

SEARCH REPORT

Application Number

LH 21
LT 2020535

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
A	SUHAIL A ET AL: "Reduction of polymer residue on wet-transferred CVD graphene surface by deep UV exposure", APPLIED PHYSICS LETTERS, A I P PUBLISHING LLC, US, vol. 110, no. 18, 4 May 2017 (2017-05-04), XP012218603, ISSN: 0003-6951, DOI: 10.1063/1.4983185 [retrieved on 2017-05-04] * abstract; bridging paragraph of p. 183103-1, right column-p.183103-2, left column; figure 1(a) to 1(c) * -----	1-4	INV. C01B32/194 H01L21/027 B82Y30/00 C23C16/26 C23C16/56
A	YANG XIAOJIAN ET AL: "Removing contaminants from transferred CVD graphene", NANO RESEARCH, TSINGHUA UNIVERSITY PRESS, CN, vol. 13, no. 3, 14 February 2020 (2020-02-14), pages 599-610, XP037271023, ISSN: 1998-0124, DOI: 10.1007/S12274-020-2671-6 [retrieved on 2020-02-14] * page 600, left column, paragraph 2-3; Scheme 1; 4.3 Degradation of PMMA * ----- -/--	1-4	TECHNICAL FIELDS SEARCHED (IPC) C01B G03F H01L B82Y C23C
The present search report has been drawn up for all claims			
Munich		Date of completion of the search 19 March 2021	Examiner Eggers, Karin
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>			

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EPO FORM 1503 11.08 (P04C80)

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A	JEONG HEE JIN ET AL: "Improved transfer of chemical-vapor-deposited graphene through modification of intermolecular interactions and solubility of poly(methylmethacrylate) layers", CARBON, ELSEVIER OXFORD, GB, vol. 66, 29 September 2013 (2013-09-29), pages 612-618, XP028758983, ISSN: 0008-6223, DOI: 10.1016/J.CARBON.2013.09.050 * the whole document *	1-4	TECHNICAL FIELDS SEARCHED (IPC)
A	KIM JIN HONG ET AL: "Facile Dry Surface Cleaning of Graphene by UV Treatment", JOURNAL OF THE KOREAN PHYSICAL SOCIETY, KOREAN PHYSICAL SOCIETY, KR, vol. 72, no. 9, 28 April 2018 (2018-04-28), pages 1045-1051, XP036490371, ISSN: 0374-4884, DOI: 10.3938/JKPS.72.1045 [retrieved on 2018-04-28] * I.INTRODUCTION, II EXPERIMENTAL PROCEDURE *	1-4	
A	CN 102 849 732 B (UNIV BEIJING) 7 January 2015 (2015-01-07) * paragraphs [0004] - [0011]; claim 1; figure 1 *	1-4	
The present search report has been drawn up for all claims			
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19-03-2021

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
CN 102849732 B	07-01-2015	NONE	

WRITTEN OPINION

File No. LH21	Filing date (<i>day/month/year</i>) 08.07.2020	Priority date (<i>day/month/year</i>)	Application No. LT2020535
International Patent Classification (IPC) INV. C01B32/194 H01L21/027 B82Y30/00 C23C16/26 C23C16/56			
Applicant Valstybinis mokslinių tyrimų institutas Fizinių ir technologijos mokslų centras			

This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of the opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the application
- ☒ Box No. VIII Certain observations on the application

	Examiner Eggers, Karin
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WRITTEN OPINION**Box No. I Basis of this opinion**

1. This opinion has been established on the basis of the latest set of claims filed before the start of the search.
2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the application, this opinion has been established on the basis of:
 - a. type of material:
 - ☐ a sequence listing
 - ☐ table(s) related to the sequence listing
 - b. format of material:
 - ☐ on paper
 - ☐ in electronic form
 - c. time of filing/furnishing:
 - ☐ contained in the application as filed.
 - ☐ filed together with the application in electronic form.
 - ☐ furnished subsequently for the purposes of search.
3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
4. Additional comments:

Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	1-4
	No: Claims	
Inventive step (IS)	Yes: Claims	1-4
	No: Claims	
Industrial applicability (IA)	Yes: Claims	1-4
	No: Claims	

2. Citations and explanations

see separate sheet

WRITTEN OPINION

Application number
LT2020535

Box No. VIII Certain observations on the application

see separate sheet

Re item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1 Reference is made to the following documents:

- D1 SUHAIL A ET AL: "Reduction of polymer residue on wet-transferred CVD graphene surface by deep UV exposure",
APPLIED PHYSICS LETTERS, A I P PUBLISHING LLC, US,
vol. 110, no. 18, 4 May 2017 (2017-05-04), XP012218603,
ISSN: 0003-6951, DOI: 10.1063/1.4983185
[retrieved on 2017-05-04]
- D2 YANG XIAOJIAN ET AL: "Removing contaminants from transferred CVD graphene",
NANO RESEARCH, TSINGHUA UNIVERSITY PRESS, CN,
vol. 13, no. 3, 14 February 2020 (2020-02-14), pages 599-610,
XP037271023,
ISSN: 1998-0124, DOI: 10.1007/S12274-020-2671-6
[retrieved on 2020-02-14]
- D3 JEONG HEE JIN ET AL: "Improved transfer of chemical-vapor-deposited graphene through modification of intermolecular interactions and solubility of poly(methylmethacrylate) layers",
CARBON, ELSEVIER OXFORD, GB,
vol. 66, 29 September 2013 (2013-09-29), pages 612-618,
XP028758983,
ISSN: 0008-6223, DOI: 10.1016/J.CARBON.2013.09.050
- D4 KIM JIN HONG ET AL: "Facile Dry Surface Cleaning of Graphene by UV Treatment",
JOURNAL OF THE KOREAN PHYSICAL SOCIETY, KOREAN PHYSICAL SOCIETY, KR,
vol. 72, no. 9, 28 April 2018 (2018-04-28), pages 1045-1051,
XP036490371,
ISSN: 0374-4884, DOI: 10.3938/JKPS.72.1045
[retrieved on 2018-04-28]
- D5 CN 102 849 732 B (UNIV BEIJING) 7 January 2015 (2015-01-07)

- 1.1 In D1 (bridging § of p. 183103-1, right column-p. 183103-2, left column), it is described a process of producing a graphene monolayer on a copper foil by a CVD process, the subsequent coating of a PMMA layer onto the graphene monolayer/copper foil to form a PMMA/graphene/copper multi film structure, the subsequent removal of the copper foil by wet-etching so that a PMMA/graphene multi film structure remains, the wet-transfer of the PMMA/graphene multi film structure onto a SiO₂/Si substrate so that a PMMA/graphene/SiO₂/Si multi film structure is formed, and the subsequent removal of the PMMA transfer film by the following procedure steps:
- irradiation of the PMMA film of the PMMA/graphene/SiO₂/Si multifilm structure with DUV (wavelength of 254 nm) light,
 - treatment of the PMMA/graphene/SiO₂/Si multifilm structure with an acetone solution to dissolve the irradiated PMMA,
 - rinse of the graphene/SiO₂/Si multifilm structure with a mixture of isopropanol/deionized water (IPA and DIW), and
 - drying the graphene/SiO₂/Si multifilm structure in vacuum atmosphere.
- 1.2 D2 is related to the removal of contaminants, notably of the PMMA transfer film from transferred CVD graphene layers. In the document, different techniques for reducing the amount of contaminants onto transferred graphene layers are described.
- On p. 600, left column, §2-3 and Scheme 1, it is described the general procedure of the fabrication and transfer a graphene layer onto a target substrate, whereas a PMMA film is used as a transfer film. It is also disclosed the routine procedure of the removal of the PMMA film in an acetone bath after the graphene layer has been transferred onto a target substrate.
- In table 1, the authors of D2 have further collected different methods to minimize PMMA and other residuals on transferred CVD graphene layers. Amongst others, the pretreatment of the PMMA film with UV radiation, ion beam or heat before the PMMA is finally stripped off in a solvent, is mentioned.
- Under item "4.3 Degradation of PMMA", the degradation of the PMMA film by DUV light (254 nm light) followed by a removal in a solvent solution (e.g. acetone) is described in more details.
- 1.3 D3 is related to an improved transfer of CVD graphene through modification of intermolecular interactions and solubility of poly(methylmethacrylate) layers. It ("2.1. Graphene growth and transfer", fig. 1(a) to (c)) discloses the coating of a 1st PMMA layer onto a graphene layer which is deposited onto a copper film,

the DUV irradiation (λ 256 nm) of the 1st PMMA layer, the subsequent coating of a 2nd sacrificial PMMA layer onto the first PMMA layer, the etch removal of the copper film, the transfer of the graphene layer together with two PMMA layers onto a target substrate, and the final removal of the PMMA layers in a mixed solvent composed of isopropyl alcohol (IPA), acetone, and methyl isobutyl ketone (MIBK).

- 1.4 In D4, it is described a method of facile dry surface cleaning of graphene containing PMMA residuals by DUV treatment. Under item "II. EXPERIMENTAL PROCEDURE", it is described that after the transfer of the graphene monolayer onto a SiO₂ substrate, the PMMA layer is irradiated with UV-C radiation using a Xe lamp under atmospheric condition and by controlling the irradiation time.
- 1.5 D5 (claim 1, fig. 1) is related to a method realizing the asymmetric covalent modification of a single-layer graphene. It (claim 1) discloses a procedure of removing the PMMA layer from a graphene surface utilizing an acetone steam.

- 2 The removal method of the polymethylmethacrylate from a graphene surface of claim 1 differs at least from the methods disclosed in D1-D4 in that the treatment after the DUV irradiation is carried out in a mixture of an alcohol and deionized water.

The PMMA removal procedure of D5 neither discloses the DUV irradiation step with a UV-C band source nor the post treatment with an alcohol/deionized water mixture as defined in the method of claim 1.

Thus, it is shown that the subject-matter of claim 1 is novel over the above cited prior art.

- 3 The problem to be solved by the present invention is a further improved easy usable method for removing PMMA from a graphene surface.

The removal method of claim 1 comprises an inventive step over the above prior art because none of D1-D5 discloses a development step of the DUV irradiated PMMA/graphene/substrate with a mixture of an alcohol and deionized deionized water. Therefore, any combination of the teaching of D1 to D5 would not arrive at the removal method as described in claim 1.

- 4 Claims 2-4 are directly or indirectly dependent on claim 1 and as such also meet the requirements of novelty and inventive step.

Re Item VIII

Certain observations on the application

- 5 The wording of claim 1 is not very clear and contains several clerical errors. The applicants are therefore asked to revise **the whole wording** without departing from the scope of the originally filed application.

Notably, the following part "subjecting the PMMA layer on graphene onto substrate" of the claim wording is unclear.

For example, the description, p. 4, l. 13-33 may be taken as a basis for the revision of the claim wording.

The present deficient claim wording may impede a direct grant of the application in the regional phase.

- 6 Also, the term "polimethylmethicrylate" used in the title, the description and in the claims should be replaced by "polymethylmethacrylate" (clerical error).
- 7 Figure 5 is illegible and should be replaced by a better copy.