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The Director

of the United States Patent and Trademark Office has received an application for a patent for a new and useful invention. The title and description of the invention are enclosed. The requirements of law have been complied with, and it has been determined that a patent on the invention shall be granted under the law.

Therefore, this United States

Patent

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Katherine Kelly Vidal

DIRECTOR OF THE UNITED STATES PATENT AND TRADEMARK OFFICE

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If the application for this patent was filed on or after June 8, 1995, the term of this patent begins on the date on which this patent issues and ends twenty years from the filing date of the application or, if the application contains a specific reference to an earlier filed application or applications under 35 U.S.C. 120, 121, 365(c), or 386(c), twenty years from the filing date of the earliest such application (“the twenty-year term”), subject to the payment of maintenance fees as provided by 35 U.S.C. 41(b), and any extension as provided by 35 U.S.C. 154(b) or 156 or any disclaimer under 35 U.S.C. 253.

If this application was filed prior to June 8, 1995, the term of this patent begins on the date on which this patent issues and ends on the later of seventeen years from the date of the grant of this patent or the twenty-year term set forth above for patents resulting from applications filed on or after June 8, 1995, subject to the payment of maintenance fees as provided by 35 U.S.C. 41(b) and any extension as provided by 35 U.S.C. 156 or any disclaimer under 35 U.S.C. 253.



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(12) **United States Patent**
Shim et al.

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(45) **Date of Patent:** **Nov. 21, 2023**

(54) **LAYERED COMPOUND AND NANOSHEET CONTAINING INDIUM AND ARSENIC, AND ELECTRICAL DEVICE USING THE SAME**

(58) **Field of Classification Search**

CPC ... B82Y 30/00; C01G 15/006; C01P 2004/24; H01L 29/0665

See application file for complete search history.

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B82Y 30/00 (2011.01)

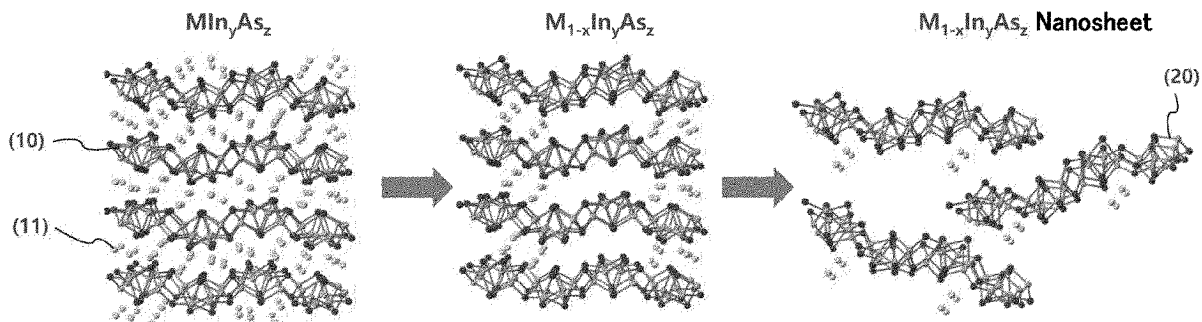
(52) **U.S. Cl.**

CPC **C01G 15/006** (2013.01); **H01L 29/0665** (2013.01); **B82Y 30/00** (2013.01); **C01P 2004/24** (2013.01)

(57) **ABSTRACT**

Proposed are a layered compound having indium and arsenic, a nanosheet that may be prepared using the same, and an electrical device including the materials. Proposed is a layered compound represented by [Formula 1] $\text{Na}_{1-x}\text{In}_y\text{As}_z$ ($0 \leq x < 1.0$, $0.8 \leq y \leq 1.2$, $1.2 \leq z \leq 1.8$).

12 Claims, 12 Drawing Sheets



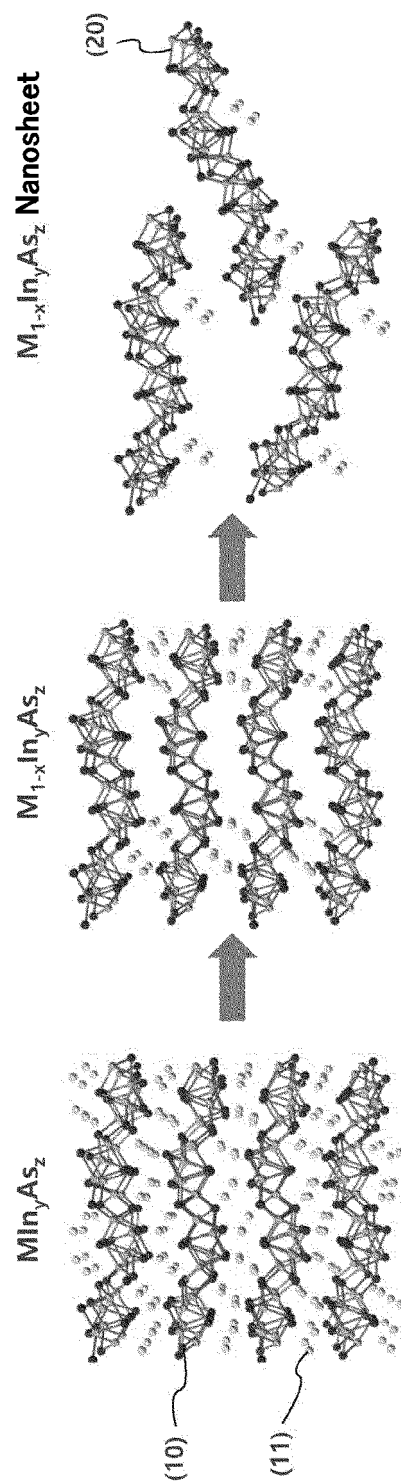


FIG. 1

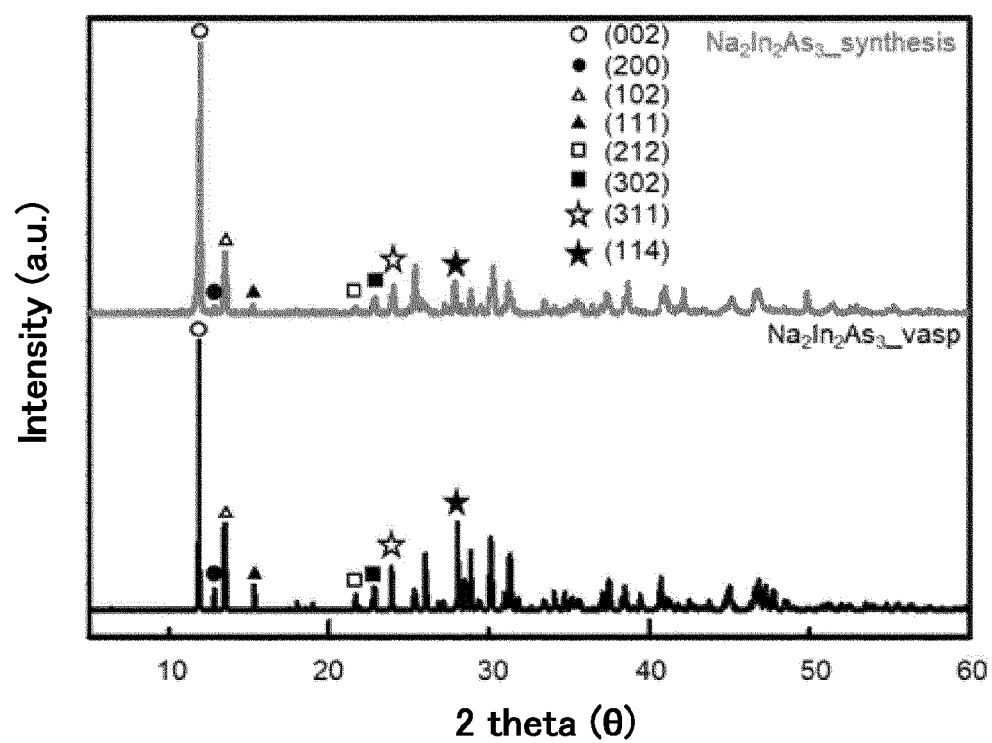
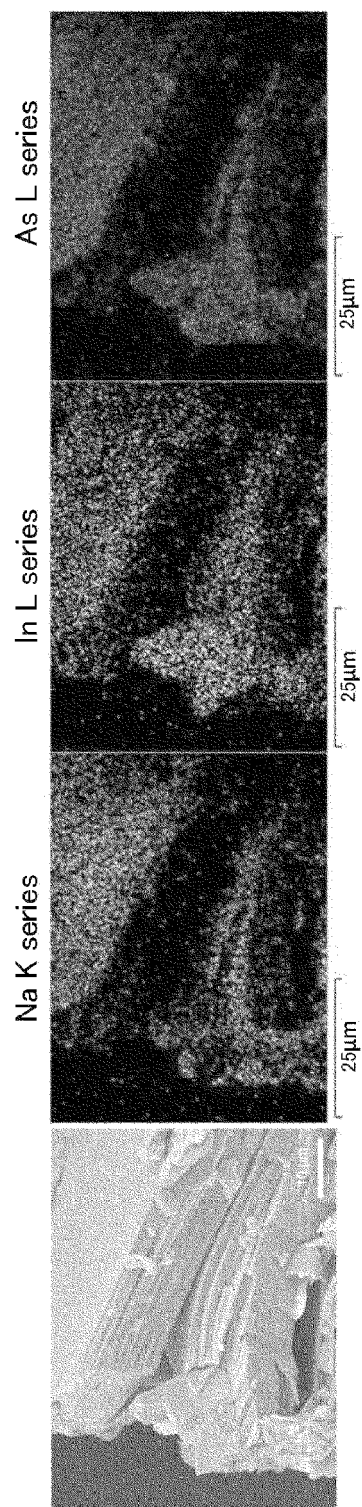


FIG. 2

**FIG. 3**

	Measurement value	Calculation value
(100)	13.755 Å	13.775 Å
(020)	3.538 Å	3.516 Å
(120)	3.22 Å	3.406 Å

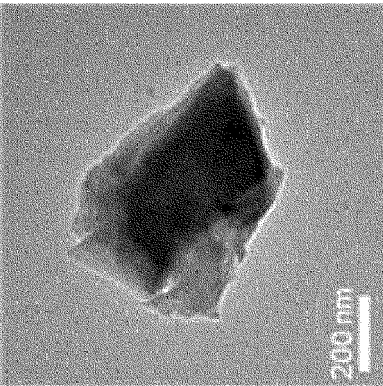
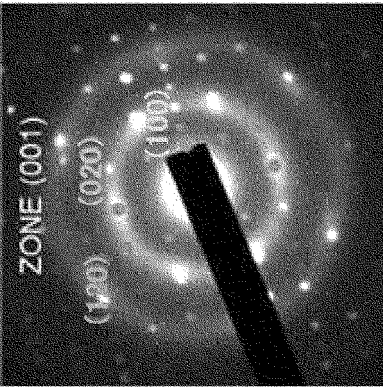


FIG. 4

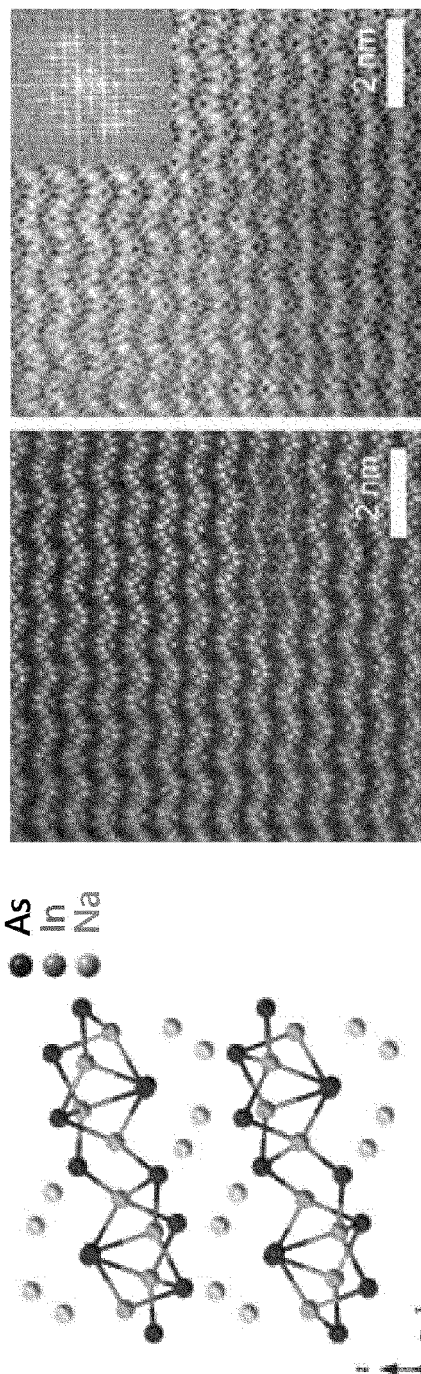


FIG. 5

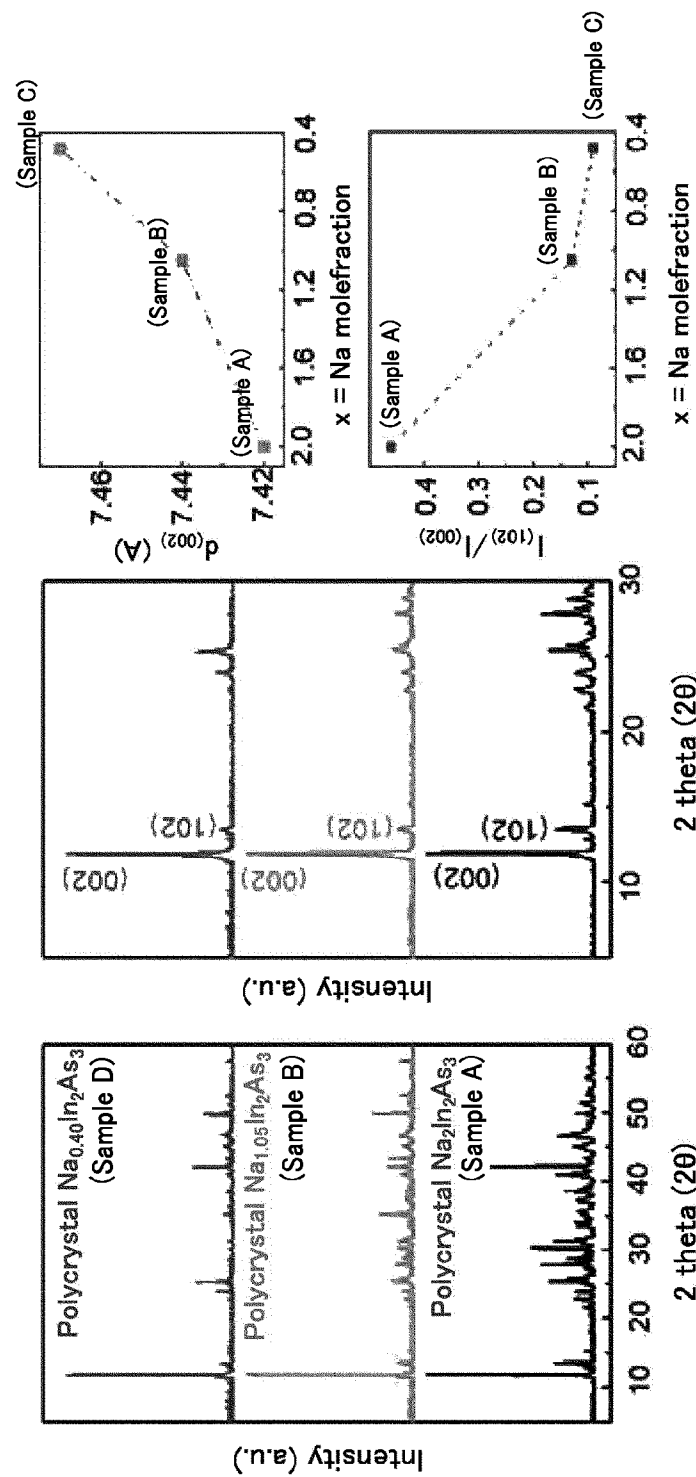


FIG. 6

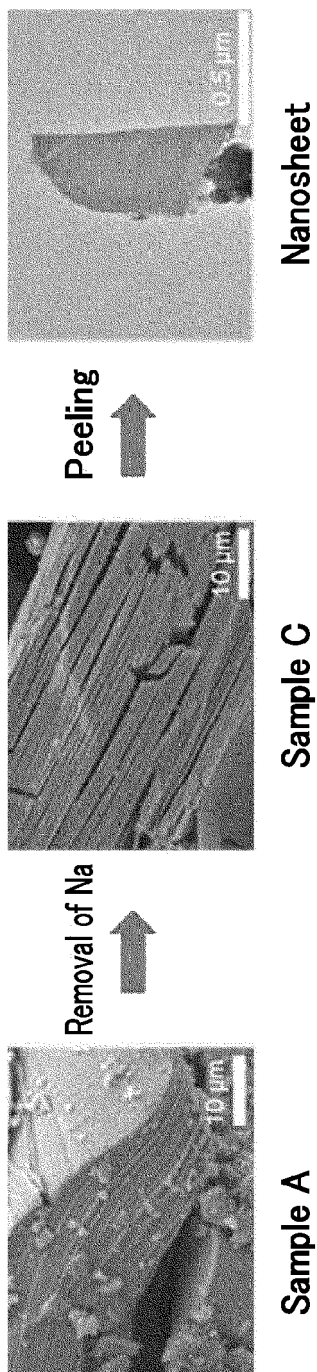
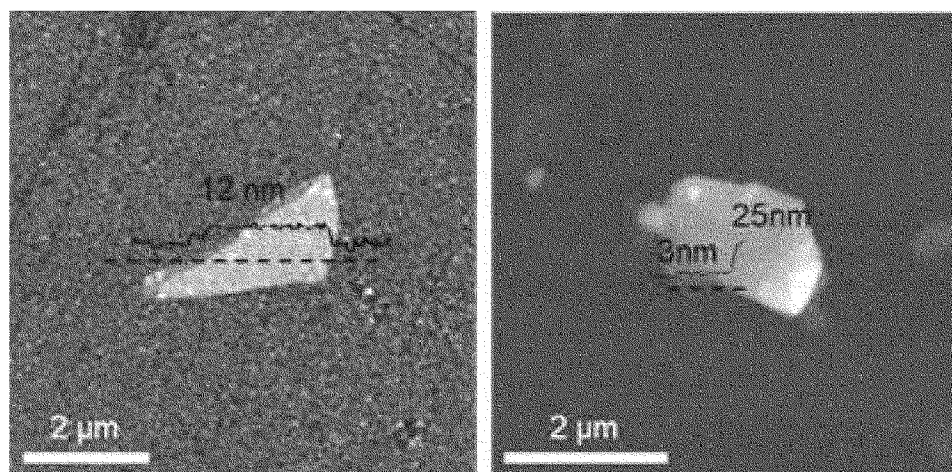


FIG. 7

**FIG. 8**

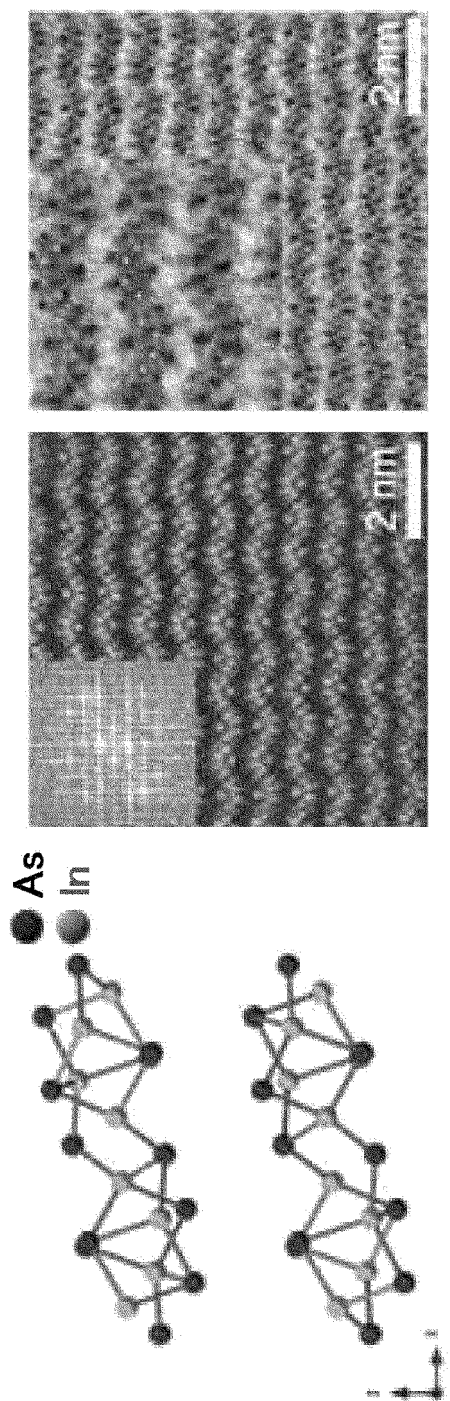


FIG. 9

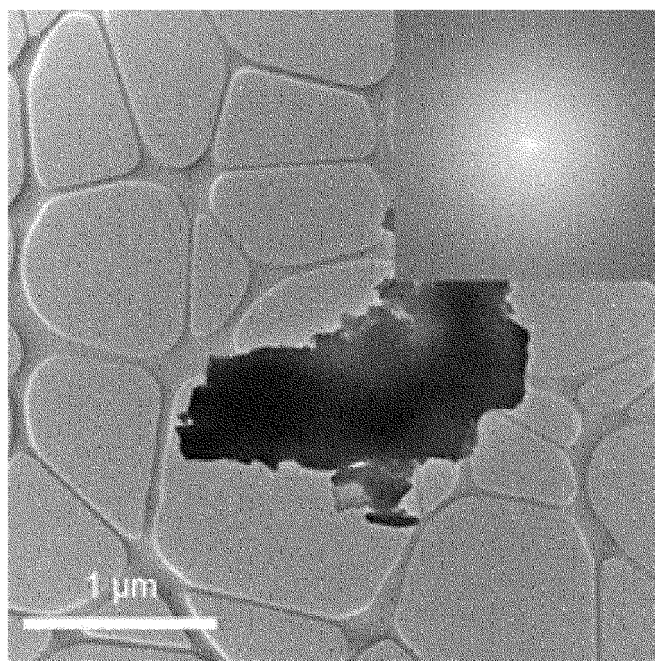
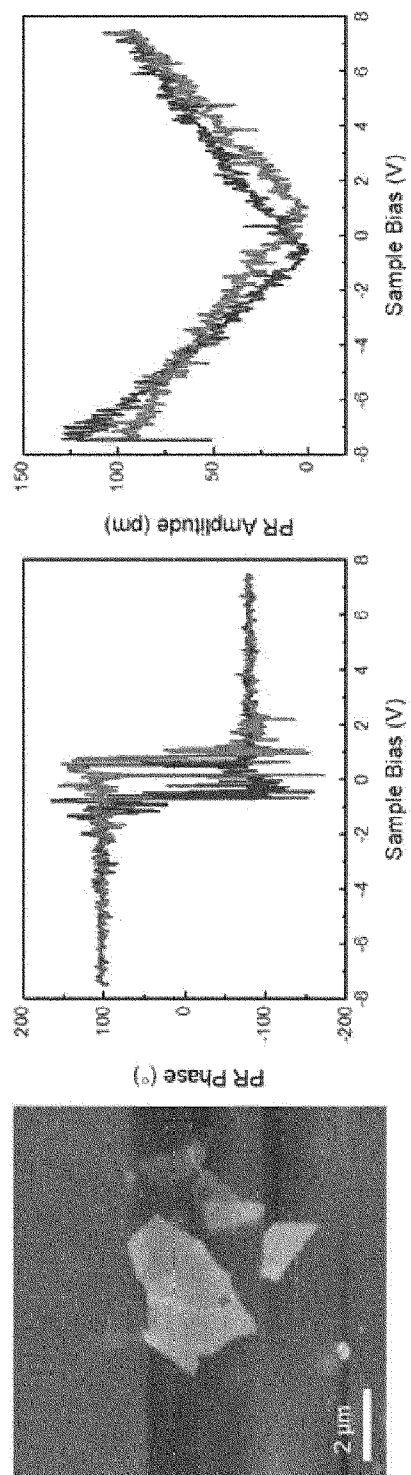


FIG. 10

**FIG. 11**

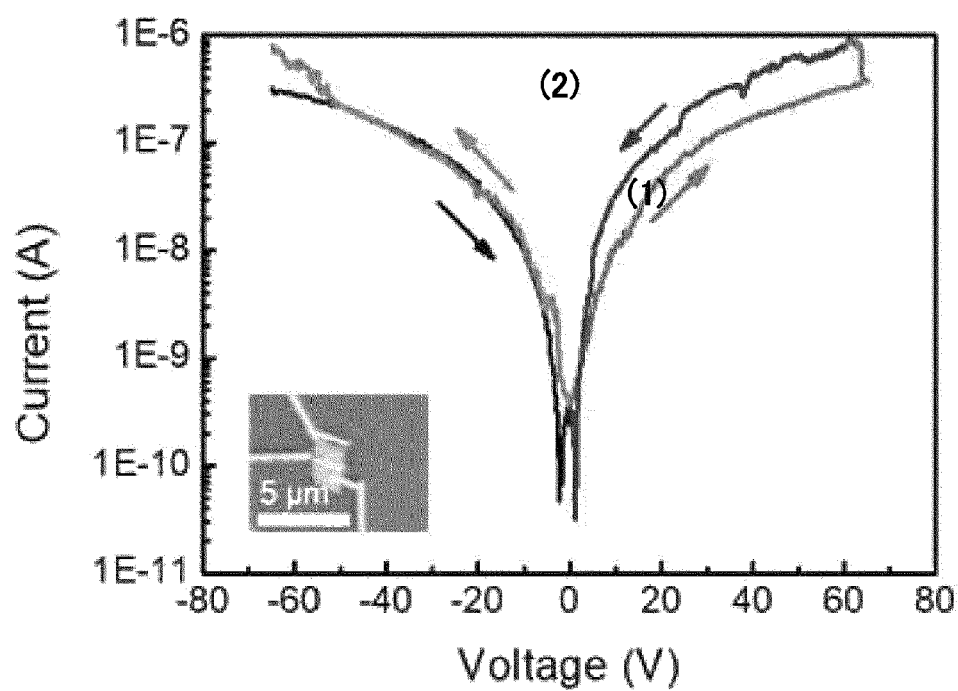


FIG. 12

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LAYERED COMPOUND AND NANOSHEET CONTAINING INDIUM AND ARSENIC, AND ELECTRICAL DEVICE USING THE SAME

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to a layered compound and a nanosheet containing indium and arsenic, and an electrical device using the same, and more particularly, to a layered compound and a nanosheet containing an alkali metal or alkaline earth metal and containing indium and arsenic having various electrical properties, and an electrical device using the same.

2. Description of the Related Art

Layered compounds connected to interlayers through van der Waals bonds may show various properties, and the layered compounds may be delaminated through physical or chemical methods to prepare two-dimensional (2D) nanosheets having a thickness of several to hundreds of nanometers, and thus, active research into the layered compounds is underway.

In particular, low-dimensional materials such as nanosheets are expected to have innovative new functions that existing bulk materials fail to provide, and are highly likely to serve as next-generation future materials instead of the existing materials.

However, up until now, the layered compounds having a two-dimensional crystal structure are limited to materials such as graphite, transition metals, and chalcogen compounds to hardly develop into materials of various compositions.

Meanwhile, indium arsenide is widely used in high-power, high-frequency electrical devices as a compound semiconductor material but ternary indium arsenide having a layered structure is not specifically known till now.

Ternary indium arsenide compounds having a layered structure, unlike existing indium arsenide compounds having a different crystal structure, are expected to allow diversified application, and to be applicable to new areas that have not been reached before.

SUMMARY OF THE INVENTION

According to an embodiment of the invention, there are provided a layered Group III-V compound having indium and arsenic, a nanosheet that may be prepared using the same, and an electrical device including the materials.

According to an embodiment of the invention, there is provided a layered compound represented by [Formula 1] $\text{Na}_{1-x}\text{In}_y\text{As}_z$ ($0 \leq x < 1.0$, $0.8 \leq y \leq 1.2$, $1.2 \leq z \leq 1.8$).

According to an embodiment of the invention, there is provided a nanosheet including a compound represented by [Formula 1] $\text{Na}_{1-x}\text{In}_y\text{As}_z$ ($0 \leq x < 1.0$, $0.8 \leq y \leq 1.2$, $1.2 \leq z \leq 1.8$), and prepared through a physical or chemical peeling method.

According to an embodiment of the invention, there is provided an electrical device including the layered compound or nanosheet as described above.

In addition, the electrical device may be a memristor.

A layered compound and a nanosheet provided through an embodiment of the invention may have various electrical properties, thereby enabling the development of new electrical devices.

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BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a conceptual diagram of a layered compound and a nanosheet prepared according to an embodiment of the invention;

FIG. 2 is a graph showing XRD diffraction patterns of a layered compound according to an embodiment of the invention;

FIG. 3 shows a scanning electron microscopy (SEM) image of a layered compound according to an embodiment of the invention and results of energy dispersive spectroscopy (EDS) analysis;

FIG. 4 shows results of transmission electron microscopy (TEM) analysis of a layered compound according to an embodiment of the invention;

FIG. 5 shows a schematic view of the structure of $\text{Na}_2\text{In}_2\text{As}_3$ according to an embodiment of the invention and results of scanning transmission electron microscopy (STEM) analysis;

FIG. 6 is shows results of XRD analysis of a layered compound according to an embodiment of the invention;

FIG. 7 is SEM and TEM images of a layered compound and a nanosheet according to an embodiment of the invention;

FIG. 8 shows an atomic force microscopy (AFM) image of a nanosheet according to an embodiment of the invention and a line-profile therefrom;

FIG. 9 shows results of STEM analysis of a layered compound according to an embodiment of the invention;

FIG. 10 shows results of TEM analysis of a layered compound according to an embodiment of the invention;

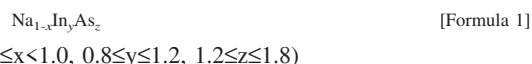
FIG. 11 shows results of evaluation on ferroelectric properties of a nanosheet according to an embodiment of the invention through piezoresponse force microscopy (PFM); and

FIG. 12 is a graph of changes in current according to voltage for a nanosheet according to an embodiment of the invention.

DETAILED DESCRIPTION OF THE EXEMPLARY EMBODIMENTS

Hereinafter, configuration and operation of embodiments of the invention will be described with reference to the accompanying drawings. In the following description, when it is determined that the specific description of the known related art unnecessarily obscures the gist of the invention, the detailed description thereof will be omitted. In addition, when an element "includes" a component, it may indicate that the element does not exclude another component unless explicitly described to the contrary, but can further include another component.

The layered compound or nanosheet according to an embodiment of the invention may be represented by Formula 1 below.



In general, InAs is a zinc blende crystal structure, which is incapable of having a layered structure, and accordingly, peeling InAs to form a nanosheet was hardly achievable.

In order to overcome the limitation, inventors of the invention have come up with an idea of adding additive elements to In_yAs_z to place the additive elements between In_yAs_z layers so as to prepare a layered compound in which the In_yAs_z layers are connected. To this end, the inventors have calculated to create a layered material having a new

composition and a crystal structure, and as a result, they have succeeded to synthesize a previously unreported new composition of a layered $\text{Na}_2\text{In}_2\text{As}_3$ so as to prepare a layered compound having a composition of Formula 1 above.

In the layered compound having the composition of Formula 1, Na is positioned between the In_yAs_z layers to weakly bond the In_yAs_z layers through van der Waals bonds, and a plane on which Na is positioned forms a cleavage plane that is easily cleaved along the plane.

Meanwhile, in the composition of Na in the $\text{Na}_{1-x}\text{In}_y\text{As}_z$ layered compound or nanosheet, x may be 0 according to Formula 1 described above, and as described above, $\text{Na}_2\text{In}_2\text{As}_3$ is a previously unreported new synthesized material where x is 0 in Formula 1. Even without the removal of Na, the plane containing Na may be peeled off as a cleavage plane that forms weak van der Waals bonds.

In the layered compound according to an embodiment of the invention, as described above, Na is positioned between the In_yAs_z layers to weakly bond the In_yAs_z layers through the van der Waals bonds, and along this cleavage plane, the plane may thus be easily peeled off into the In_yAs_z layers through either or both physical or chemical methods, and the more Na is removed, the easier the peeling is. Accordingly, through a physical or chemical peeling method, an In_yAs_z nanosheet may be easily obtained from the layered compound, and in this case, Na may partially remain in the In_yAs_z nanosheet.

With the continuous removal of the additive element Na, the distance between the In_yAs_z compound layers gradually become greater to weaken the interlayer bond force, and eventually the bond between the layers breaks down, which may cause cracks between the layers. Therefore, the layered structure of the layered compound described in the invention includes a case where repeating two-dimensional In_yAs_z layers are interlayer-bonded through van der Waals bonds by additive element Na as well as a case where the interlayer bonding force between In_yAs_z layers is removed to increase the interlayer distance, thereby causing cracks. As such, Na is removed to weaken the interlayer bond, and accordingly, easier peeling to prepare a nanosheet may be achievable.

The nanosheet prepared through the peeling from the layered compound may be a single layer of In_yAs_z , but may be formed when a plurality of layers overlaps to be several hundreds of nm thick. In general, nanosheets may exhibit anisotropy according to a two-dimensional shape only when a thickness to a lateral width is less than a certain level. To this end, the ratio of a thickness (d) to a width (L) of a nanosheet (d/L) is preferably 0.1 or less. A width of the nanosheet prepared through an embodiment of the invention may be 5 μm or greater, and thus, a thickness of the nanosheet is preferably 500 nm or less. In this case, Na may partially remain in the In_yAs_z nanosheet.

As such, the nanosheet according to an embodiment of the invention refers to a sheet peeled off from a layered compound through a physical or chemical method, and includes being formed as a plurality of In_yAs_z layers in addition to being formed as a single In_yAs_z layer.

A conceptual view of examples of the layered compound and the nanosheet is shown in FIG. 1, which shows that an additive element, Na 11, is positioned between In_yAs_z layers 10 of NaIn_yAs_z to keep the bond between the In_yAs_z layers 10, and in this case, the removal of Na 11 allows the layers to switch to $\text{Na}_{1-x}\text{In}_y\text{As}_z$, and to weaken the bond between the In_yAs_z layers 10, and thus to be easily peeled off physically or chemically, thereby, in the end, developing into

a In_yAs_z nanosheet 20. Nanosheets prepared using this way may still contain some Na 11.

Therefore, x may satisfy $0.1 \leq x \leq 0.9$ to ensure easy peeling and to prevent the breakdown of the layered structure or changes in the crystal structure due to excessive removal of Na. In this case, the crystal structure of the layered compound may have a space group of $\text{P2}_1/\text{c}$. The nanosheet peeled off from the layered compound having the range of x described above may equally satisfy $0.1 \leq x \leq 0.9$.

In addition, residual Na in the $\text{Na}_{1-x}\text{In}_y\text{As}_z$ layered compound or nanosheet may be in the range of $0.3 \leq x \leq 0.8$ according to Formula 1 below.

In the layered compound, in which an additional element, Na is partially removed and a certain amount of Na remains, Na which is an additional element remaining between the layers, becomes movable to exhibit various electrical properties. Therefore, it may be preferable that the additive element is removed in a certain fraction or greater from the $\text{Na}_{1-x}\text{In}_y\text{As}_z$ compound and the rest some remain. x for this may have a range of $0.3 \leq x \leq 0.8$.

In Formula 1, y may satisfy $0.8 \leq y \leq 1.2$, and z may satisfy $1.2 \leq z \leq 1.8$, and y and z may have slight changes due to defects in initially prepared $\text{Na}_2\text{In}_2\text{As}_3$, and the removal of Na may cause slight changes in the ratio of In to As during the removal process, and thus, values of y and z in $\text{Na}_{1-x}\text{In}_y\text{As}_z$ may change within a range that does not alter the crystal structure for a given amount of Na.

Meanwhile, a strong acid such as nitric acid or hydrochloric acid may be used for the removal of additive elements, and as the additive elements are removed through the strong acid, the place where the additive elements are removed is replaced with hydrogen ions contained in the strong acid which is then bonded thereto, and thus a layered compound containing hydrogen and a nanosheet prepared using the compound may be provided.

The layered compound containing hydrogen ions or the nanosheet therefrom may be represented by Formula 2 below.



$$(0 \leq x < 1.0, 0.8 \leq y \leq 1.2, 1.2 \leq z \leq 1.8, 0 < n \leq x)$$

In this case, hydrogen ions replace Na, an additive element, and are added in less than the amount of Na removed.

The range of x, an amount from which Na is removed, may be $0.1 \leq x \leq 0.9$, and more preferably may be $0.3 \leq x \leq 0.8$. As described above, when some of the additive elements are removed and the rest some remain, the layered structure of the initial layered compound, NaIn_yAs_z , is kept as it is, and as the additive element, Na, is partially removed, the interlayer bonding force is weakened to easily peel the compound off into the In_yAs_z layers, thereby exhibiting various electrical properties through the residual additive elements.

In addition, n above may have the same value as x, and hydrogen ions may replace the removed additive elements to be contained in the layered structure compound.

The layered compound and the nanosheet described above exhibit various properties as a result of analysis, and these properties will be described below.

The layered compound and the nanosheet described above may have a space group of $\text{P2}_1/\text{c}$ in XRD measurement using $\text{CuK}\alpha$ rays.

Meanwhile, in XRD measurement using $\text{CuK}\alpha$ rays, the layered compound or nanosheet described above may have peaks at the positions of $2\theta = 11.9^\circ \pm 0.50^\circ$, $12.8^\circ \pm 0.50^\circ$, $13.5^\circ \pm 0.50^\circ$, $15.3^\circ \pm 0.50^\circ$, $21.6^\circ \pm 0.50^\circ$, $22.7^\circ \pm 0.50^\circ$, $23.8^\circ \pm 0.50^\circ$, and $27.8^\circ \pm 0.50^\circ$, and the peaks may have an

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intensity of 1% or greater (preferably 3% or greater, more preferably 5% or greater) with respect to a peak having the greatest intensity.

Meanwhile, as the additive elements are removed from the layered compound or nanosheet, slight changes in the XRD measurement peak may be observed, and according to the changes, in the XRD measurement using CuK α rays, the layered compound has a $I_{(102)}/I_{(002)}$ value of 0.40 or less which is a peak intensity of a (102) plane to a peak intensity of a (002) plane. This is caused when the interlayer distance gradually increases due to the removal of the additive elements from the layered compound, and the same is true for the nanosheet.

The layered compound in which the additional element, Na partially remains, and the nanosheet using the compound may exhibit various electrical properties due to the residual Na.

The layered compound or nanosheet as described above may exhibit various electrical properties due to a unique layered structure and residual additive elements.

First, the layered structure compound or the nanosheet according to an embodiment of the invention exhibits ferroelectric-like properties.

Ferroelectric properties are generally found in oxides of an asymmetric structure such as BaTiO₃ of a perovskite structure, and are found according to changes in the position of Ba located at the center.

However, the layered structure compound or the nanosheet according to an embodiment of the invention does not have the asymmetric structure, but nevertheless exhibits ferroelectric properties. Despite the fact that the layered structure compound or the nanosheet does not have an asymmetrical structure, the layered structure compound or the nanosheet still exhibit ferroelectric-like properties since the position of the residual additive elements moves according to an external electric field.

The ferroelectric-like properties of the layered compound or nanosheet according to an embodiment of the invention enables application to various electrical devices.

In addition, the layered structure compound or the nanosheet according to an embodiment of the invention may exhibit resistance switching properties.

When a material has resistance switching properties, current does not increase linearly according to voltages applied to the material, but when an initial voltage is applied, the material keeps a high resistance state to have an insignificant increase in the current and then when the material reaches a certain critical point, the material switches to a low resistance state to have a sharp increase in the current.

These resistance switching properties are generally found in oxides, and recently, using these properties, memory devices such as a memristor capable of storing information like a flash memory have been actively developed, and, through the resistance switching properties, the layered compound and the nanosheet of an embodiment of the invention may be actively used in the development of memory devices such as the memristor.

Example

1) Synthesis of Na₂In₂As₃ Having a Layered Structure

Na, In, and As were weighed at a molar ratio of 2:2:3, mixed, and then put into an alumina crucible. Next, the mixture was placed in a quartz tube which was then double-sealed to block outside air. The process was performed in a glove box under argon atmosphere. Thereafter, the resultant was put at a temperature raised to 1000° C. in a box furnace,

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kept for 12 hours, cooled to 500° C. at a temperature reduction rate of 5° C./h, then kept for 100 hours at 500° C., and cooled to room temperature to obtain Na₂In₂As₃.

2) Removal of Na

The layered Na₂In₂As was subjected to reaction over time in a 0.25M HCl solution diluted with ethanol to remove Na therefrom. The results are shown in the table below. In Table 1, the residual Na represents the results obtained through EDS analysis.

TABLE 1

Name of sample	Removal of additive elements	Reaction time	Residual Na (at %)
Sample A	—	—	28
Sample B	Hydrochloric acid	0.5 hours	17.4
Sample C	Hydrochloric acid	1 hour	10.7
Sample D	Hydrochloric acid	1.5 hours	8.8
Sample E	Hydrochloric acid	4 hours	1.5

3) Process of Preparing Nanosheets

The samples prepared as in Table 1 above were irradiated with ultrasonic waves in ethanol to prepare peeled nanosheets using a tape.

The inventors have calculated to project a layered structure using vienna ab initio simulation package (VASP) for a previously unreported new Na₂In₂As₃ compound, and as a result, they have found out that the layered structure had a structure of P2₁/c similar to known Na₂Al₂As₃ and Na₂Ga₂As₃.

FIG. 2 shows XRD diffraction patterns (Na₂In₂As₃_vasp) of Na₂In₂As₃ projected through the calculation using VASP and XRD rotation patterns of Sample A (Na₂In₂As₃_synthesis) synthesized through the method described above. When comparing the peaks of the calculated data with the peaks of the data for Sample A, which is an actual synthesized compound, it was found that (002), (200), (102), (111), (212), (302), (311), and (114) were detected. The 2 θ angles of the planes were 11.9°, 12.8°, 13.5°, 15.3°, 21.6°, 22.7°, 23.8°, and 27.8°, respectively.

FIG. 3 shows a scanning electron microscopy (SEM) image of the synthesized Sample A and results of energy dispersive spectroscopy (EDS) analysis. From the EDS results, it was found that the synthesized Sample A was composed of Na, In, and As.

FIG. 4 shows results of transmission electron microscopy (TEM) analysis of Sample A. As a result of selected area electron diffraction (SAED) analysis through TEM for Sample A, patterns in which the space group of P2₁/c was present in the (001) direction were measured, and the distance between each (100), (020), and (120) plane was found to be similar in calculation and measurements.

FIG. 5 shows a schematic view of the structure of Na₂In₂As₃ and results of scanning transmission electron microscopy (STEM) analysis. The results of STEM analysis found that the synthesized Sample A had a P2₁/c space group.

As such, the results of FIGS. 2 to 5 found that the synthesized Sample A was Na₂In₂As₃, a layered material having a new composition and a crystal structure having a P2₁/c space group.

FIG. 6 shows changes in XRD peaks according to the removal of Na. In Sample A in which Na was not removed, the interplanar distance of the (002) plane was 7.42 Å, and as Na was removed, the interplanar distance gradually increased up to 7.47 Å. Changes in the XRD peaks as well were observed according to the changes in the interplanar

distance, and it was found that with the removal of Na, the size of the peaks of the (102) plane to the peaks of the (002) plane gradually decreased. Accordingly, the value of $I_{(102)}/I_{(002)}$ was 0.46 in Sample A, decreased to 0.13 in Sample B and to 0.09 in Sample D. In addition, when the XRD peaks were compared after the removal of Na, the (002) and (102) planes showed the same peaks, indicating that the crystal structure having a $P2_1/c$ space group was kept.

FIG. 7 shows a nanosheet prepared by removing Na from Sample A to become Sample C, and being peeled off from Sample C using a tape. In Sample A, a cleavage plane between the layers was observed, but in Sample C, as Na was removed, the interlayer distance increased, thereby forming cracks.

FIG. 8 shows an atomic force microscopy (AFM) image of a nanosheet prepared by being peeled off from Sample C and a line-profile therefrom. It was confirmed that a nanosheet was peeled off to have a thickness of 10 nm to 30 nm.

FIG. 9 shows results of scanning transmission electron microscopy (STEM) analysis of Sample D. The data showed that Na was partially removed and there were no changes in the crystal structure even after the removal.

FIG. 10 is results of TEM analysis of Sample E. It was found that an amorphous structure appeared with the excessive removal of Na.

Ferroelectric properties were measured through piezoresponse force microscopy (PFM) for the nanosheet peeled off from Sample C, and the results are shown in FIG. 11. It was found that the nanosheet had similar ferroelectric-like properties.

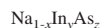
In addition, current changes according to voltages were measured for the nanosheet peeled off from Sample C, and results are shown in FIG. 12.

It was found that at an initial voltage, the nanosheet kept a high resistance state 1, indicating a low current flow, but when the voltage was greater than a certain level, the nanosheet switched to a low resistance state 2, indicating a sharp increase in the current, and the same properties were shown in an opposite electrode direction, thereby showing resistance switching properties.

It was found that using the resistance switching properties, the nanosheet would be applied as a memristor device, which is being actively developed as a neuromorphic memory device.

What is claimed is:

1. A layered compound represented by Formula 1 below:



($0.3 \leq x \leq 0.8$, $0.8 \leq y \leq 1.2$, $1.2 \leq z \leq 1.8$) wherein the layered compound exhibits ferroelectric-like properties.

2. The layered compound according to claim 1, wherein the layered compound further comprises H.

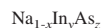
3. The layered compound according to claim 1, wherein, in XRD measurement using $\text{CuK}\alpha$ rays, the layered compound has peaks at positions of $2\theta = 11.9^\circ \pm 0.50^\circ$, $12.8^\circ \pm 0.50^\circ$, $13.5^\circ \pm 0.50^\circ$, $15.3^\circ \pm 0.50^\circ$, $21.6^\circ \pm 0.50^\circ$, $22.7^\circ \pm 0.50^\circ$, $23.8^\circ \pm 0.50^\circ$, and $27.8^\circ \pm 0.50^\circ$, the peaks having an intensity of 1% or greater with respect to a peak having the greatest intensity.

4. The layered compound according to claim 1, wherein a crystal structure of the layered compound represents a space group of $P2_1/c$.

5. The layered compound according to claim 1, wherein, in XRD measurement using $\text{CuK}\alpha$ rays, the layered compound has a $I_{(102)}/I_{(002)}$ value of 0.40 or less which is a peak intensity of a (102) plane to a peak intensity of a (002) plane.

6. The layered compound according to claim 1, wherein the layered compound exhibits resistance switching properties.

7. A nanosheet comprising a compound represented by Formula 1 below, and prepared through a physical or chemical peeling method:



($0.3 \leq x \leq 0.8$, $0.8 \leq y \leq 1.2$, $1.2 \leq z \leq 1.8$)

wherein the compound exhibits ferroelectric-like properties.

8. The nanosheet according to claim 7, wherein a crystal structure of the compound exhibits a space group of $P2_1/c$.

9. The nanosheet according to claim 7, wherein, in XRD measurement using $\text{CuK}\alpha$ rays, the compound has a $I_{(102)}/I_{(002)}$ value of 0.40 or less which is a peak intensity of a (102) plane to a peak intensity of a (002) plane.

10. The nanosheet according to claim 7, wherein the compound exhibits resistance switching properties.

11. The nanosheet according to claim 7, wherein the nanosheet has a thickness of 500 nm or less.

12. An electrical device comprising the layered compound according to claim 1.

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