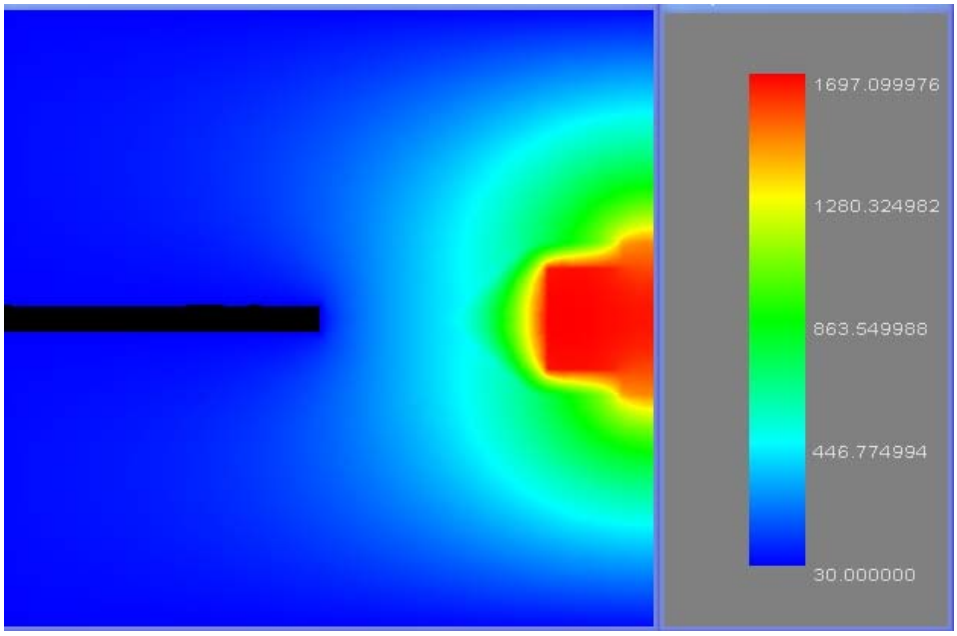
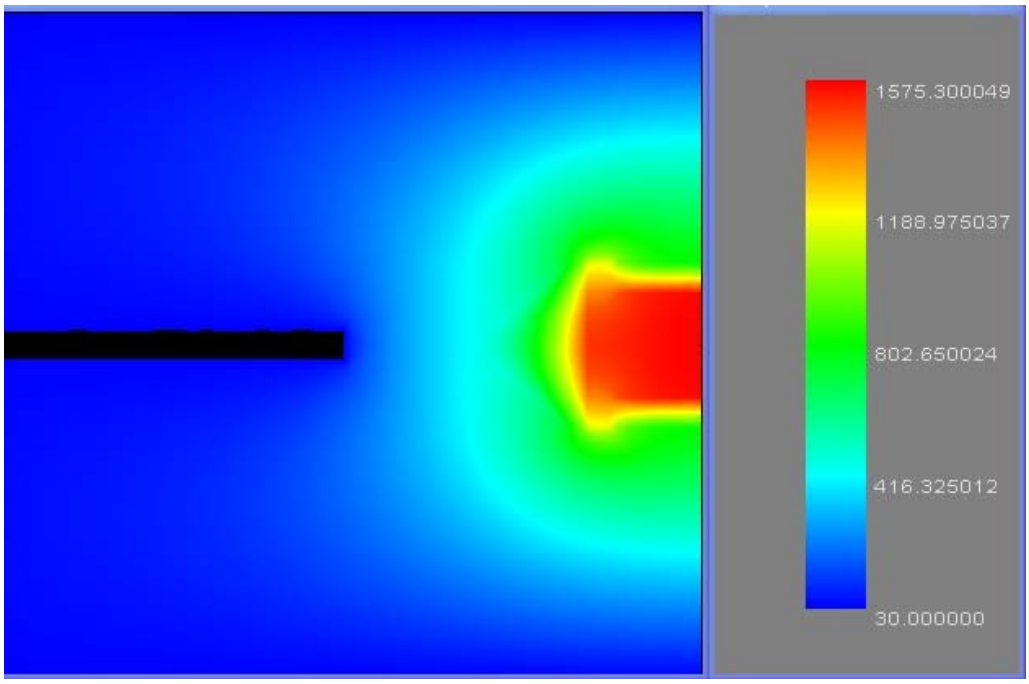


Figure 3: Temperature profile (K) with heat source of 15 W/mm^3. Graphite discs at the center of the calcite rings – as normally applied in industry.

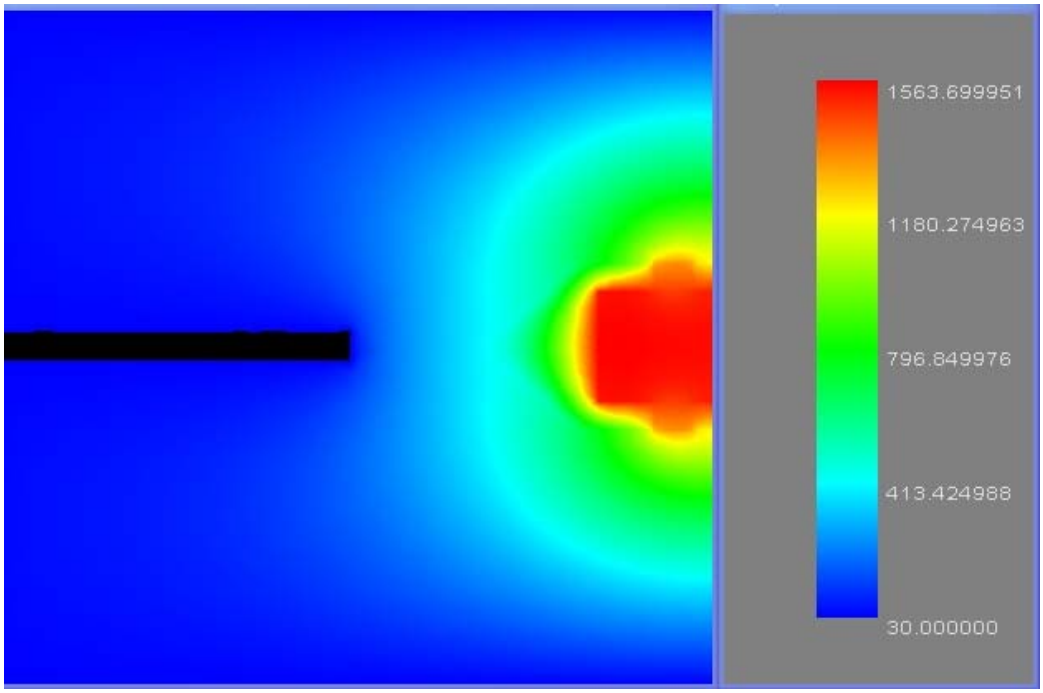
Figure 4 shows that no significative change occurred in the temperature distribution at the reactive mixture (region where the diamond synthesis takes place) following modification in the shape/location of the calcite rings. However, a reduction in the reactive mixture temperature occurred after the modification. This implies in a higher application of power as a source of heat. On the other hand, the result of the simulations, indicates that a displacement in the temperature gradient within the graphite discs, takes place towards a location far from the center of the anvils.

Temperature gradients in the anvils are very much unwanted since they reduce the expected lifetime. Moreover, the more distant the gradients are from the center of the anvils, the more difficult is to overcome them.

Figure5 shows, for comparison, the results given in the work of Novikov.[9] It should be noticed that this result is similar to the ones shown in the present work and illustrate in Figures 3 and 4.



(a)



(b)

Figure 4: Temperature profile (K) with heat source of 15 W/mm^3. (a) Graphite discs at the outside of the calcite rings; and (b) graphite discs at the center of calcite rings.

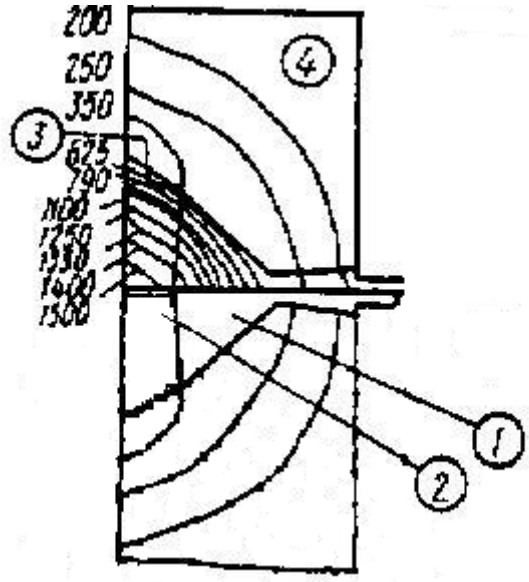


Figure 5: Temperature profile (200K - 1500K) given in the work of Novikov.[9] 1- calcite container; 2- reaction mixture; 3- isotherms in reaction mixture; 4-anvils.

6 CONCLUSIONS

The results obtained for the finite element computational simulation of the temperature distribution inside the high pressure chamber are in agreement with data known in the literature regarding this subject.

Based on the model developed, the temperature distribution can be simulated aiming at a better process performance by changing the design of the chamber. The primary objective is to keep the reactive mixture under the most possible homogeneous distribution of temperature. This would result in lower temperature gradients and, consequently, associated with more perfect diamonds with better quality and properties. No less important is the fact that greater temperature gradients acting on the hard metal anvils are to be avoided since they reduce the lifetime and raise the cost of the equipment since the anvil is one of the most expensive part and has a great influence on the final price of synthesized diamonds.

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