



<p style="text-align: center;">FORM 2 THE PATENTS ACT, 1970 (39 OF 1970) & The Patents Rules, 2003 COMPLETE SPECIFICATION (Refer section 10 and rule 13)</p>
<p>TITLE OF THE INVENTION:</p> <p>COMPOSITION FOR SUSTAINED RELEASE OF CARBONATE AND A WATER PURIFICATION DEVICE BASED ON THE SAME WITH ENHANCED BIOCIDAL ACTIVITY</p>
<p>2. APPLICANT:</p> <p>(A) NAME: INDIAN INSTITUTE OF TECHNOLOGY MADRAS</p> <p>(B) NATIONALITY: Indian</p> <p>(C) ADDRESS: INDIAN INSTITUTE OF TECHNOLOGY MADRAS IIT P.O Chennai - 600 036</p>
<p>3. Preamble to the Description</p> <p style="text-align: center;">COMPLETE SPECIFICATION</p> <p>The following specification particularly describes the invention and the manner in which is to be performed.</p>

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COMPLETE SPECIFICATION

TITLE OF THE INVENTION

- 5 **COMPOSITION FOR SUSTAINED RELEASE OF CARBONATE AND A WATER PURIFICATION DEVICE BASED ON THE SAME WITH ENHANCED BIOCIDAL ACTIVITY**

FIELD OF INVENTION

- 10 The present invention articulates a scaffold composition of a material that releases, in a sustained manner for prolonged usage, an anion of choice, in specific carbonate ions (CO_3^{2-}), entrapped within the designed architecture into water for its purification. The composition that releases CO_3^{2-} in conjunction with M^{n+} acts as an enhancing system to increase the biocidal properties of transition metal ions, especially silver ions at lowest possible
- 15 concentrations, whose concentration in free state is smaller due to its reactions with interfering species usually found in real water. The present invention articulates the method of preparation of a pellet adjunct releasing CO_3^{2-} which reinforces the microbial disinfection of M^{n+} in water in a fast and complete fashion and a water purification device based on the same. The composition can be tuned to release another ion of interest from the same carrier
- 20 composite and it is demonstrated here for potassium release, without disturbing the carbonate release and the composite does not release any other unwanted ions in significant quantities from its framework.

BACKGROUND OF THE INVENTION

- 25 Transition metal ions, specifically, silver and copper have been traditionally utilized for disinfection of water. There are several mechanisms associated with the biocidal property of silver and copper and is covered in several recent articles (Pradeep T, Anshup, Thin Solid Films, 2009, 517, 24, 6441–6478; Feng QL et al, J Biomed Mater Res., 2000, 52, 662-668; Z. Xiu et al, Nano Lett., 2012, 12, 4271–4275). It has recently been learnt through several
- 30 detailed studies that dissolution of anions in water can reduce the consumption of silver up to 60% used for disinfection purposes in view of the anticipated crisis in silver production through their synergic effect (WO 2015/059562 A1).

It is therefore important to develop new ion releasing compositions containing anions, in particular, carbonate ion, which can provide enhancement in the disinfection ability of metals

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in diverse conditions of water quality. For a water purification device using such materials to be effective, the compositions should release ion/ions steadily, at the required level and this must occur over extended period at room temperature.

5 Prior Art

Water purification in various parts of world uses several disinfectants, while their by-products (known as disinfection by-products, DBPs) turn out to be lethal on extended use. Several trihalomethanes, haloacetic acids, haloacetonitriles, haloketones and other DBPs were found to cause colon, rectal and bladder cancers and adverse reproductive disorders, because of which the US EPA initiated a rule in the year 1992 to evaluate the need for additional controls for microbial pathogens and DBPs. The goal of this rule was to develop an approach that would reduce the level of exposure from disinfectants and DBPs, simultaneously controlling microbial pathogens (Sedlak D. L, & von Gunten U, Science, 2011, 331, 42-43). Biochemical properties of silver in the ionic form, against pathogens like bacteria, virus, fungi and protozoa have been widely studied and the reaction products silver may produce are less harmful compared to other disinfectants which produce DBPs.

Traditionally, noble metals, especially silver, are used as disinfectants in delivering safe portable water worldwide against severe gastroenteritis-causing pathogens. The biochemical property of metals in ionic form, against pathogens like bacteria, virus, fungi, protozoa, etc. is widely studied. Silver for water purification is one of the oldest known technologies and its use dates back to 500 BC. A recent review covers the importance of silver in water purification (Pradeep T, Anshup, Thin Solid Films, 2009, 517, 24, 6441–6478).

Amongst several transition metal ions that are usually found in nature, silver ion is the only example which has its chloride salt highly insoluble in water (solubility = 1.9 mg/L at 25 °C). This solubility limit is seemingly designed by nature for a specific reason: to reduce the silver concentration in water which limits the mobility of silver in living bodies.

Active-silver, the concentration of effective ionic silver available for interaction with microbes in test water conditions varies with water quality. Available free silver continues to decrease with increasing concentration of Cl^- in water. For a specific silver ion concentration, the antimicrobial performance is in the order, seawater \leq high organic matter-containing

water \leq high divalent cation-containing water \leq synthetic water due to the speciation of Ag^+ (Zhang H, Smith J A, Craver V O, Water Res. 2012, 46, 691-699).

There are several mechanisms associated with the biocidal property of silver and copper and are covered in several recent articles (Pradeep T, Anshup, Thin Solid Films, 2009, 517, 24, 6441-6478; Feng QL et al, J Biomed Mater Res., 2000, 52, 662-668; Z. Xiu et al, Nano Lett., 2012, 12, 4271-4275). Antimicrobial activity of various transition metals is well-reported in the literature. Silver and copper have been of special interest, largely because they have no known long-term health effects in humans at the concentrations of use as well as their large disinfection potential. However, other transition metals are not so effective disinfectants, especially with regards to enteric microorganisms (Muller H E, Zentralbl Bakteriologie Mikrobiologie Hyg B., 1985, 182, 95-101). Anti-bacterial effect of transition metals is usually named as oligodynamic effect, as they are most effective at low concentrations (because of solubility limits imposed by various anions, they can't exist as ions at higher concentrations in real water). It is suggested that toxicity of metal ions for fungi goes in the following order: $\text{Ag} > \text{Hg} > \text{Cu} > \text{Cd} > \text{Cr} > \text{Ni} > \text{Pb} > \text{Co} > \text{Au} > \text{Zn} > \text{Fe} > \text{Mn} > \text{Mo} > \text{Sn}$ (Martin, H. 1969. In D. C. Torge-son (ed.), Fungicides, vol. 11. Academic Press Inc., New York).

While the precise mechanism of silver ion attack on bacteria is not known, however, based on its known strong binding with sulphur, it is suggested that silver binds with sulphur containing enzymes and proteins (Bragg PD, Rainnie DJ, Can J Microbiol., 1974, 20, 883-9).

Interaction of silver with other components in the bacterial cell membrane through release of K^+ ion or through hydrogen bonding is also suggested (Schreurs WJ, Rosenberg H, J Bacteriol., 1982, 152, 7-13). It is difficult to ascertain the precise mechanism of silver's antimicrobial activity because most of such studies are conducted at higher silver ion concentrations at which it may undergo precipitation through interaction with cellular sulphur containing compounds. Conducting studies at low concentrations of silver is quite important for understanding the mechanism but it requires significant experimental care.

Use of water soluble monovalent metal carbonates (for example, Na_2CO_3) in water purification is generally for alkalization and water softening (e.g., European patent application EP0812808B1). Prior art reports of using metal carbonates (such as partially soluble magnesium or calcium carbonate) along with transition metals having antibacterial activity for drinking water is limited to them being slow dissolving tablets as an indicator for volume of water passed (e.g., WO 2006/070953, WO 2013/046213). It has recently been

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learnt through our earlier work that dissolution of anions in water can reduce the consumption of silver up to 60% through their synergic effect (Swathy J.R., et al Sci reports, 2014, 4:7161). In view of the anticipated crisis in silver production, this finding is important in the context of sustainable use of silver for water purification.

- 5 Our previous invention, WO2015/059562 A1 describes that carbonate in water can enhance biocidal activity of silver (Ag^+) (where Ag^+ is released at 50 ppb concentration). However, materials for obtaining sustainable and effective concentration of carbonate in water are required for this invention to be effective.

Thus the present work is built upon our previous applications which relates to the design of a material which releases anion, in particular carbonate (CO_3^{2-}), specifically in a sustained fashion for improving biocidal activity of metal ions in water for prolonged period of time. The material serves as a reinforcement acting along with our previous invention WO2012/140520 A2 which releases sustained silver ions in water. It is also learnt from these prior art that the presence of carbonate in water can reduce the interfering peripheral membrane proteins on the cell surface thus making the cell naked for the action of silver ion. Thus a robust thousand fold enhanced antimicrobial activity of silver is obtained at a 60% lesser concentration, in presence of carbonate. It is important to note that such a composition should be permitted for use in water, especially drinking water.

Therefore, the main object of the present invention is to develop an effective, simple and cost-effective composition based on anions or cations of interest and more particularly carbonate and potassium ions for obtaining a resilient antimicrobial activity at lowest possible silver ion concentration.

Another object of the present invention is to develop a water purification device based on the prepared composition. An object of such a device is to ensure constant release of ions from the composition in water, over prolonged use. A representative diagram of the water purification device is also described.

SUMMARY OF THE INVENTION

It is understood from prior art that a sustained anion releasing material in continuous flow of water is significant for complete biocidal action of several biocides. Therefore, the present invention demonstrates a composition containing CO_3^{2-} homogenized in the

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aggregated network of amended material, composed of a residual amorphous phase. The confined CO_3^{2-} ions get released in a sustained order, offering strong antimicrobial activity even in presence of interfering species, usually found in water.

One aspect of the invention shows the material in conjunction with the M^{n+} releasing system achieves a complete biocidal activity with higher efficiency of the metal ion while reducing the overall required M^{n+} by 60%. M^{n+} and CO_3^{2-} are present in widely separated concentration window (e.g., typical concentration of M^{n+} in $\text{M}^{n+}/\text{CO}_3^{2-}$ is in the range of 1-10 μM whereas typical concentration of CO_3^{2-} is in the range of 100-1000 μM).

Another aspect of the present invention describes the method of adding the composition to water in such a way that a constant and stable release of CO_3^{2-} is obtained. The composition is thereby demonstrated for use as an accompaniment in a water purification device.

Another embodiment of the present invention demonstrates that the release from this material can enhance the overall killing efficiency of $\text{M}^{n+}/\text{CO}_3^{2-}$ significantly by 1000 fold compared to the killing efficiency obtained with transition metal ions alone (more particularly silver and copper ions).

In another embodiment the material is proved to be inert and just the carbonate is released, whereas, other unwanted scaffold compositions are not released and thus making it safe for water purification.

In other embodiment, the present invention shows that it can be modified in several ways to tune the release of alternative ions of interest, mainly potassium, into water. The present invention also demonstrates that the killing efficiency using the composition is affected in the case of depleted ingredient in the composition, which may be utilized as an indication to replenish the composition in a water purification device.

BRIEF DESCRIPTION OF THE DRAWINGS

The accompanying figures, which are incorporated in and constitute a part of this specification, illustrate several aspects and together with the description serve to explain the principles of the invention.

FIG. 1: A schematic representation of the method involved in the preparation of the amalgamated composite loaded with carbonate, in accordance with an embodiment of the present invention.

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FIG. 2: Scanning electron microscopic (SEM) image of the granular particles of the composite loaded with CO_3^{2-} confirmed by Raman spectrum in the inset and a corresponding energy dispersive X-ray spectrum (EDS) based elemental composition in accordance with various aspects of the present invention.

5 **FIG. 3:** (A) X-ray photoelectron spectra (XPS) of the CO_3^{2-} releasing composite loaded with CO_3^{2-} before and after heating. (B) and (C) show the deconvoluted XPS spectra of carbon and oxygen, respectively. (D) Powder XRD pattern of the material before and after heating in accordance with various aspects of the present invention.

FIG. 4: Batch trials of sustained carbonate release from the granular particles of the composite loaded with CO_3^{2-} for several trials in accordance with various aspects of the present invention.

FIG. 5: Performance of a water purification device containing the composite loaded with CO_3^{2-} solely releasing sustained CO_3^{2-} during the passage of 250 L of water in accordance with various aspects of the present invention.

15 **FIG. 6:** Comparison of bacterial killing efficiency of silver ions in the presence and absence of CO_3^{2-} wherein the proposed composite is a source of CO_3^{2-} releasing material as claimed in accordance with an embodiment of the present invention.

FIG. 7: Comparison of bacterial killing efficiency of Zn^{2+} and Cu^{2+} ions in the presence and absence of CO_3^{2-} wherein the proposed granular composite is a source of CO_3^{2-} releasing material as claimed in accordance with an embodiment of the present invention.

FIG. 8: Rate of viral killing efficiency by M^{n+} and composition ($\text{M}^{n+}/\text{CO}_3^{2-}$), wherein M^{n+} represent d-block cations and the proposed granular composite as a source of CO_3^{2-} , on Bacteriophage MS2 in accordance with an embodiment of the present invention.

25 **FIG. 9:** Scanning electron microscopic (SEM) image of the amended granular particles of the composite loaded with CO_3^{2-} and K^+ , presence of CO_3^{2-} confirmed by Raman spectrum in the inset and a corresponding EDS spectrum based elemental composition in accordance with various aspects of the present invention.

FIG. 10: (A) X-ray photoelectron spectra XPS survey spectra of the amended granular particles of the composite loaded with CO_3^{2-} and K^+ , before and after heating. (B) and (C) shows the deconvoluted XPS spectra of carbon and oxygen, respectively. (D) Powder XRD

pattern of the material before and after heating in accordance with various aspects of the present invention.

FIG. 11: Batch trials of sustained carbonate release from the amended granular particles of the pellet composite loaded with CO_3^{2-} and K^+ for prolonged period of time in accordance with various aspects of the present invention.

FIG. 12: Batch trials of sustained potassium release from the amended granular pellet composite loaded with CO_3^{2-} and K^+ , for prolonged period of time in accordance with various aspects of the present invention.

FIG. 13: Controlled release of selective cations from the amended granular pellet composite loaded with Ca^{2+} and K^+ , where release of K^+ is enhanced while release of Ca^{2+} is controlled for prolonged period of time in accordance with various aspects of the present invention.

FIG. 14: Performance of a water purification device containing amended granular composite loaded with CO_3^{2-} and K^+ , solely releasing sustained CO_3^{2-} during the passage of 250 L in accordance with various aspects of the present invention.

FIG. 15: Rate of bacteria killing efficiency by M^{n+} and amended composition loaded with CO_3^{2-} and K^+ ($\text{M}^{n+}/\text{CO}_3^{2-}$), wherein M^{n+} represent d-block cations and the proposed granular composite as a source of CO_3^{2-} on *E. coli* in accordance with an embodiment of the present invention.

FIG. 16: Rate of antiviral efficiency of M^{n+} and composition loaded with CO_3^{2-} and K^+ ($\text{M}^{n+}/\text{CO}_3^{2-}$), wherein M^{n+} represent d-block cations and the proposed granular composite as a source of CO_3^{2-} , on Bacteriophage MS2 in accordance with an embodiment of the present invention.

FIG. 17: Schematic representation of a water purification device based on the proposed granular composite according to an illustrative embodiment of the invention.

DETAILED DESCRIPTION OF THE INVENTION

The present invention relates to the material releasing sustained CO_3^{2-} due to its augmentation ability in presence of various interfering species usually found in water. Utility of the composition is demonstrated based on a number of aspects such as ability to release CO_3^{2-} for an extended period, immobilization of other carrier composition and the release of specific ion of interest, antimicrobial activity even at low concentrations of the M^{n+} and activity against diverse types of microorganisms. These properties of the composition are

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demonstrated through use of *E.coli* and MS2 bacteriophage as model organisms for gram negative bacteria and virus, respectively.

Experimental methods

The present invention can be understood more readily by reference to the following detailed description of the invention and the examples included therein.

Before the present compounds, compositions, articles, systems, devices, and/or methods are disclosed and described, it is to be understood that they are not limited to specific synthetic methods unless otherwise specified. It is also to be understood that the terminology used herein is for the purpose of describing particular aspects only and is not intended to be limiting. Although any methods and materials similar or equivalent to those described herein can be used in the practice or testing of the present invention, example methods and materials are now described. All publications mentioned herein are incorporated by reference to disclose and describe the methods and/or materials in connection with which the publications are cited.

Disclosed are the components to be used to prepare the compositions of the invention to be used within the methods disclosed herein. The specification and the appended claims represent the composite material pellet having a source of carbonate ion in the material as chosen from the following: sodium carbonate, barium carbonate, strontium carbonate, ammonium carbonate, calcium carbonate, potassium carbonate and zinc carbonate or a combination thereof. The other components of the composition include SiO_2 , Al_2O_3 , Na_2CO_3 , K_2CO_3 (1:0.06:0.2:0.06 wt%) unless otherwise specified as a modified composition.

Example 1

FIG. 1 illustrates the scheme utilized in accordance with an embodiment of the present invention for the preparation of a scaffold composite as a carrier composition entrapping an unmodified form of CO_3^{2-} . The steps involved are sequentially described where the intricate scaffold composite is an aggregated network of amended material which is synthesized by melting silica, aluminum and anion source are homogenized and compactly packed in template molds and heated at melting temperatures in the range of 600 - 1200°C wherein the anion is from any source of CO_3^{2-} which does not decompose at this melting temperatures. The composition used for making the composite is presented in Experimental Methods.

Example 2

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FIG. 2 shows the Scanning electron microscopic (SEM) image of the granular particles of the composite and its chemical composition. Carbonate amalgamated pellet was granulated (40 – 100 μm) and FIG. 2(A) confirms the porous and crystalline morphology of the composite indicated by arrows, wherein the patterns around the particles are the carbon substrate used for mounting the material. FIG. 2(B) illustrates the granular form of the composition wherein the inset in FIG. 2(B) illustrates the Raman feature specific for carbonate. Elemental composition confirms the presence of essential elements: carbon, oxygen, silica, potassium and sodium shown in FIG. 2(C). Insets show the elemental mapping for an illustrative CO_3^{2-} impregnated composite and the scaffold compositions.

Example 3

FIG. 3 shows X-ray photoelectron spectra (XPS) of the initial components and the designed CO_3^{2-} releasing composite material. FIG. 3(A) shows the survey spectra and FIG. 3(B) depicts the deconvoluted C1s XPS spectra showing the presence of C-C peak at 284.6 eV, carbonate peak at 289.3 eV and the potassium K2p_{3/2} peak at 293.0 eV and K2p_{1/2} peak at 295.8 eV. This clearly indicates that carbonate and a small portion of potassium are entangled in the SiO_2 matrix. It is further confirmed from the deconvoluted spectra of oxygen, as shown in FIG. 3(C) where O1s peaks appear in the range between 531 – 532 eV for metal carbonates and 532 – 533 eV for SiO_2 . FIG. 3(D) shows the X-ray diffraction pattern of the amended granular composite before and after heating in accordance with the various aspects of the present disclosure. The peak corresponding to the template composition before heating has melted and resulted in a single compound depicted in the XRD pattern of the material after heating. The X-ray diffraction peaks of the claimed material are observed at 2θ (degree) values of 22.00, 28.47, 31.49, 36.13, 36.41, 47.10 and 48.65 which correspond to the (101), (111), (102), (200), (112), (113) and (212) planes of the cristobalite phase of SiO_2 (JCPDS 39-1425).

Example 4

FIG. 4 demonstrates the sustained release of carbonate from the amended pellet composite releasing carbonate, when immersed in water. In an aspect, 10 mg of the carbonate amalgamated pellet granulated as 40 - 100 μm particles was shaken briefly in 10ml of milliQ water. The carbonate release in water was inferred after different standing time of the material in water. Carbonate concentration present in the test water was calculated based on the inorganic carbon concentration measured at each step using Shimadzu TOC-V_{CPH} model. TOC analyzer is a versatile instrument to quantify the total carbon (TC) and total inorganic

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carbon (TIC) separately and difference between TC and IC gives the TOC value. The instrument was calibrated earlier using anhydrous sodium carbonate and anhydrous sodium hydrogen carbonate.

Example 5

FIG. 5 describes the release of carbonate from the designed material. In an aspect, 1L of tap water with 1g of the carbonate releasing material was shaken briefly. Typically, 15 minutes of standing time, unless otherwise mentioned, was provided for the required sustainable release of the composition. After 15 minutes of standing time, the TDS and the pH change in the water were measured as indications of release of carbonate into the water. The performance of the 1g/l of amended scaffold material releasing sustained CO_3^{2-} was evident up to ~50 L passage of water. After every 10 cycles of the above procedure, the material was dried for 18 - 20 hours and was tested for further CO_3^{2-} release. After every step of drying the sample, an increased release of CO_3^{2-} in the first 15 minutes exposure was observed (shown as a spike in the graph at trial numbers 1, 11, 21 and 31).

Example 6

FIG. 6 describes the method for measuring the enhancement in bacterial killing efficiency of silver ion in presence of the pellet, in comparison to discrete silver ion. In an aspect, 100 mL of tap water (typically containing *E. coli* concentration of 1×10^5 CFU/mL, unless otherwise mentioned) with Ag^+ (40 ppb) and Ag^+ (40 ppb)/ CO_3^{2-} (15-20 ppm released from 100 mg pellet) was separately shaken. Typically, one hour of standing time, unless otherwise mentioned, is provided for the exposure of microorganisms to the biocidal composition. After one hour of standing, 1 mL from the samples was plated along with agar on a sterile petri dish using the pour plate method. After 48 h of incubation at 37 °C, the colonies were counted and recorded.

Example 7

FIG. 7 describes the method of measuring the enhancement in bacterial killing efficiency of the composition in comparison to corresponding transition metal ion alone. In an aspect, 100 mL of tap water (typically containing *E. coli* concentration of 1×10^5 CFU/mL, unless otherwise mentioned) was separately shaken with M^{n+} , $\text{M}^{n+}/\text{CO}_3^{2-}$, where M^{n+} was externally spiked {concentration used: copper (500 ppb), zinc (1 ppm), carbonate (~20 ppm)} and CO_3^{2-} was released from the composite. 1 mL of the sample was plated along with nutrient agar on a sterile petri dish using the pour plate method after a contact time of 0, 30 and 60 minutes.

After 48 hours of incubation of plating at 37 °C, the colonies were counted and recorded.

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Example 8

FIG. 8 describes the method of measuring the enhancement in viral killing efficiency of transition metal ion with the composition in comparison to corresponding transition metal ion alone. In an aspect, 100 mL of tap water (typically containing MS2 bacteriophage concentration of 1×10^3 PFU/mL, unless otherwise mentioned) was separately shaken with M^{n+} , M^{n+}/CO_3^{2-} released from material {concentration used: copper (500 ppb), zinc (1 ppm), silver (30 ppb) carbonate (~20 ppm)}. After one hour of standing, 1 mL from the samples were plated along with soft agar and host cells on a sterile petri dish using plaque assay method. After 16 h of incubation at 37 °C, the plaques were counted and recorded.

Example 9

FIG. 9 shows the scanning electron microscopic (SEM) image of the pellet composite loaded with CO_3^{2-} and K^+ , and its chemical composition. Carbonate amalgamated pellet was granulated (40 – 100 μm). FIG. 9(A) confirms the porous and crystalline morphology of the composite indicated by arrows, wherein the patterns around the particles are the carbon substrate used for mounting the material. FIG. 9(B) illustrates the granular form of the composition wherein the inset in FIG. 9(B) illustrates the Raman feature specific for carbonate. Elemental composition confirms the presence of essential elements: carbon, oxygen, silicon, potassium, calcium and sodium shown in FIG. 9(C). Insets show the elemental mapping for an illustrative CO_3^{2-} and K^+ impregnated composite and the scaffold compositions.

Example 10

FIG. 10 shows X-ray photoelectron survey (XPS) spectra of the initial components and the amended CO_3^{2-} and K^+ releasing composite material. FIG. 10(A) shows the survey spectra and FIG. 10(B) depicts the deconvoluted C1s XPS spectra showing the presence of C-C at 284.9 eV; carbonate peaks at of 289 eV. The presence of potassium is shown by $K2p_{3/2}$ peak at 293.0 eV and $K2p_{1/2}$ peak at 295.8 eV. This clearly indicates that carbonate and a greater portion of potassium are entangled in the SiO_2 matrix. It is further confirmed from the deconvoluted spectra of oxygen, as shown in FIG. 10(C) where O1s peaks appear in the range between 531 – 533 eV for metal carbonates and SiO_2 , respectively. FIG. 10(D) shows the X-ray diffraction pattern of the amended granular composite material before and after heating in accordance with the various aspects of the present disclosure. The peak corresponding to the template composition on heating, has melted and resulted in a compound depicted in the XRD pattern of the material.

Example 11

FIG. 11 demonstrates the sustained release of carbonate from the amended scaffold composite modified to release potassium and carbonate, when immersed in water. In an aspect, 10mg of the carbonate amalgamated pellet granulated as 40 – 100 μm particles was shaken briefly in 10ml of milliQ water. The carbonate release in water was inferred after different standing time of the material in water. Carbonate concentration present in the test water was calculated based on the inorganic carbon concentration measured at each step using Shimadzu TOC-V_{CPH} model. TOC analyzer is a versatile instrument to quantify the total carbon (TC) and total inorganic carbon (TIC) separately and difference between TC and IC gives the TOC value. The instrument was in prior calibrated using anhydrous sodium carbonate and anhydrous sodium hydrogen carbonate.

Example 12

FIG. 12 demonstrates the sustained release of potassium from the amended scaffold composition modified to release in a sustained order of potassium and carbonate, when immersed in water. In an aspect, 10ml of milliQ water with 10mg of the material was shaken briefly. Source of potassium ion in the material is chosen one amongst the following: potassium carbonate, potassium nitrate, potassium chloride or a combination thereof. The potassium release in water was inferred after different standing time of the material in water. Potassium concentration present in the test water was measured at each step using inductively coupled plasma spectroscopy (ICP-MS) (Perkin Elmer Nex ION 300 ICP-MS).

Example 13

FIG. 13 demonstrates the selective release of potassium and sodium while release of other cations was controlled from the modified material, when immersed in water. In an aspect, 10ml of milliQ water with 10mg of the material was shaken briefly. The example also demonstrates the control over the release of other components from the composition. The cation release in water was inferred after different standing time of the material in water. The cation concentration released in the test water was measured at each step using inductively coupled plasma spectroscopy (ICP-MS) (Perkin Elmer Nex ION 300 ICP-MS).

Example 14

FIG. 14 describes the release of carbonate from the material modified to release potassium and carbonate in a sustained manner. In an aspect, 1L of tap water with 1g of carbonate

releasing pellet was shaken briefly. Typically, 15 minutes of standing time, unless otherwise mentioned, is provided for the required sustainable release of the composition. After 15 minutes of standing time, the TDS and the pH change in the water was measured as an indication of release of carbonate into the water. The performance of 1g/l of amended scaffold composite, releasing sustained CO_3^{2-} was evident up to ~50 L passage of water. After every 10 cycles of the above procedure, the material was dried for 18 - 20 hours and was tested for further CO_3^{2-} release. After every step of drying the sample, an increased release of CO_3^{2-} in the first 15 minutes exposure was observed (shown as a spike in the graph at trial numbers 1, 11, 21 and 31).

10 Example 15

FIG. 15 describes that the enhancement in bacterial killing efficiency of the composition is not affected while the material is modified to release potassium ions along with carbonate. In an aspect, 100 mL of tap water (typically containing *E. coli* concentration of 1×10^5 CFU/mL, unless otherwise mentioned) was separately shaken with M^{n+} , $\text{M}^{n+}/\text{CO}_3^{2-}$ released from material {concentration used: copper (500 ppb), zinc (1 ppm), silver (40 ppb) carbonate (~20 ppm)}. 1 mL of the sample was plated along with nutrient agar on a sterile petri dish using the pour plate method after a contact time of 0, 30 and 60 minutes. After 48 hours of incubation of plating at 37 °C, the colonies were counted and recorded.

Example 16

FIG. 16 describes that the enhancement in virus killing efficiency of the composition is not affected while the material is modified to release potassium ions along with carbonate. In an aspect, 100 mL of tap water (typically containing MS2 bacteriophage concentration of 1×10^3 PFU/mL, unless otherwise mentioned) was separately shaken with M^{n+} , $\text{M}^{n+}/\text{CO}_3^{2-}$ released from material {concentration used: copper (500 ppb), zinc (1 ppm), silver (30 ppb), carbonate (~20 ppm)}. After 0, 30 and 60 minutes of standing time, 1 mL from the samples was plated along with soft agar and host cells on a sterile petri dish using plaque assay method. After 16 h of incubation at 37 °C, the plaques were counted and recorded.

Example 17

FIG. 17 illustrates the schematic of the water purification device in accordance with an embodiment of the present invention. The anion releasing compound is selected from the group consisting of Sodium carbonate, Barium carbonate, Strontium carbonate, Calcium carbonate, Potassium carbonate and Zinc carbonate carefully entrapped within the carrier composition without modification and the said conduit is fitted along with a silver ion

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releasing system from our prior art WO2012/140520 A2, leading to complete and enhanced killing of microorganisms especially *Escherichia coli* and Bacteriophage MS2, the surrogates for water borne pathogens (bacteria and virus, respectively) and the removal of the microbial debris by a filter connected to an output delivers pure drinking water.

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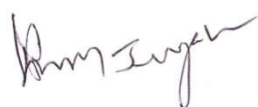
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We Claim:**CLEAN COPY**

1. A composite material with porous architecture comprises an anion source, homogenized with an intricate scaffold composite forming material prepared by heating silica and alumina, forming a rock-like solid at a temperature range of 600 - 1200°C and under ambient atmosphere, wherein the said material releases constant concentration of anions at room temperature in a sustained fashion for a prolonged period and enhances the biocidal activity of metal ions (M^{n+}), especially silver ions.
2. The material as claimed in claim 1, wherein the said anion is CO_3^{2-} selected from the group consisting of sodium carbonate, barium carbonate, strontium carbonate, ammonium carbonate, calcium carbonate, potassium carbonate and zinc carbonate.
3. The material as claimed in claim 1, wherein the said anion CO_3^{2-} releasing material fitted along with a M^{n+} releasing system leads to increase in the biocidal property, including water borne pathogens.
4. The material as claimed in claim 1, wherein the anion CO_3^{2-} releasing material increases the disinfection ability of M^{n+} up to 1000 times against gram negative bacteria and provides antiviral property to M^{n+} .
5. The material as claimed in claim 1, wherein the composite does not release any other ions from its framework other than the desired CO_3^{2-} , which is released in the range of 5 - 100 ppm in the temperature window of 10°C to 80°C in natural water and maintains a pH from about 6.5 to about 8.0.
6. The material as claimed in claim 1, wherein the release of CO_3^{2-} in water varied the electrical conductivity in the range of 100 to 1000 μS and pH in the range of 6.5 to 8.0 in cartridge mode upon soaking in continuous flow of water in a sustained mode wherein the release occurs within 5 -15 min of contact time and concentration of carbonate released is in the range of 5 to 100 ppm; the highest release occurring upon prolonged contact time.
7. The material as claimed in claim 1, wherein the concentration of CO_3^{2-} in the aqueous system of continuous water flow is from 5 ppm to 100 ppm based on the flow of the water, thereby reducing the required concentration of M^{n+} for a specific microbial load in water.
8. The material as claimed in claim 1, wherein the material when modified in its composition additionally releases different cations including potassium without affecting the release kinetics of CO_3^{2-} .

9. The material as claimed in claim 1, wherein the anion releasing material in conjunction with transition metal ion (M^{n+}) for obtaining biocidal activity in water is chosen from Fe^{3+} , Zn^{2+} , Cu^{2+} , Ag^{+} or a combination thereof at a concentration below 1 ppm.
10. A water purification device comprising a composite material as claimed in claim 1, wherein the device includes:
- a tank having an inlet and an outlet for passage of water through and
 - a filtration unit present inside the tank, the filtration unit comprising:
 - i. at least one M^{n+} releasing filtration medium for releasing metal ion in water, in specific Ag^{+} along with
 - ii. the composite material of claim 1 for releasing CO_3^{2-} , entrapped in an aggregated network of amended inert material
 - iii. wherein the filtration medium, material and the debris are disposed within a filtration unit using a debris removal disc.

Dated at Chennai this Dec 14, 2018

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COMPOSITION FOR SUSTAINED RELEASE OF CARBONATE AND A WATER PURIFICATION DEVICE BASED ON THE SAME WITH ENHANCED BIOCIDAL ACTIVITY

5

Abstract

The present invention relates to a composite material with porous architecture that releases anions in a sustained manner for prolonged usage, as an enhancing system to increase the biocidal properties of transition metal ions in water especially silver ions up to thousand fold.

10 The material comprises an anion source, homogenized in an aggregated network of amended scaffold material. The material when modified in its composition release other ions of interest from the same carrier composite and it is demonstrated here for potassium release, without disturbing the carbonate release and the composite does not release any other ions in significant quantities from its framework. The composite is further demonstrated for use as an
15 adjunct in water purification device for the fast and complete inactivation of microorganisms and as a marker for the replacement of a filter component in the water purification device.

COMPOSITION FOR SUSTAINED RELEASE OF CARBONATE AND A WATER
PURIFICATION DEVICE BASED ON THE SAME WITH ENHANCED BIOCIDAL
ACTIVITY

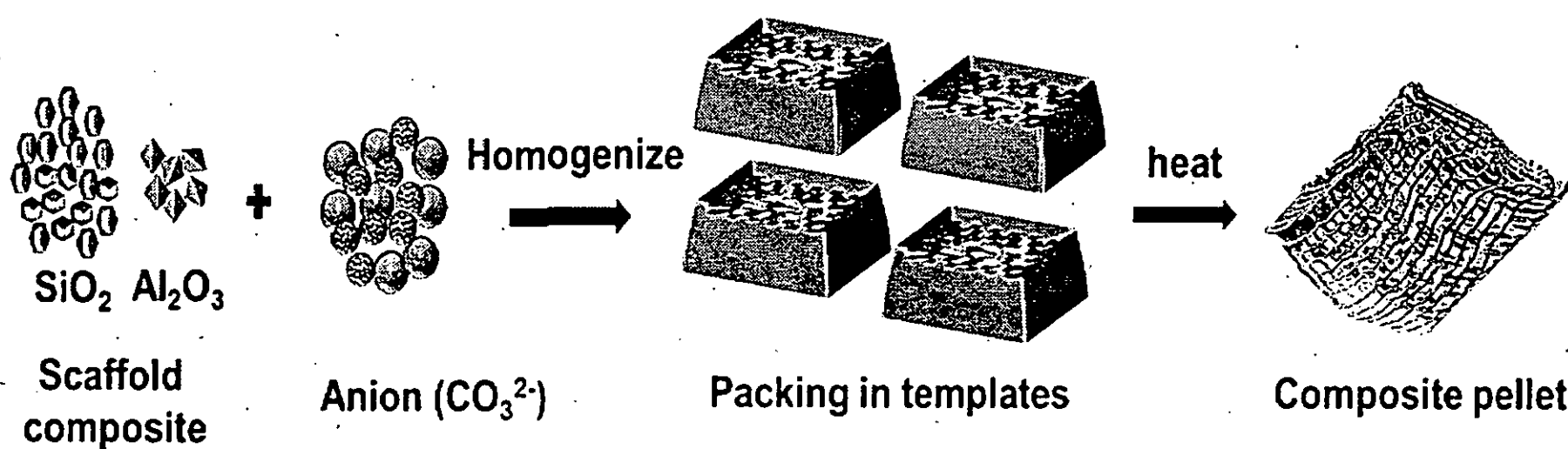


FIGURE 1

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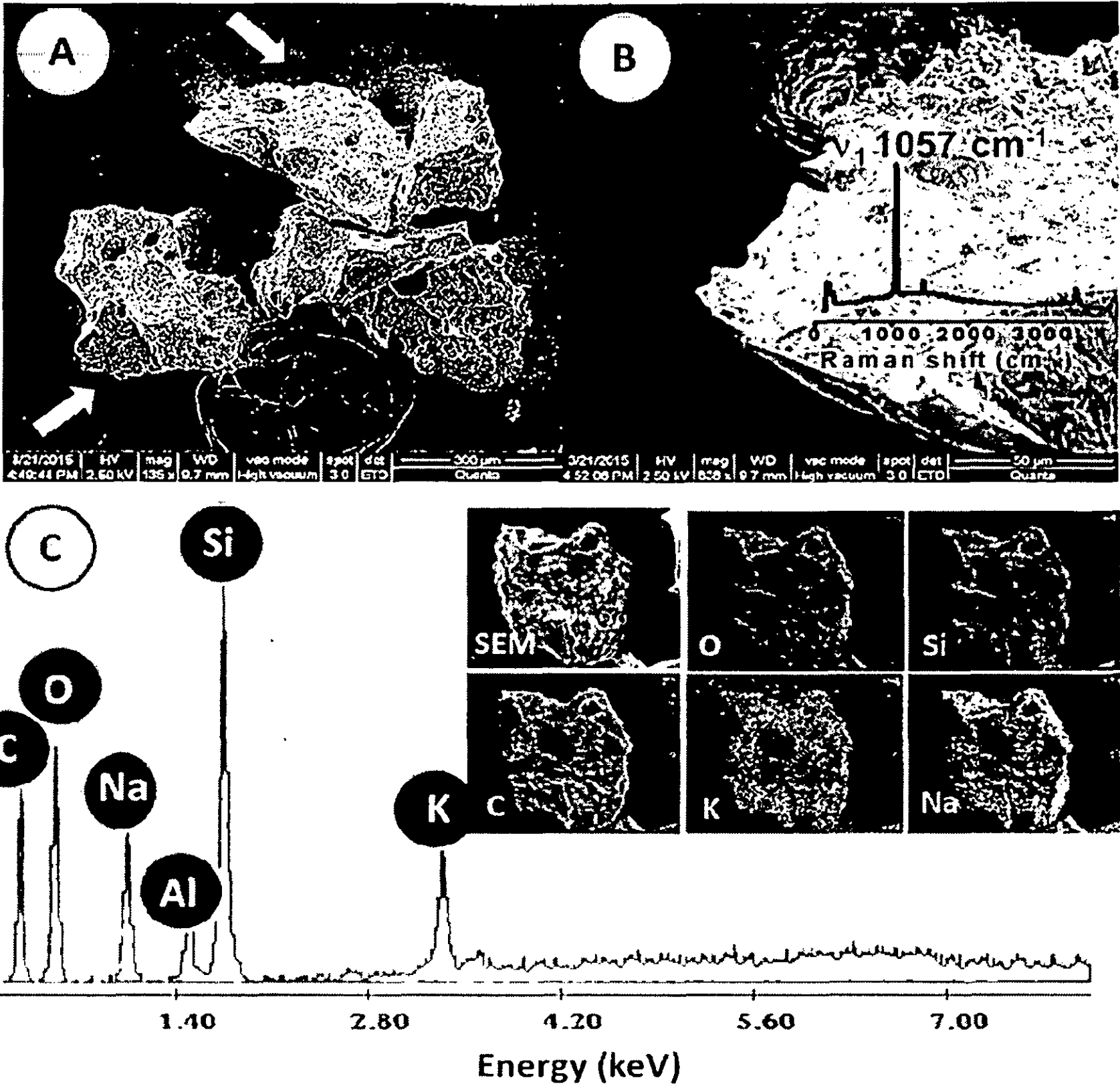
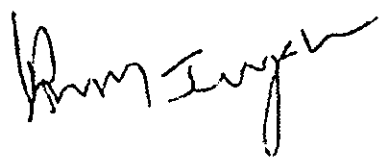


FIGURE 2

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COMPOSITION FOR SUSTAINED RELEASE OF CARBONATE AND A WATER PURIFICATION DEVICE BASED ON THE SAME WITH ENHANCED BIOCIDAL ACTIVITY

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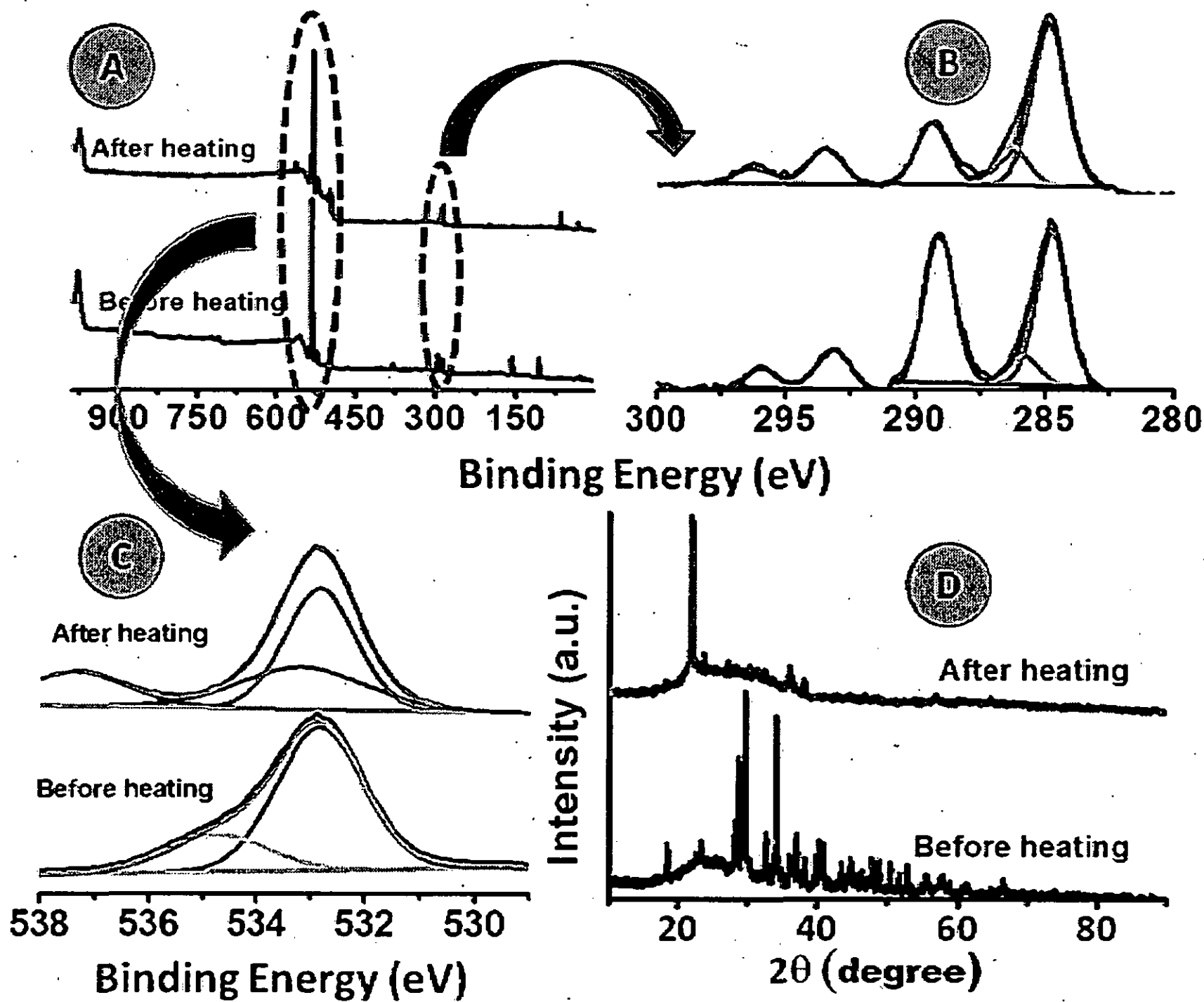


FIGURE 3

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[Handwritten Signature]

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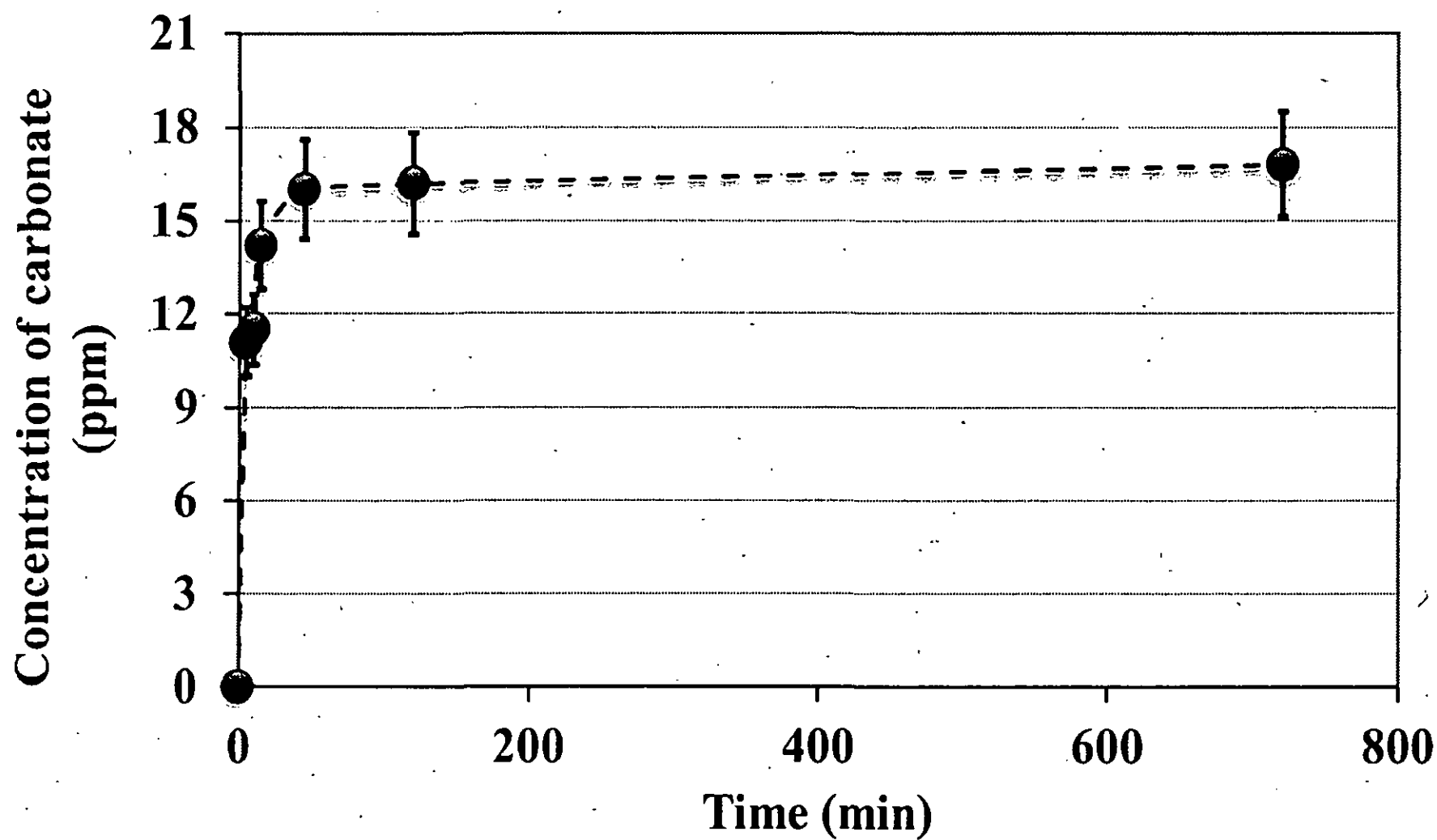


FIGURE 4

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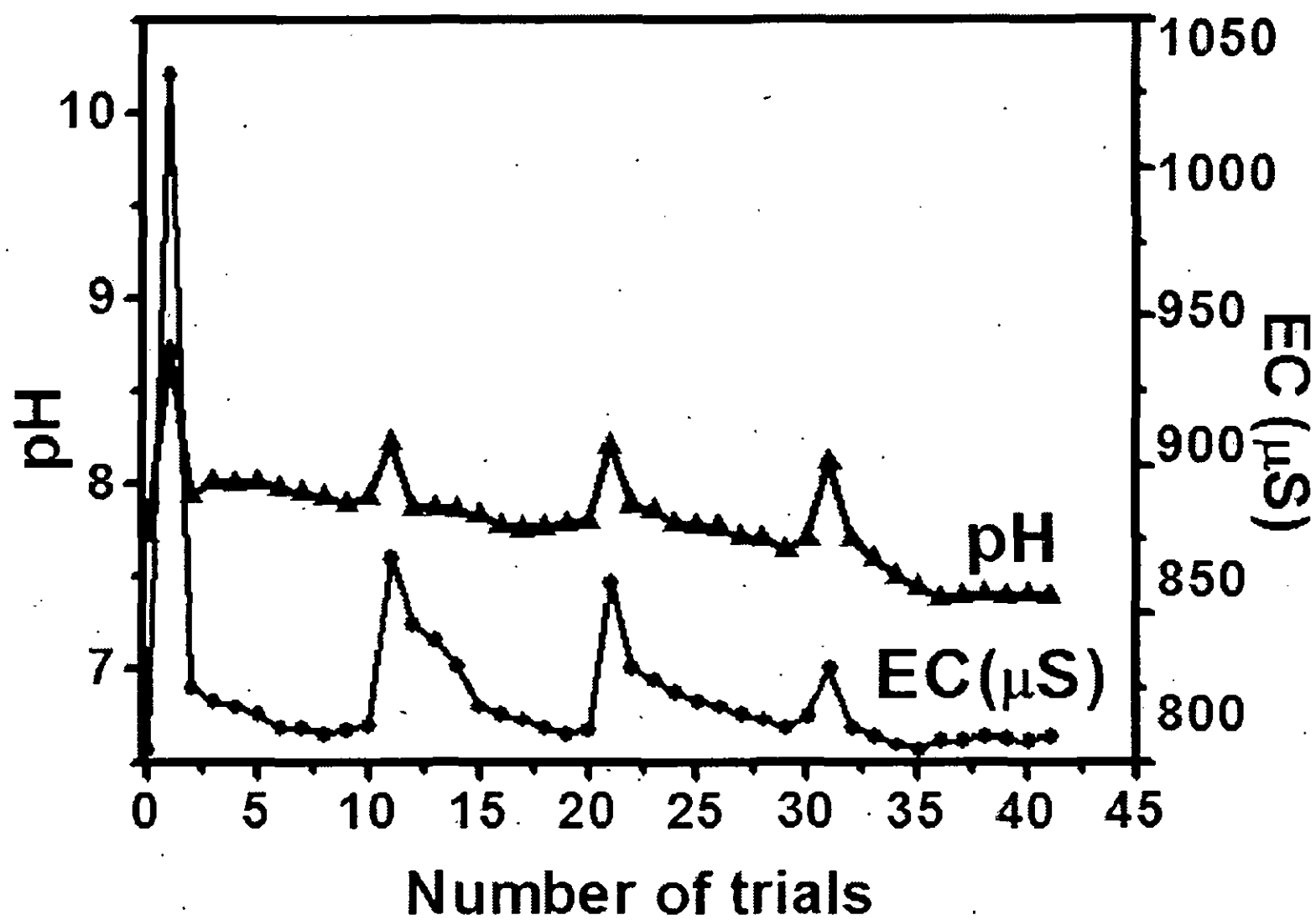


FIGURE 5

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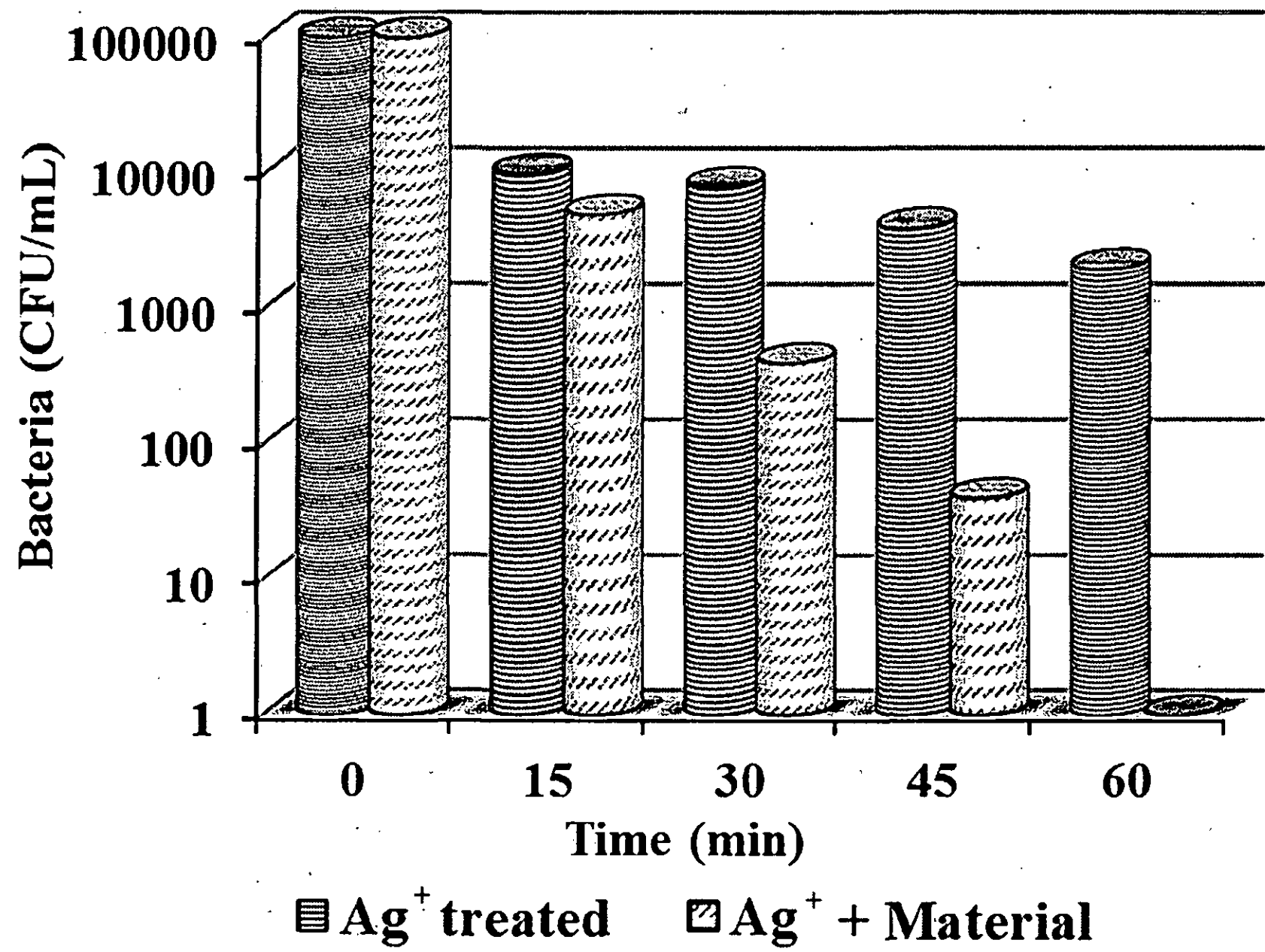
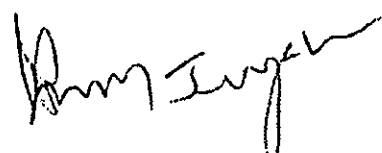


FIGURE 6

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COMPOSITION FOR SUSTAINED RELEASE OF CARBONATE AND A WATER
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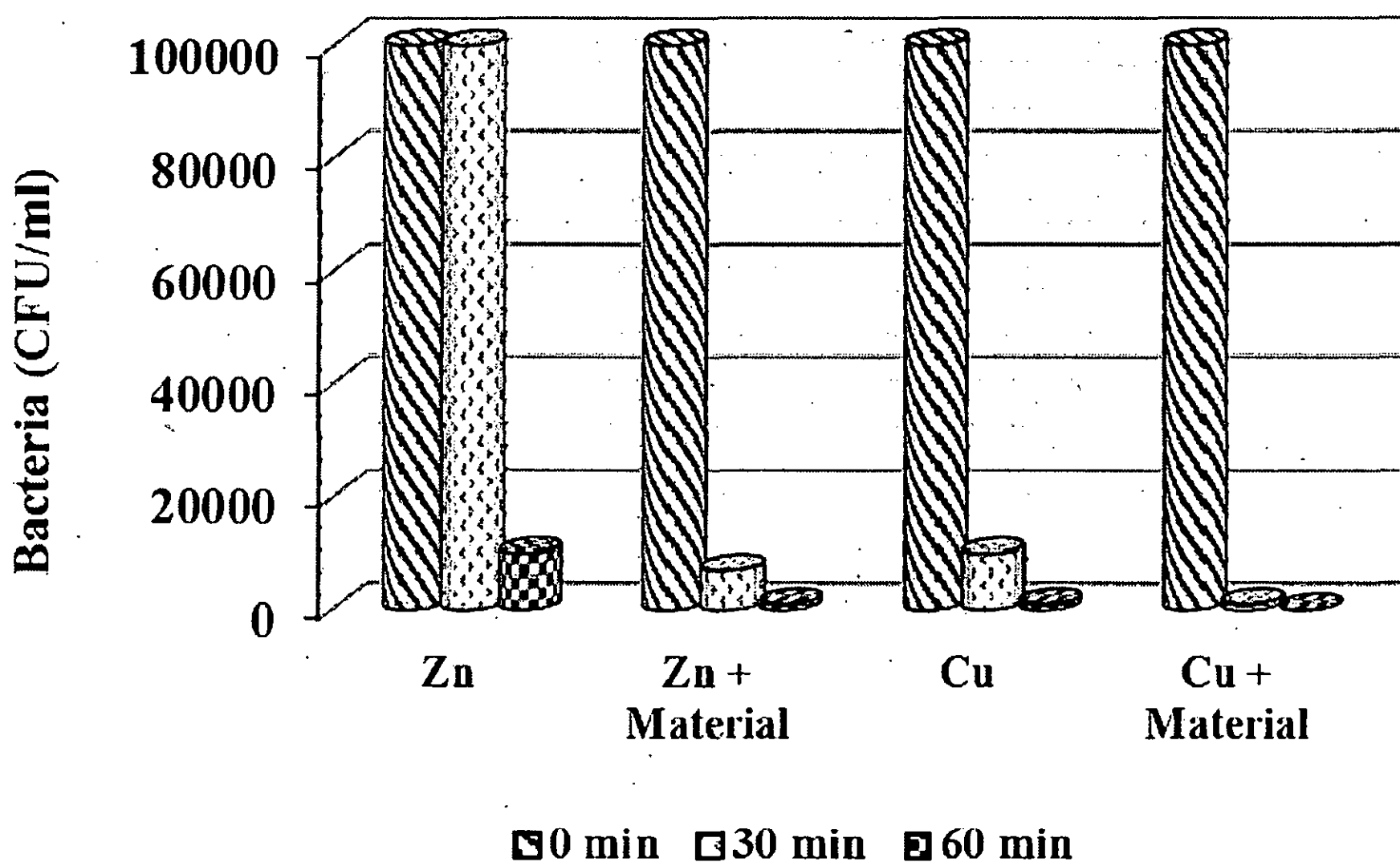


FIGURE 7

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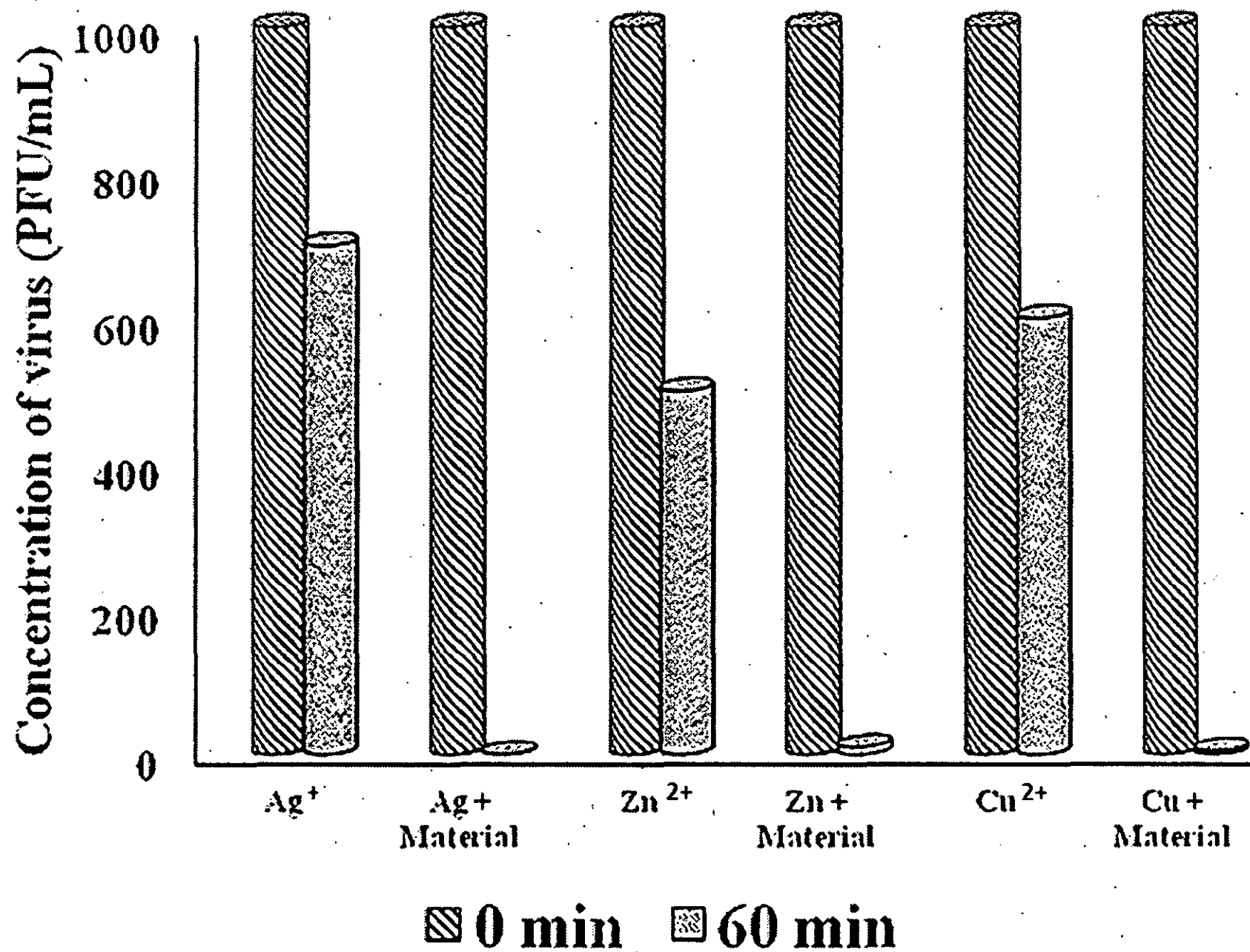


FIGURE 8

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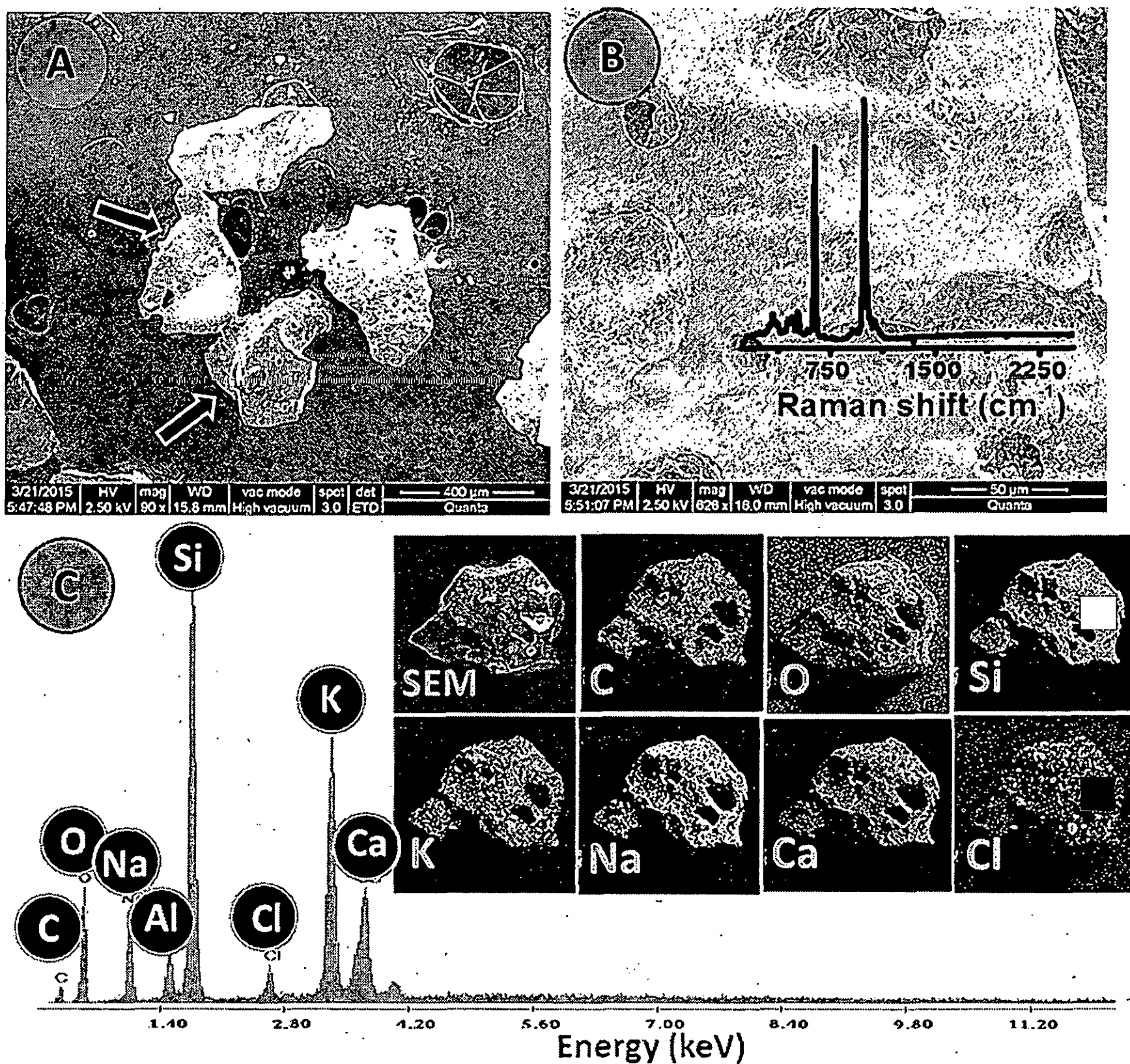


FIGURE 9

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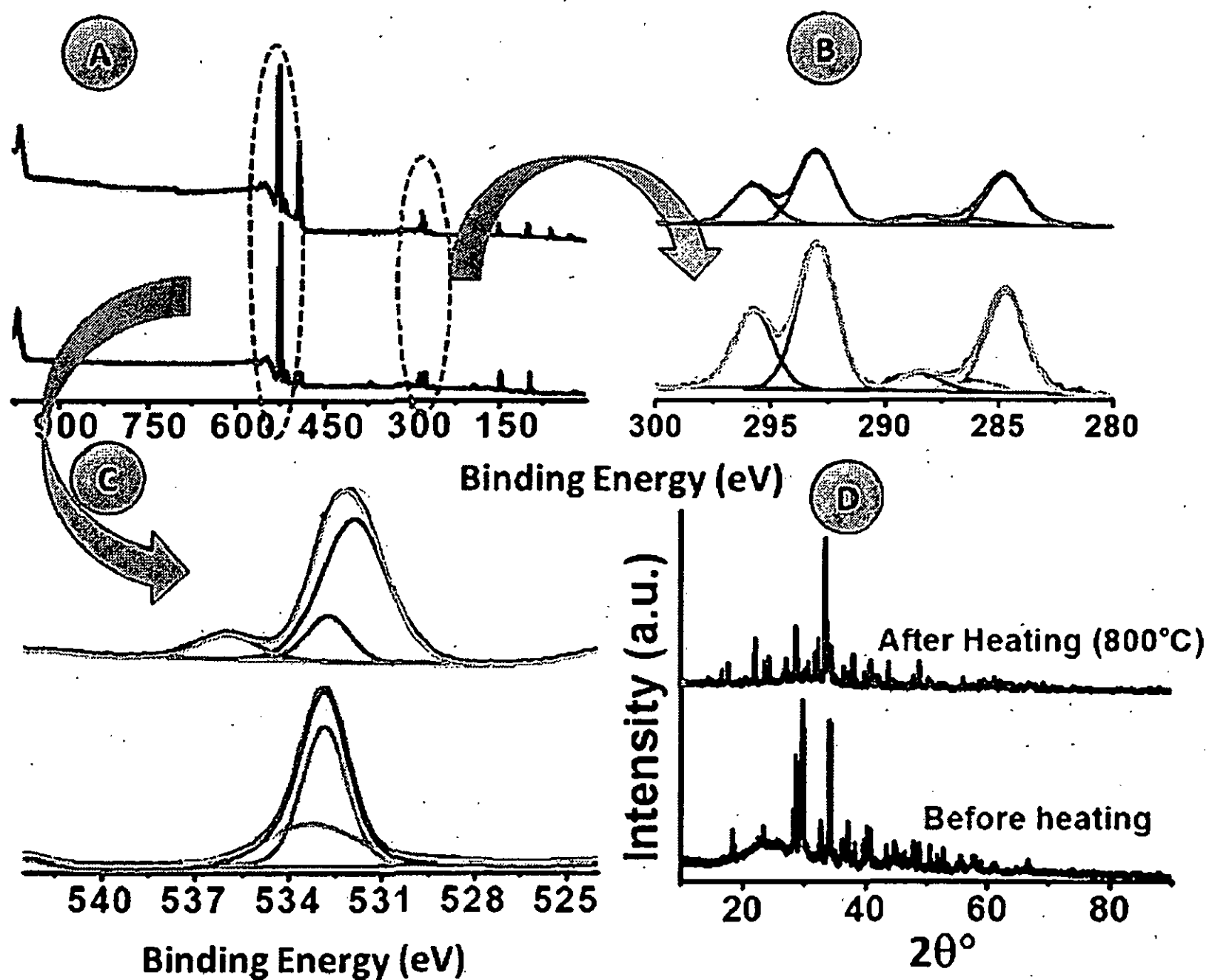


FIGURE 10

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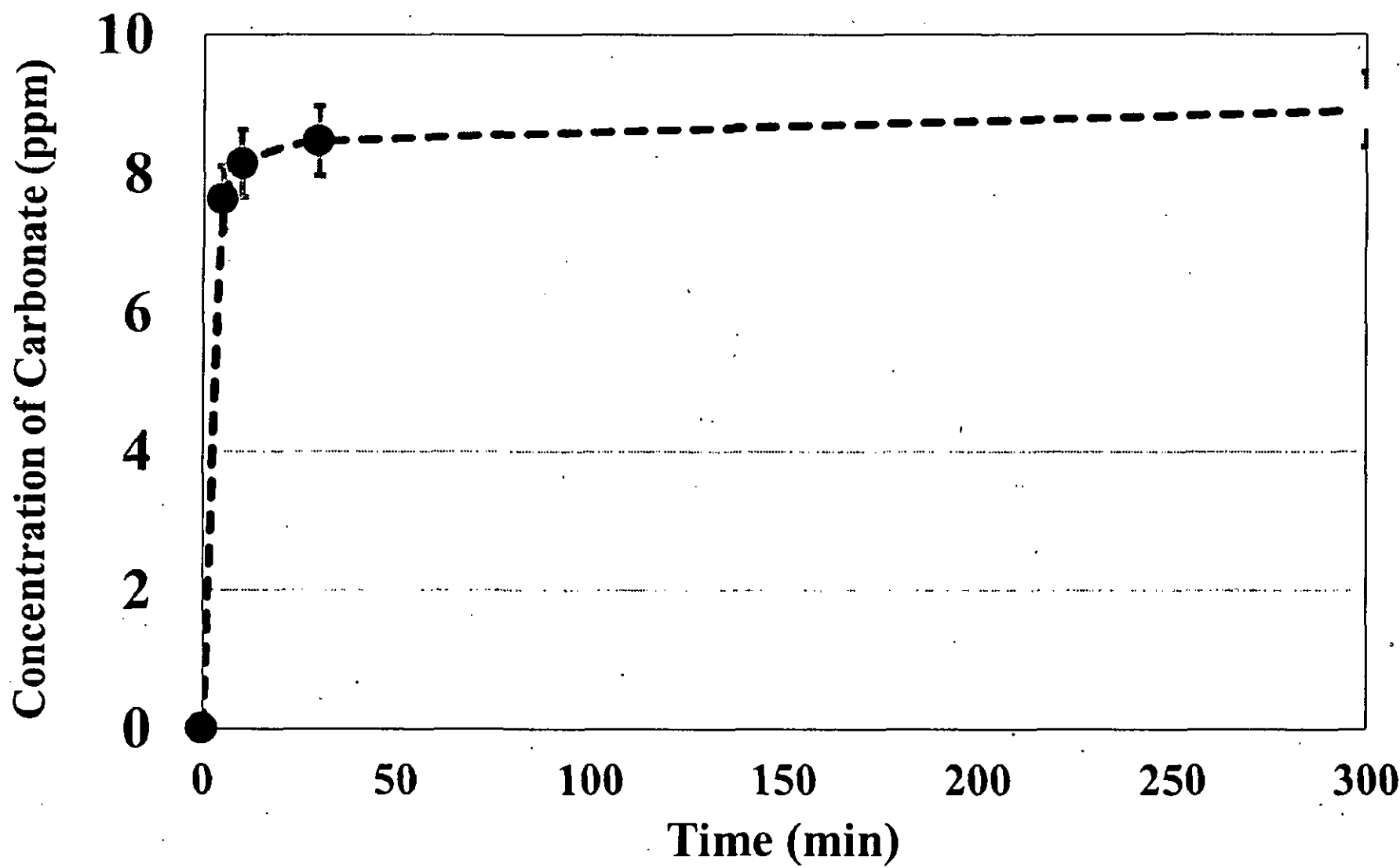


FIGURE 11

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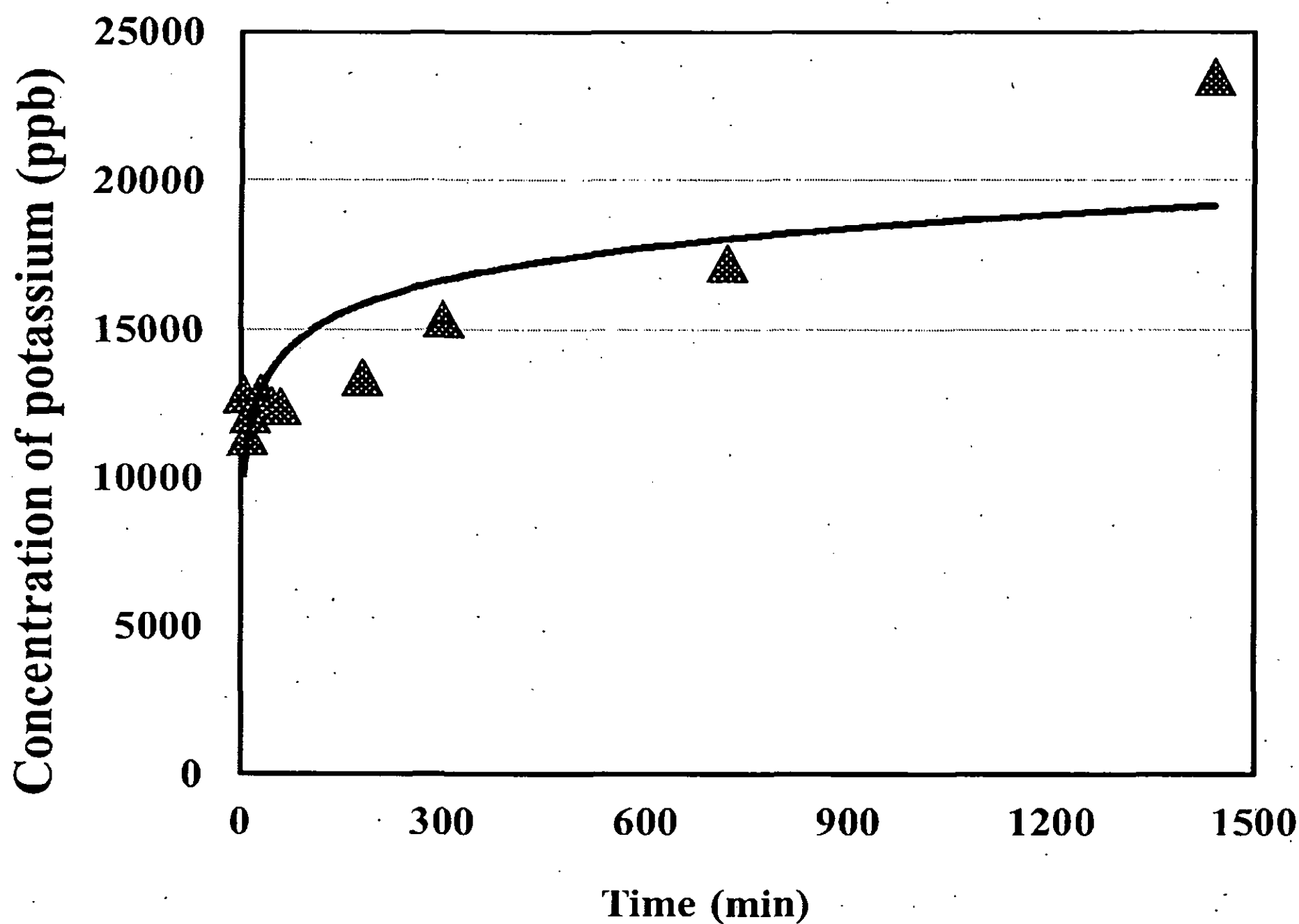


FIGURE 12

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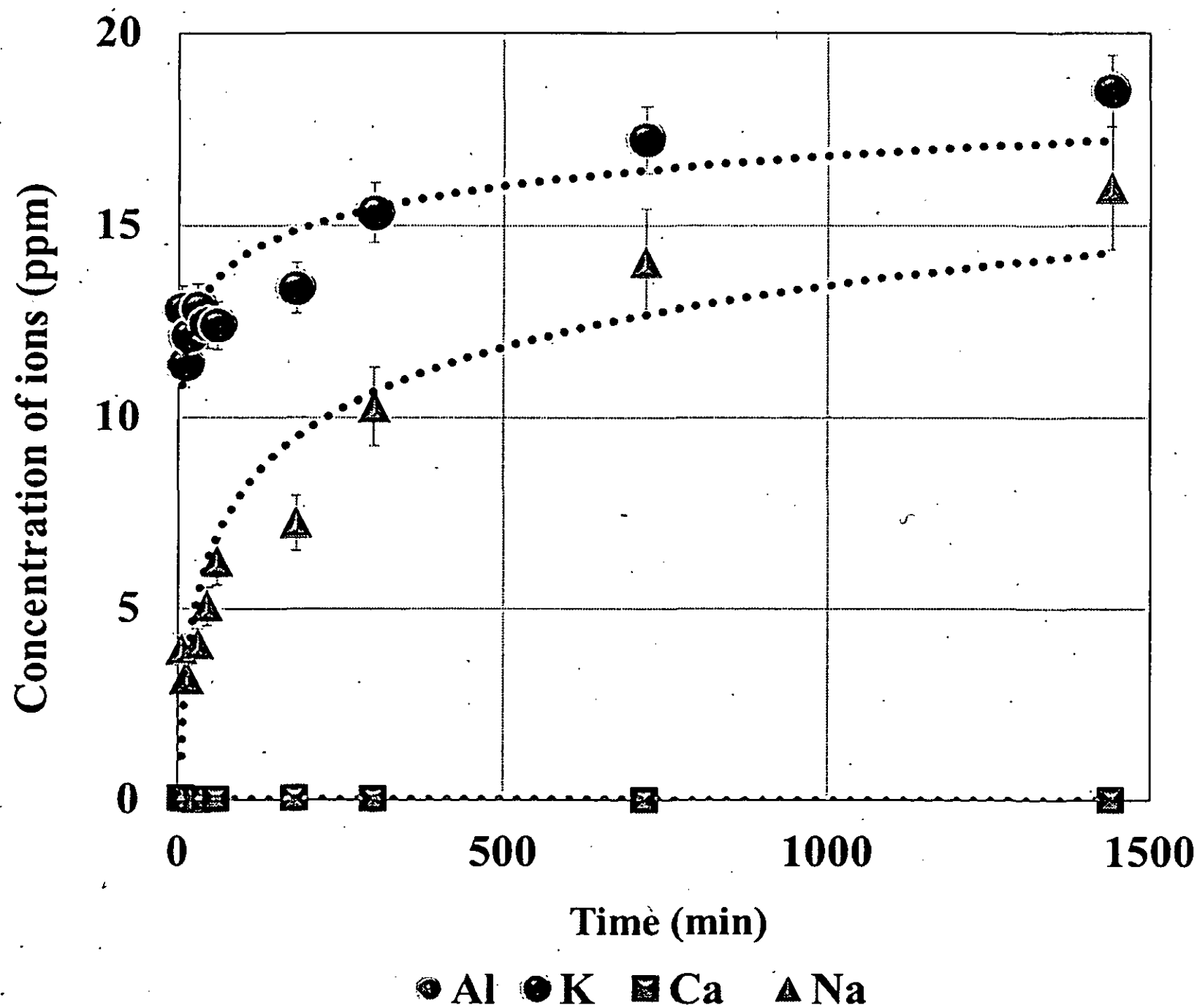


FIGURE 13

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COMPOSITION FOR SUSTAINED RELEASE OF CARBONATE AND A WATER
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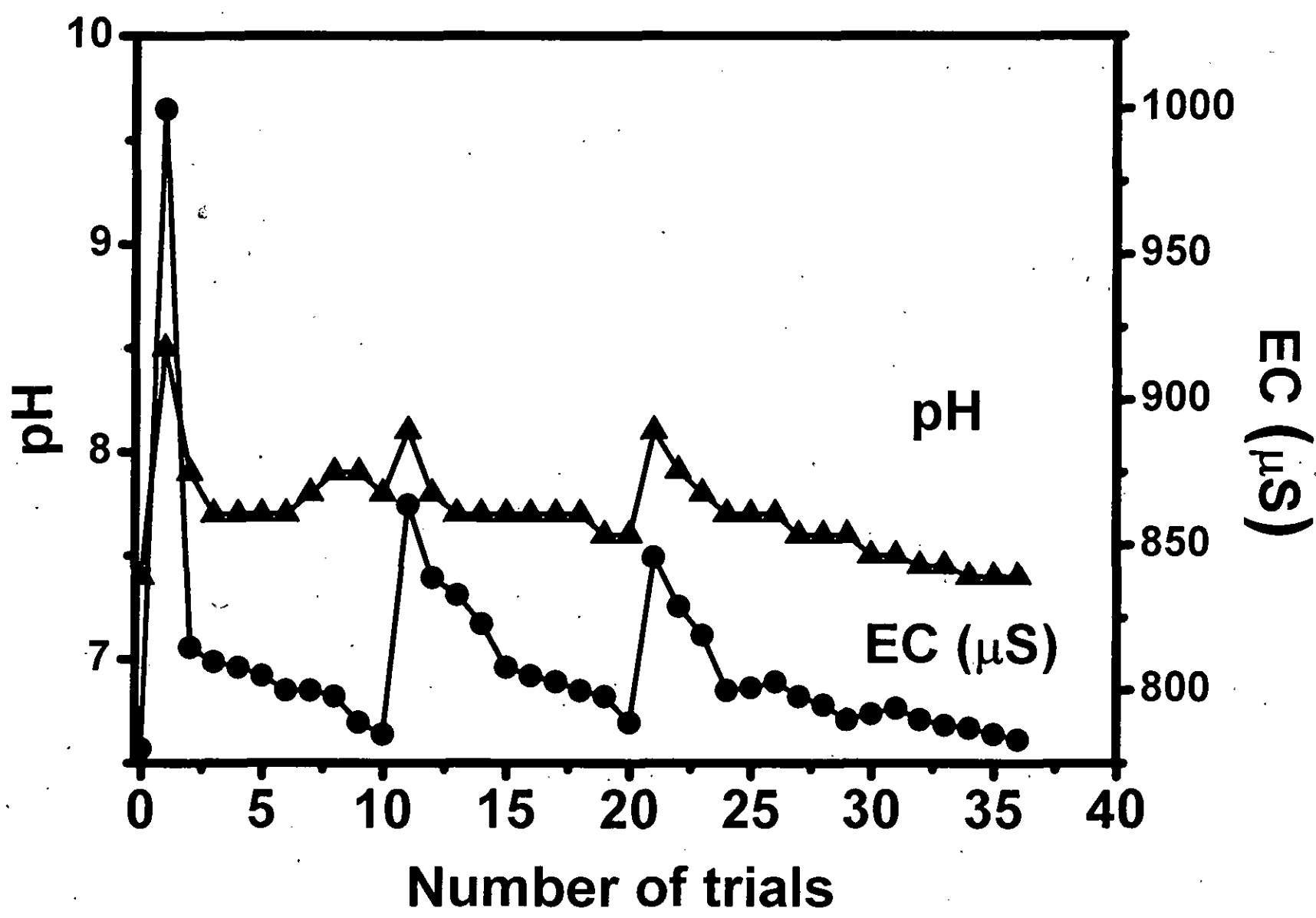


FIGURE 14

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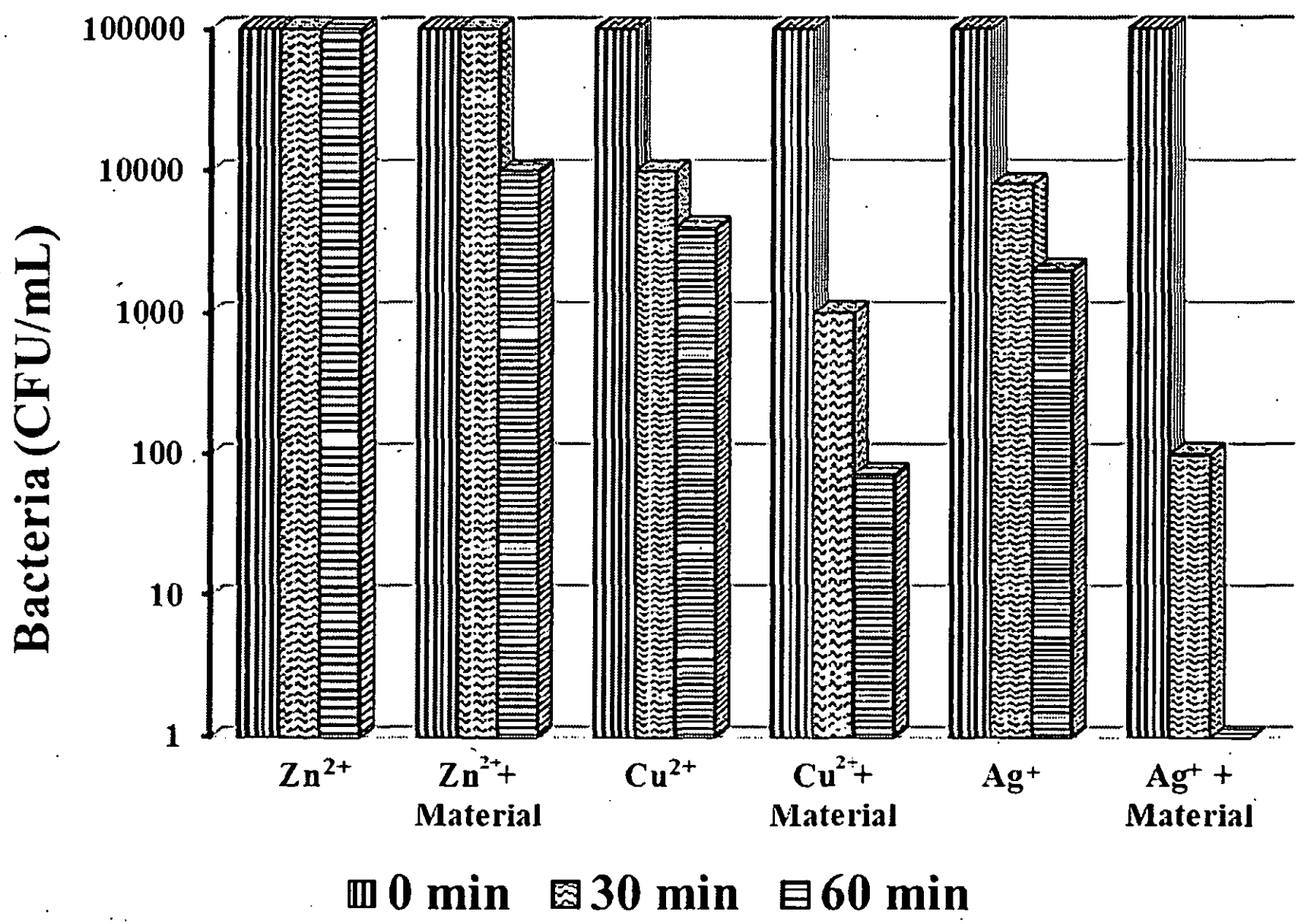
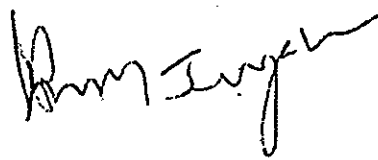


FIGURE 15

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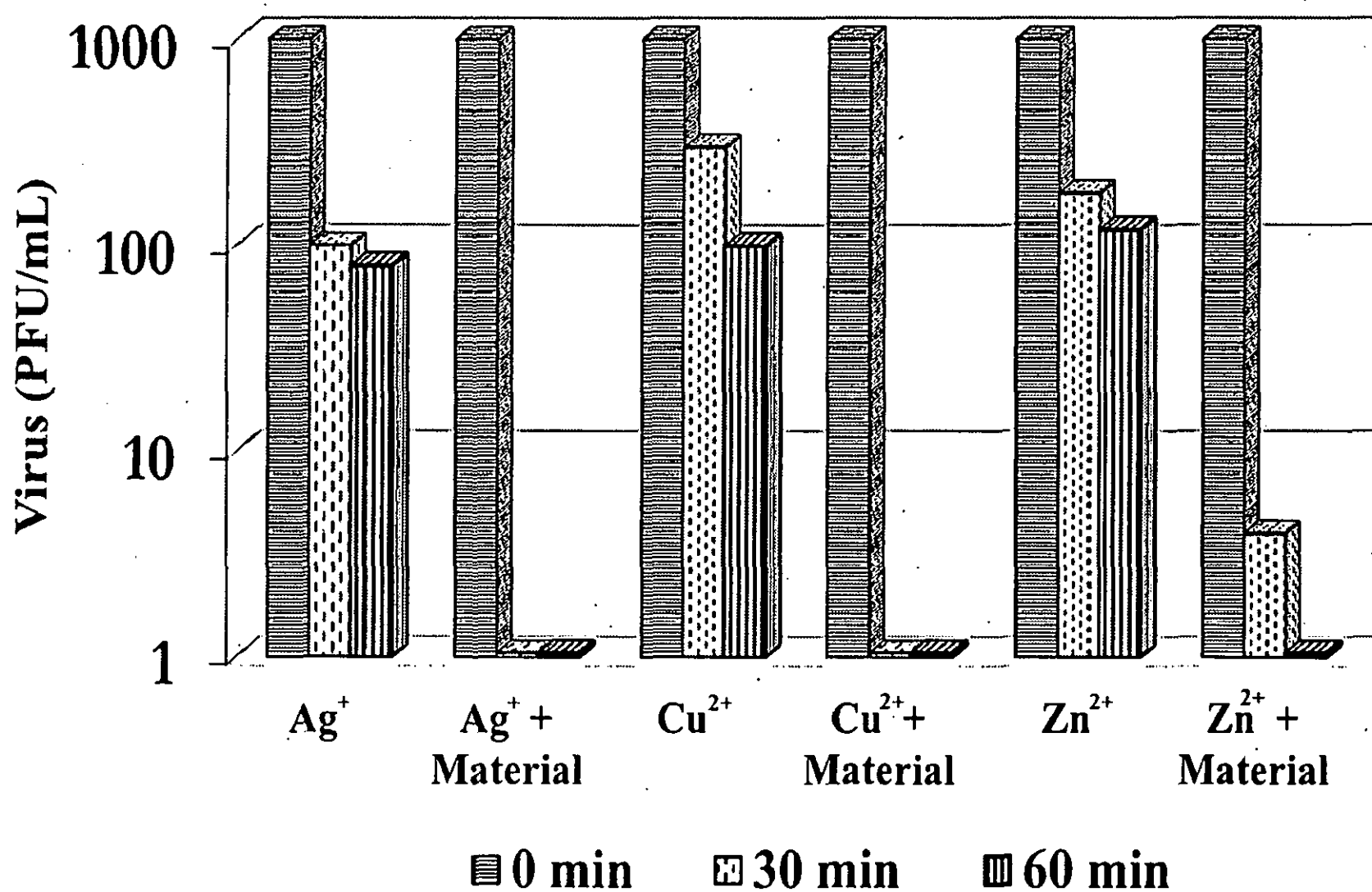


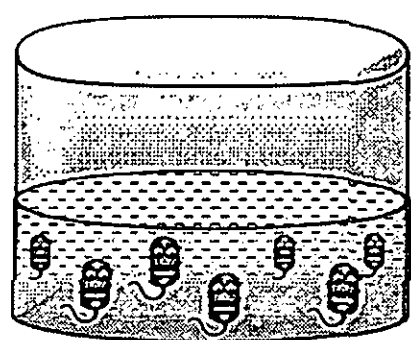
FIGURE 16

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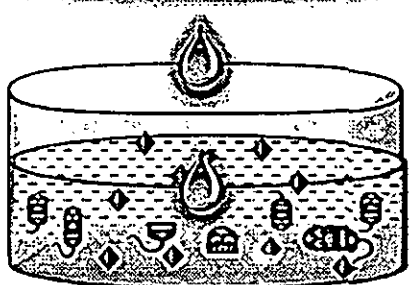
Bacteria contaminated water



Sustained CO_3^{2-} release



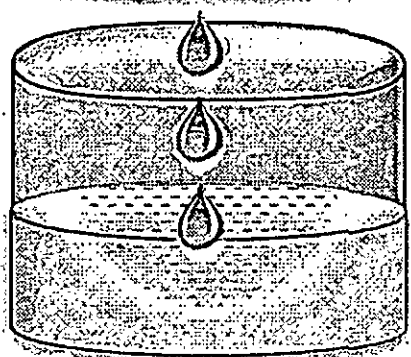
Sustained Ag^+ release



Treated water



Debris removal



Clean water

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