### **Accepted Manuscript**

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PII: S0042-207X(18)31453-2

DOI: 10.1016/j.vacuum.2018.09.055

Reference: VAC 8268

To appear in: Vacuum

Received Date: 1 August 2018

Revised Date: 26 September 2018 Accepted Date: 27 September 2018

Please cite this article as: Li F, Zhang X, Liu H, Zhao J, Xiao Y, Feng Q, Zhang J, Rapid synthesis of inorganic  $[Ca_{24}Al_{28}O_{64}]^{4+}(e^{-})_4$  electride and its performance as an electron thermal emitter, *Vacuum* (2018), doi: https://doi.org/10.1016/j.vacuum.2018.09.055.

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#### ACCEPTED MANUSCRIPT

# Rapid synthesis of inorganic $[Ca_{24}Al_{28}O_{64}]^{4+}(e^{-})_4$ electride and its performance as an electron thermal emitter

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#### **ABSTRACT**

We propose an effective and rapid route for synthesizing polycrystalline mayenite electride (C12A7:e) in dense bulk. C12A7:e bulks with high-density electron doping were obtained via a comprehensive method, which combined Ti powder treatment with spark plasma sintering (SPS) process under a  $10^{-2}$ Pa vacuum. The resulting electride, which exhibits an electron concentration of ~  $2.3 \times 10^{21}$  cm<sup>-3</sup>, an electrical conductivity of ~  $1.38 \times 10^{3}$  S/cm at 300K, and an absorption peak at 2.5eV, was obtained via SPS process at  $1150^{\circ}$ C for 20 minutes. Furthermore, thermionic emission from a flat surface of C12A7 electride was examined at temperatures of 973-1373K and acceleration electrical field of 3000-35000V/cm in a  $10^{-5}$  vacuum. A large current density of 1.68A/cm<sup>2</sup> was obtained in the thermionic emission with an work function ( $\phi_{WF}$ ) of 2.13eV for an applied electric field of 35000V/cm. Moreover, the emission with a current density of 1.68A/cm<sup>2</sup> was stably sustained for 30 h. These results not only suggest an efficient method for fabricating high-quality mayenite electrides but also pave a way for various applications including thermionic emission.

Keywords: mayenite electride; SPS process; thermionic emission.

#### 1. Introduction

C12A7:O<sup>2-</sup> oxide was first found as a typical phase in alumina cement; chemical formula of the unit cell which contains two molecules of C12A7:O<sup>2-</sup> can be expressed as  $[Ca_{24}Al_{28}O_{64}]^{4+}+2O^{2-}$  [1-2]. The unit cell is constructed by a positively charged  $[Ca_{24}Al_{28}O_{64}]^{4+}$  lattice framework, which is composed of 12 sub-nanometer-sized cages [3]. These 12 cages are closely connected in three-dimensional space by sharing an open windows which consists of Ca-O-Al-O-Al-O 6-atom annuli [4]. To maintain electrical neutrality, two additional O<sup>2-</sup> ions randomly distribute in two of these twelve cages [5]. The encaged O<sup>2-</sup> ions which are loosely bound to Ca<sup>2+</sup> cations in the cage wall can be replaced by diverse anions such as OH, H, Cl, F, O<sup>-</sup>, O<sub>2</sub><sup>-</sup> and electrons (e<sup>-</sup>) without destroying lattice structure. [6-9]. when the free O<sup>2-</sup> ions are partly or totally substituted by e<sup>-</sup>, the resultant electride can be expressed by

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