

Diamond crystallization in a CO₂-rich alkaline carbonate melt with a nitrogen additive



Alexander F. Khokhryakov^{a,b,*}, Yuri N. Palyanov^{a,b}, Igor N. Kupriyanov^a, Denis V. Nechaev^a

^a Sobolev Institute of Geology and Mineralogy, Siberian Branch of the Russian Academy of Sciences, Koptyug Pr., 3, Novosibirsk 630090, Russian Federation

^b Novosibirsk State University, 630090, Russia

ARTICLE INFO

Article history:

Received 11 April 2016

Received in revised form

31 May 2016

Accepted 2 June 2016

Communicated by: Peter Rudolph

Available online 7 June 2016

Keywords:

A1. Crystal morphology

A1. Point defects

A2. Single crystal growth

A2. High pressure and high temperature

B1. Diamond

ABSTRACT

Diamond crystallization was experimentally studied in a CO₂-bearing alkaline carbonate melt with an increased content of nitrogen at pressure of 6.3 GPa and temperature of 1500 °C. The growth rate, morphology, internal structure of overgrown layers, and defect-impurity composition of newly formed diamond were investigated. The type of growth patterns on faces, internal structure, and nitrogen content were found to be controlled by both the crystallographic orientation of the growth surfaces and the structure of the original faces of diamond seed crystals. An overgrown layer has a uniform structure on the {100} plane faces of synthetic diamond and a fibrillar (fibrous) structure on the faceted surfaces of a natural diamond cube. The {111} faces have a polycentric vicinal relief with numerous twin intergrowths and micro twin lamellae. The stable form of diamond growth under experimental conditions is a curved-face hexoctahedron with small cube faces. The nitrogen impurity concentration in overgrown layers varies depending on the growth direction and surface type, from 100 to 1100 ppm.

© 2016 Elsevier B.V. All rights reserved.

1. Introduction

Research of diamond crystallization processes in transition metal-free systems of various compositions has been actively conducted over the last two decades. These studies have mainly focused on simulating growth medium and determining P–T parameters of the natural diamond formation. Among other potential media of the natural diamond formation, ultra alkaline carbonate-fluid systems where growth occurs at the minimal P and T parameters are of special interest [1]. In addition, a number of recent works have proven the important role of both alkaline carbonate melts in the generation of proto-kimberlite melts [2–5] and carbon dioxide as the most important mantle fluid component in oxidized domains [6]. The possibility of diamond crystallization in the presence of carbon dioxide is also supported by detection of inclusions containing carbonate [7–9] and CO₂ fluid [10] in natural diamonds. Recently, we investigated the effect of P–T parameters on diamond crystallization in a CO₂-rich sodium carbonate melt [11]. The morphology, internal structure, and defect-impurity composition of diamond were studied. We found several interesting features, including the fibrillar structure of growth sectors of {100} faces and the presence of a 566 nm center in

photoluminescence spectra of produced diamonds, which is also observed in natural type Ib diamonds. However, the nitrogen impurity content in diamonds grown in the Na₂CO₃+CO₂+C system did not exceed 150 ppm. At the same time, the nitrogen impurity content in many natural diamonds exceeds 1000 ppm. The elevated nitrogen content in natural diamonds may be related to both the peculiarities of diamond growth and the possibility of an increased nitrogen content in the diamond crystallization medium. An increased nitrogen content in the Earth's mantle may be supported by the findings of iron nitrides and carbonitrides [12] and a CO₂-N₂ fluid [13] as inclusions in natural diamonds. However, involvement of a nitrogen-bearing fluid in natural mineral-forming processes and diamond crystallization has not been investigated. All preceding works on doping of diamond with nitrogen have been conducted using metal-carbon systems whose composition differs significantly from that of most natural systems. In this paper, we report the results of our first experiments on diamond crystallization in a CO₂-rich sodium carbonate melt in the presence of nitrogen additive. We studied the growth rate, morphology, internal structure, and defect-and-impurity content of the newly formed diamond.

2. Experimental procedure

Experiments on diamond crystallization were performed using a multi-anvil high-pressure “split-sphere” apparatus [14]. A

* Corresponding author at: Sobolev Institute of Geology and Mineralogy, Siberian Branch of the Russian Academy of Sciences, Koptyug Pr., 3, Novosibirsk 630090, Russian Federation.

E-mail address: khokhr@igm.nsc.ru (A.F. Khokhryakov).

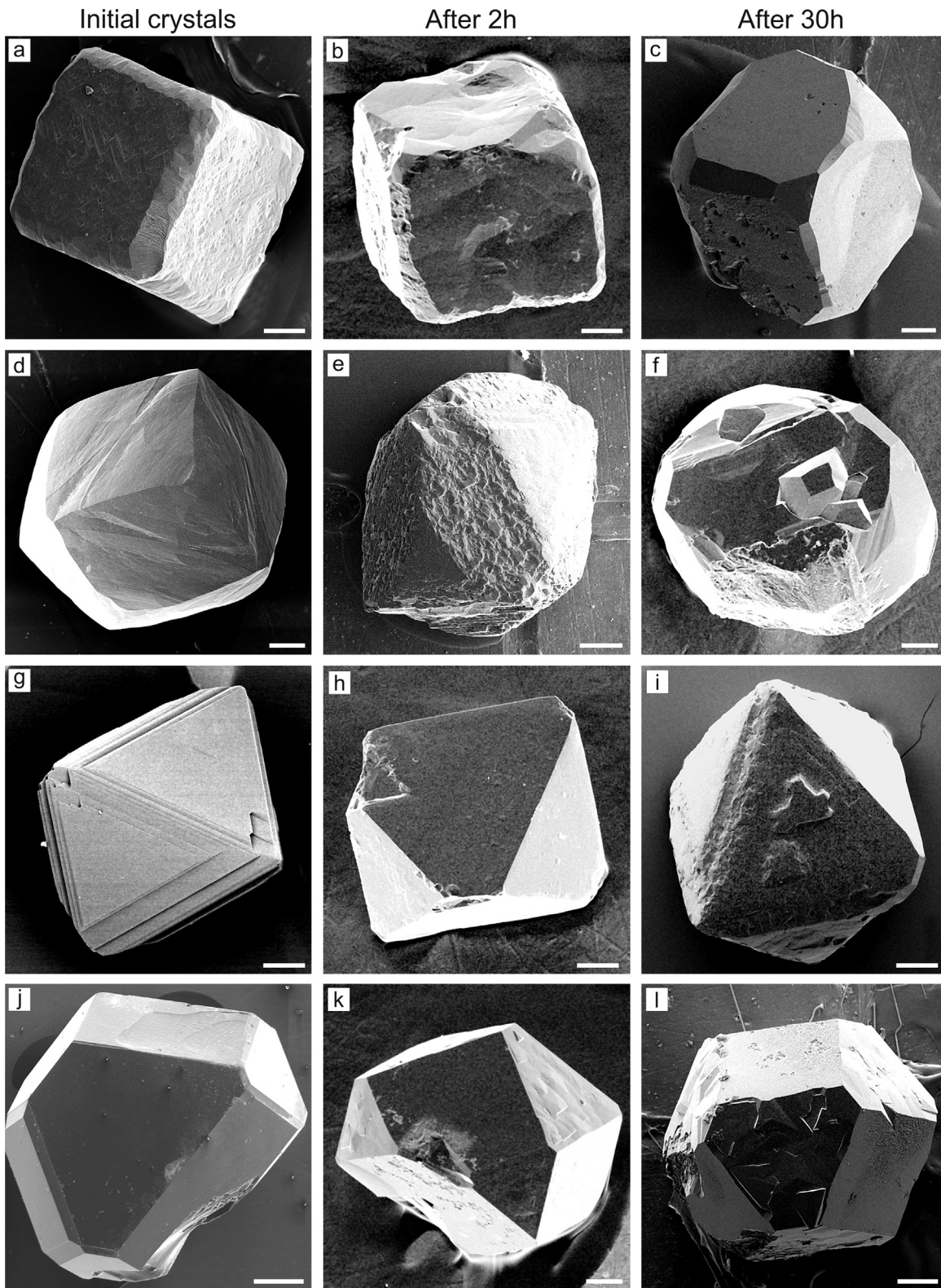


Fig. 1. SEM micrographs of the seed diamond crystals before and after experiments. Scaling bar corresponds to 100 μm .

$19 \times 19 \times 22$ mm high pressure cell in the form of a tetragonal prism was used. The design and composition of the high pressure cell are described in our previous works [15,16]. $\text{Na}_2\text{C}_2\text{O}_4$ (sodium oxalate) of 99.99% purity was used as the starting material. The use of sodium oxalate ensured diamond crystallization from a CO_2 -rich carbonate melt and did not require an additional carbon source under the applied experimental parameters [11,17]. CaCN_2

(calcium cyanamide) of 99.9% purity was added to the charge to increase nitrogen content of the growth system. Octahedral, cubic and dodecahedral natural diamonds and cube-octahedral synthetic diamonds with sizes of 0.4–0.5 mm were used as the seed crystals. The octahedral crystals had sharp edges and corners and showed triangular layers on the faces (Fig. 1g). The cubic crystals had rough cube faces with numerous square pits (Fig. 1a).

Download English Version:

<https://daneshyari.com/en/article/1789545>

Download Persian Version:

<https://daneshyari.com/article/1789545>

[Daneshyari.com](https://daneshyari.com)