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(54) **GRAPHENE NANOPLATELETS DERIVED FROM THERMOMECHANICAL EXFOLIATION OF GRAPHITE**

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(58) **Field of Classification Search**
CPC C01B 32/19; C01B 32/194–2204; C01B 32/04; C09C 1/44; C01P 2002/72; C01P 2002/82; C01P 2002/85; C01P 2002/88; C01P 2004/03; C01P 2004/04; C01P 2006/40

(73) Assignee: **Queen's University at Kingston**, Kingston (CA)

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(21) Appl. No.: **16/962,073**

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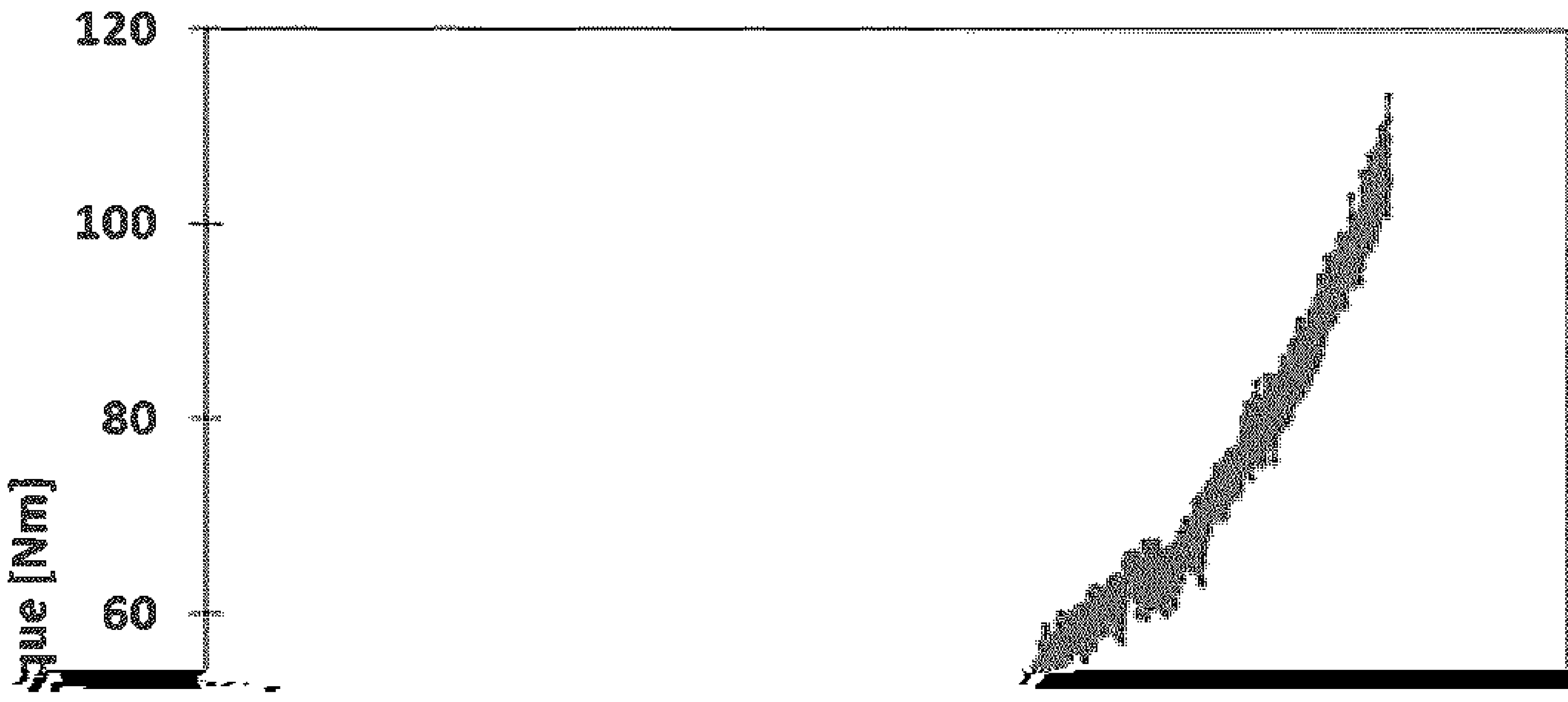
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100
80
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que [Nm]


A
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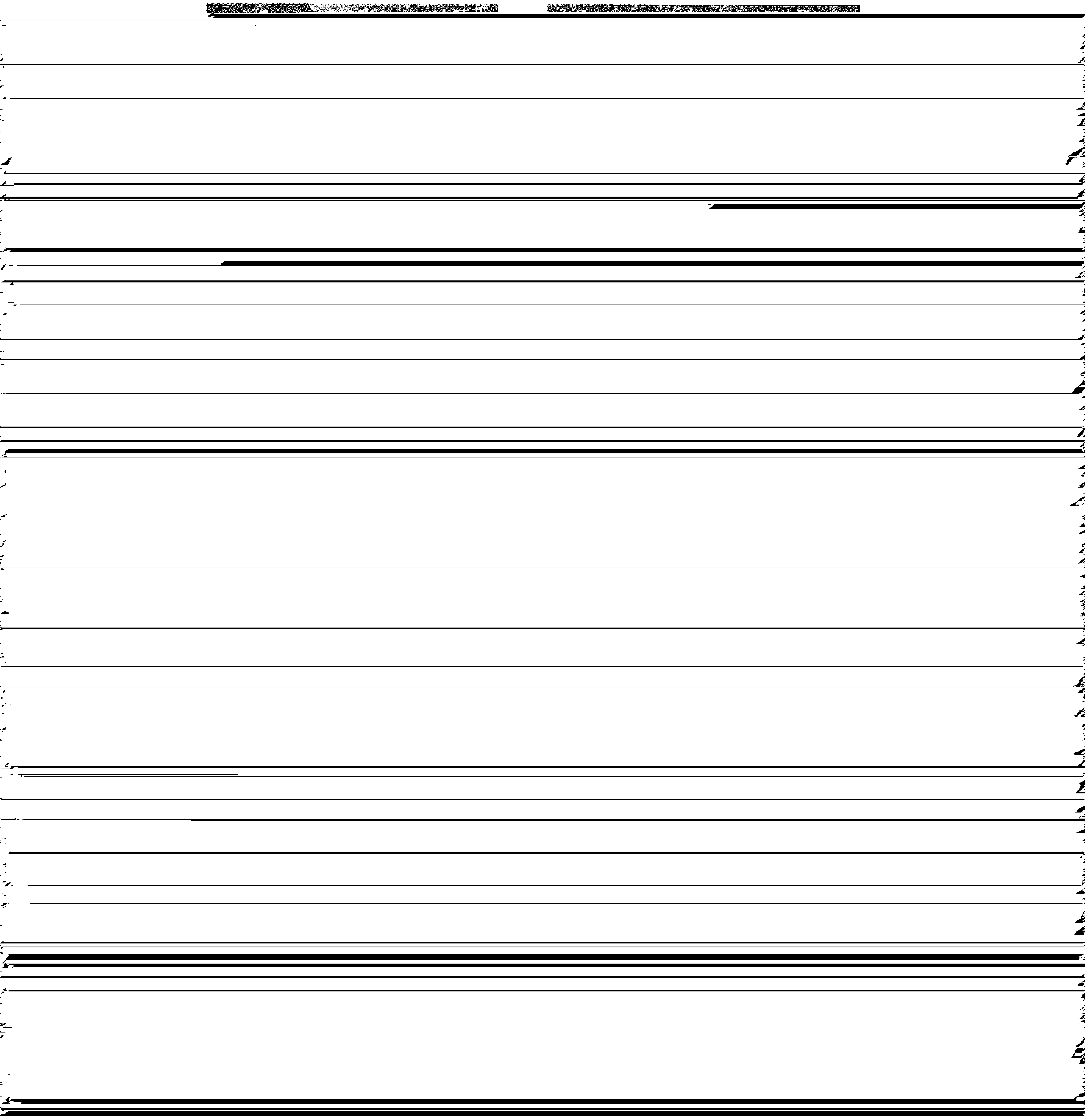


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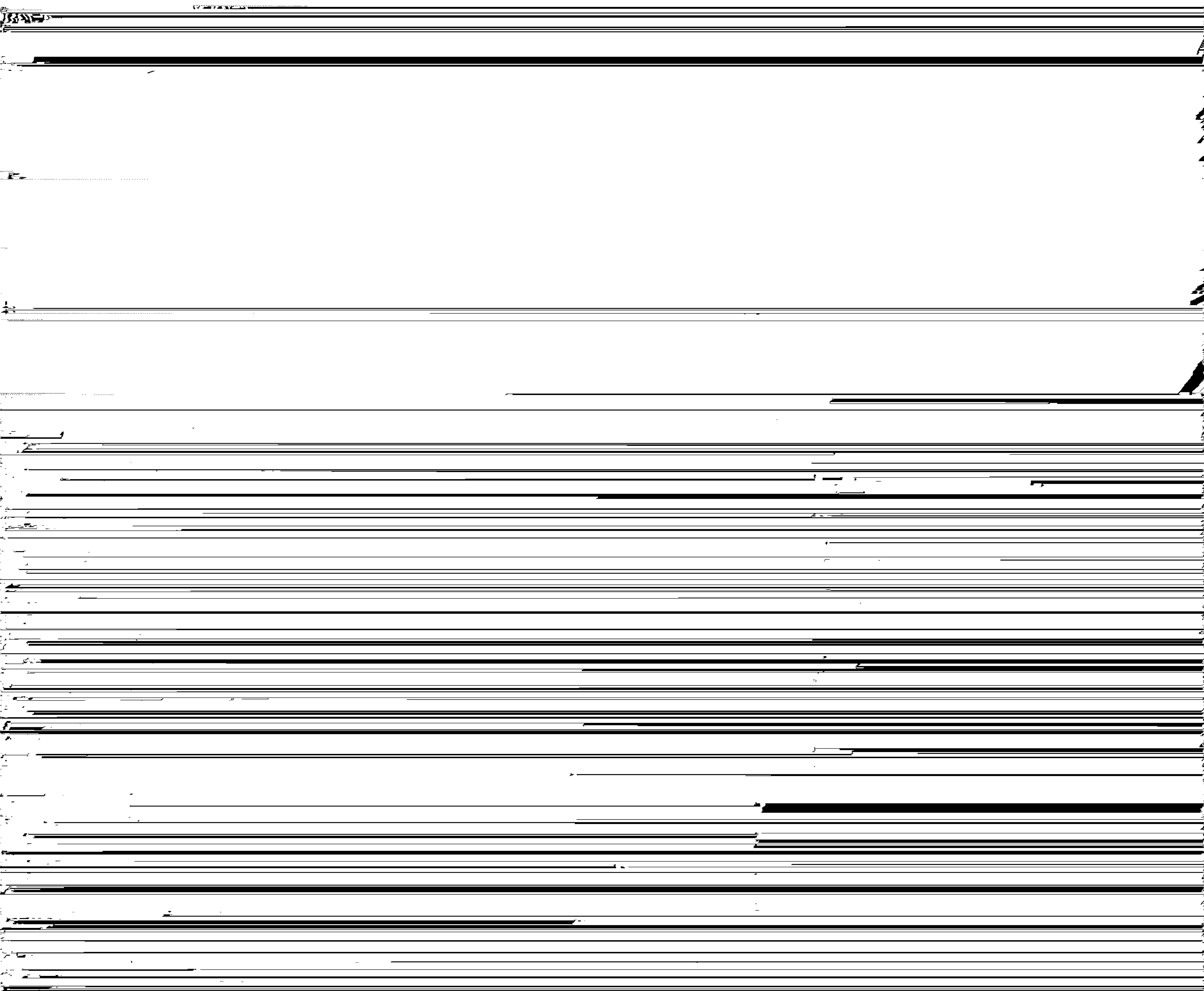
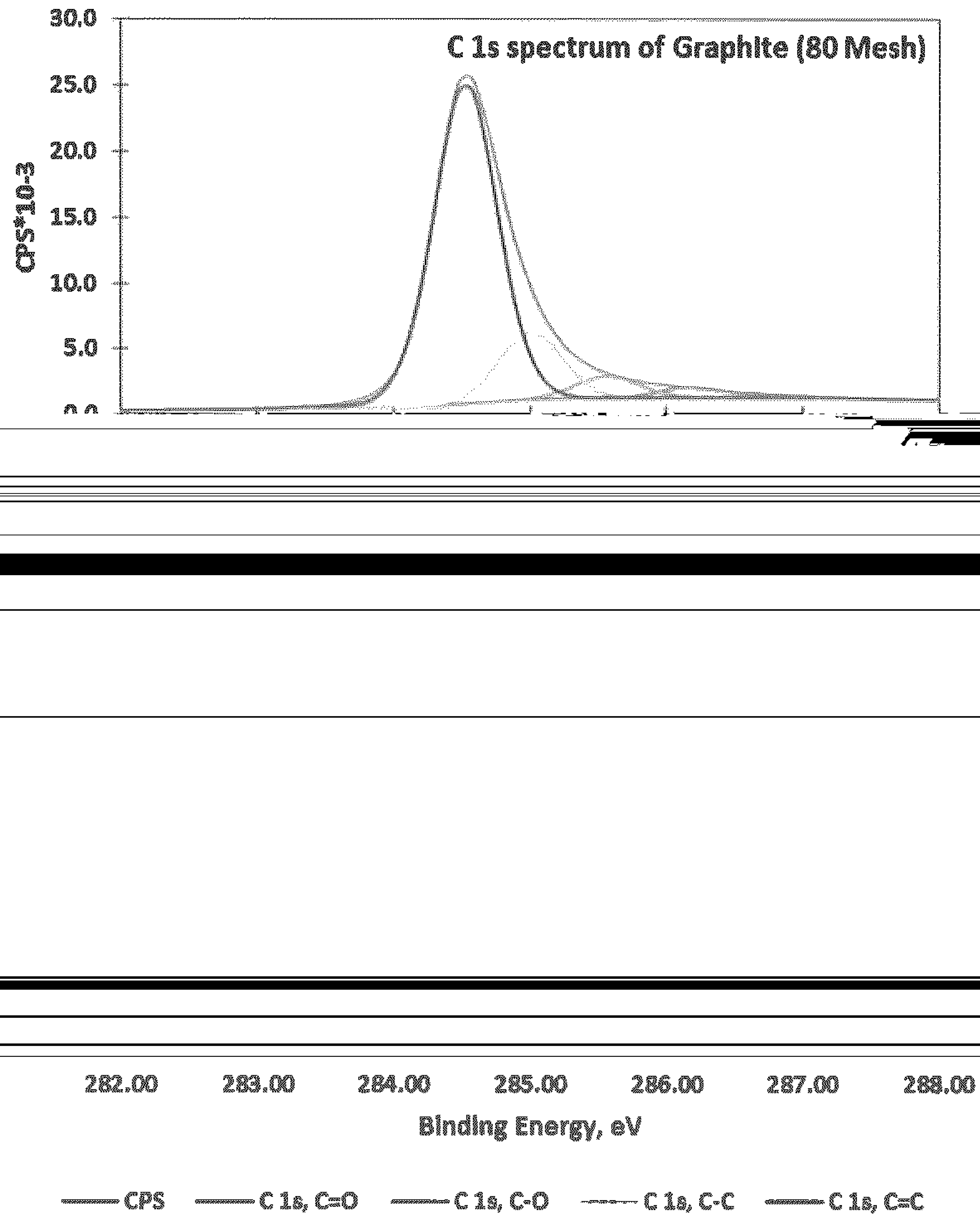
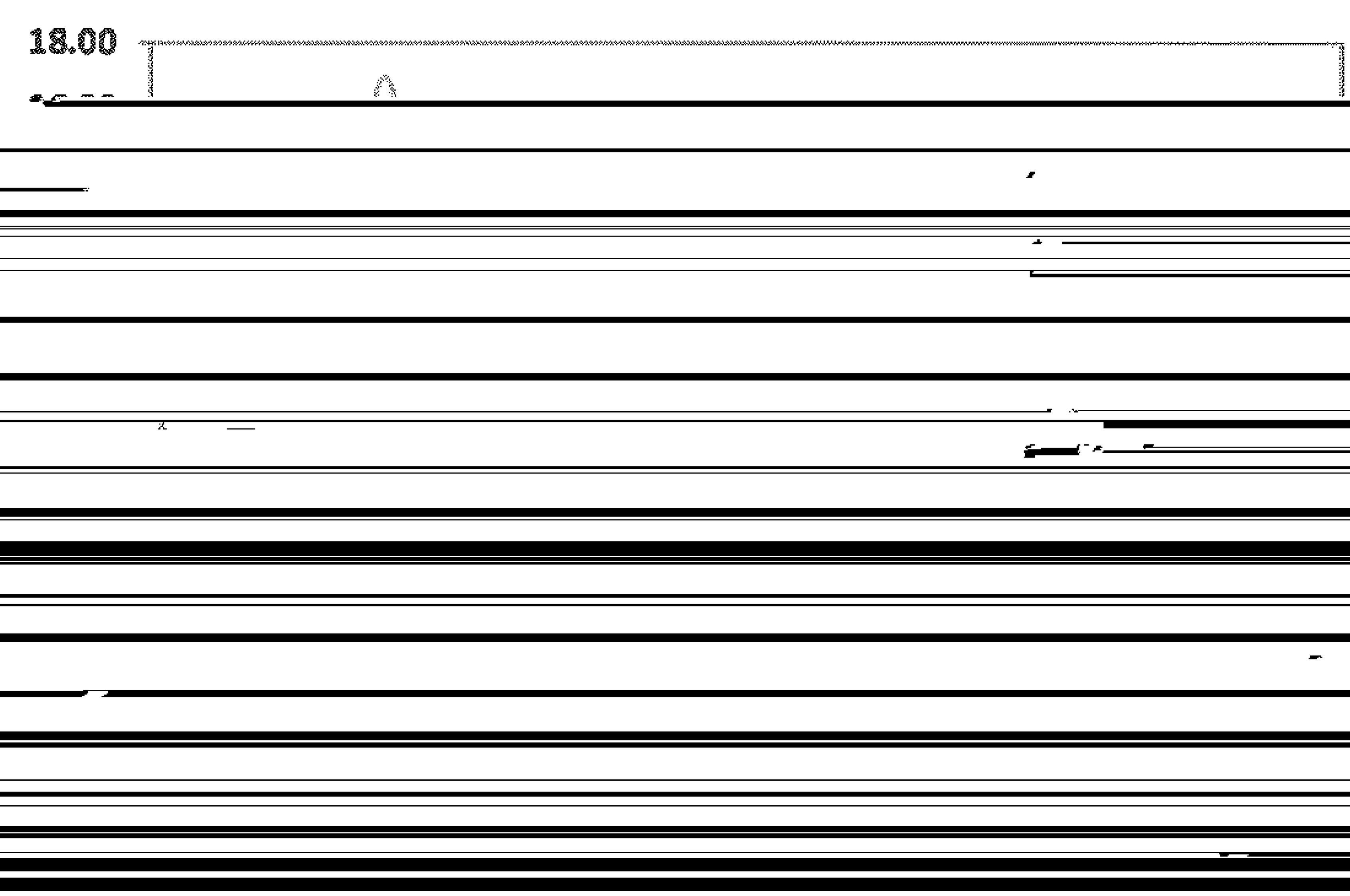




C 1s spectrum of Graphite (80 Mesh)
30.0
25.0
20.0
15.0
10.0
5.0
CPS*10-3
282.00
283.00
284.00
285.00
286.00
287.00
288.00
Binding Energy, eV
CPS
C 1s, C=O
C 1s, C-O
C 1s, C-C
C 1s, C=C

Fig. 8



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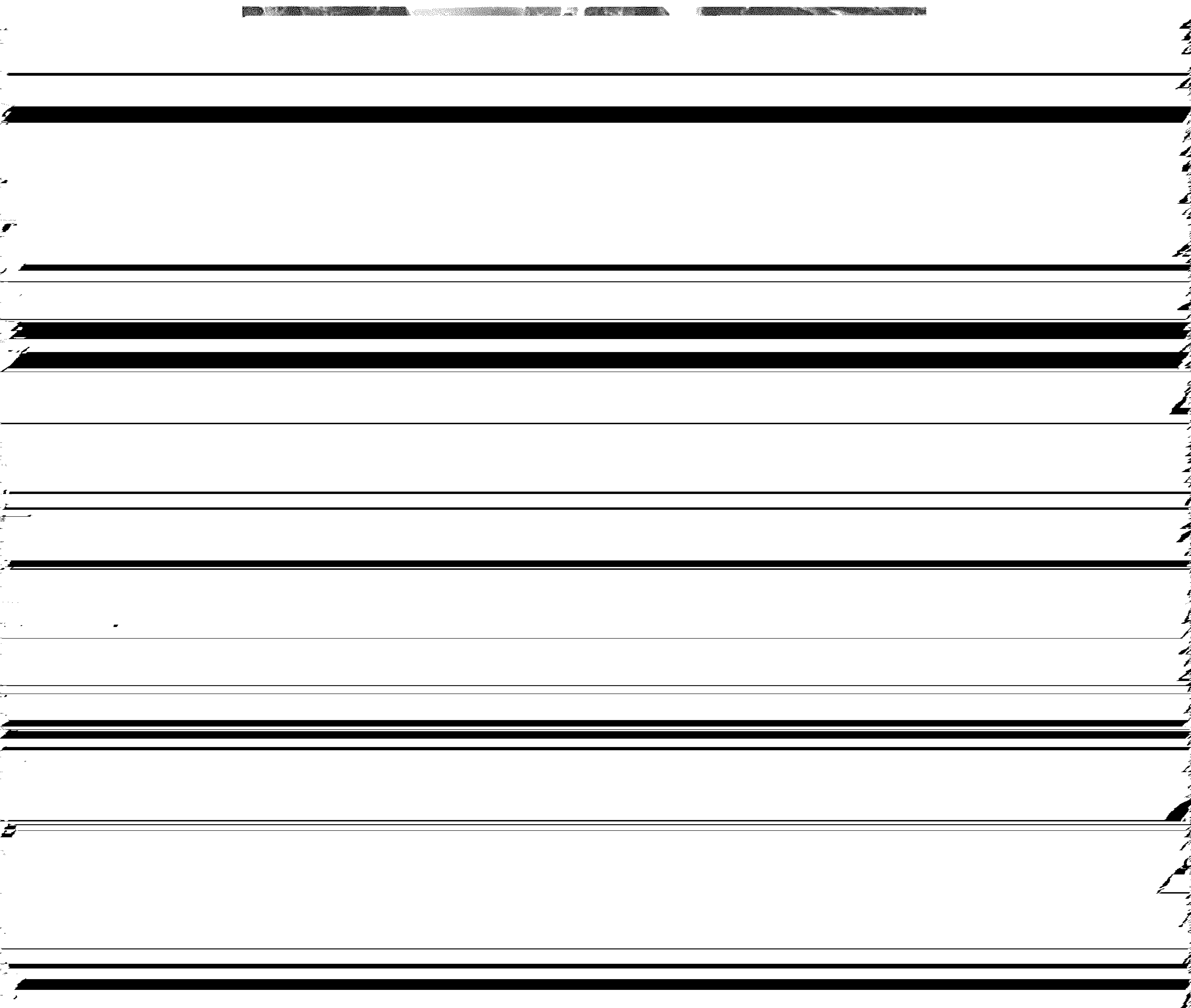

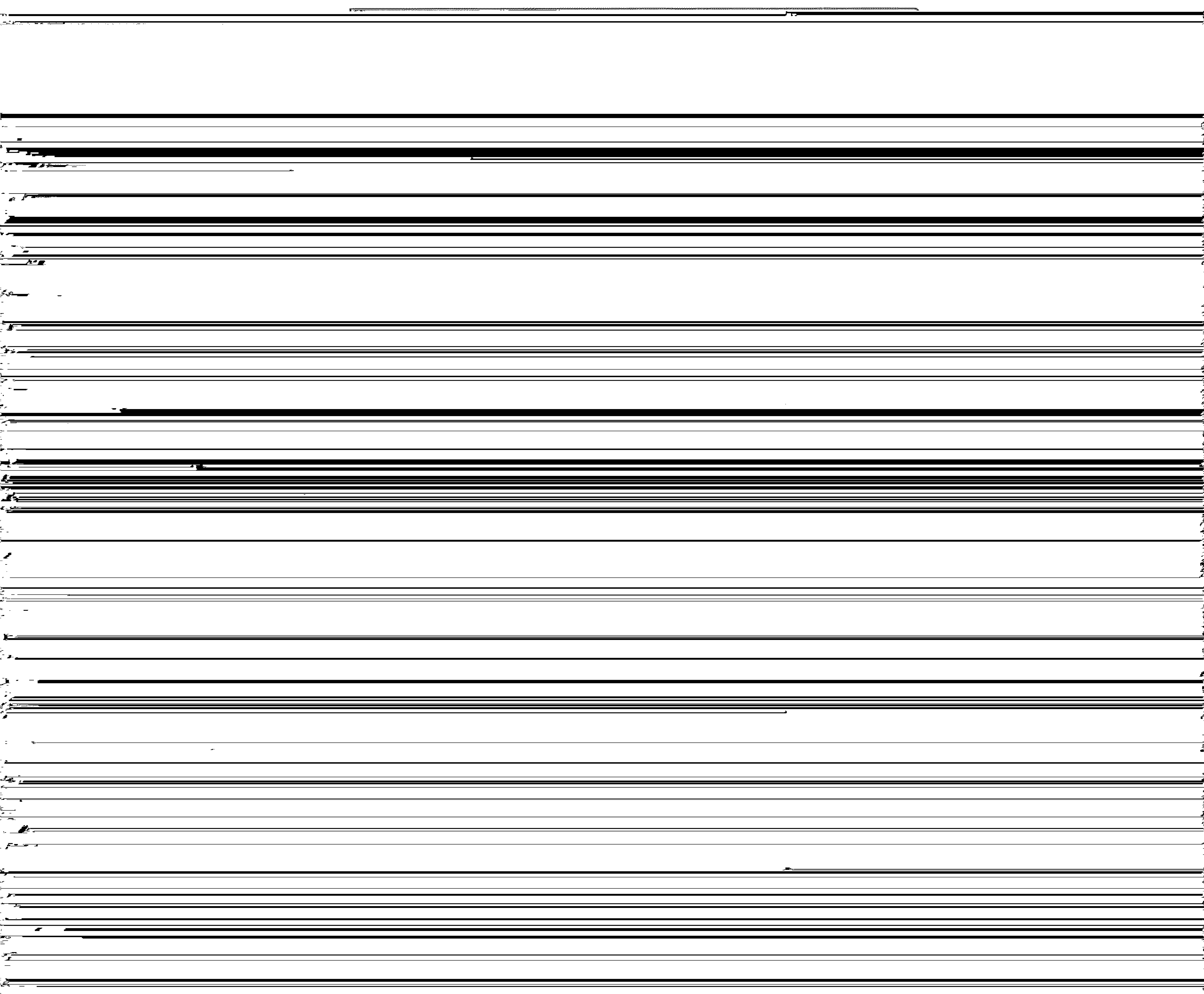

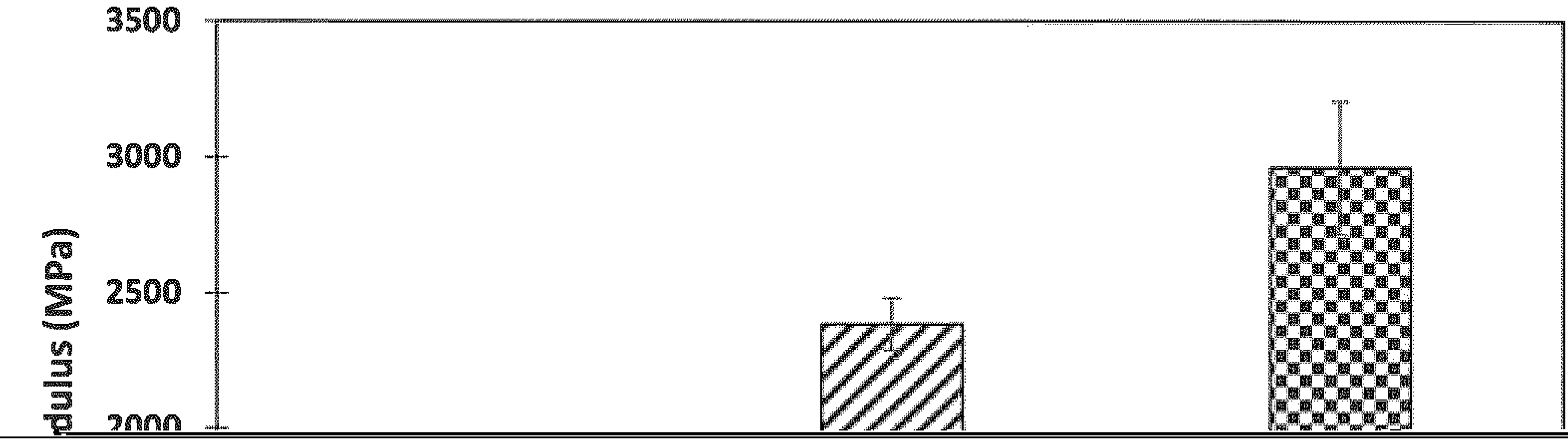
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dulus (MPa)

GRAPHENE NANOPLATELETS DERIVED FROM THERMOMECHANICAL EXFOLIATION OF GRAPHITE

FIELD

[redacted]

The invention relates to graphene nanoplatelets and meth- [redacted]

and obtaining the exfoliated product. In one embodiment, the layered material is graphite or expanded graphite and the exfoliated product is graphene and/or graphene nanoplatelets. In one embodiment, the method further includes grinding the layered material prior to shearing. In one embodi- [redacted]

shearing, and optionally heating the exfoliated product to [redacted] one embodiment, the at least one controller controls a temperature of the expandable chamber according to a selected temperature or a selected temperature range.

In one aspect, the invention provides graphene and/or graphene nanoplatelets prepared by the method of the above aspect. In one embodiment, the nanoplatelets are substantially unfunctionalized. In one embodiment, the nanoplatelets are substantially free of defects.

In one aspect, the invention provides a product comprising the graphene and/or the graphene nanoplatelets of the above aspect. In one aspect, the invention provides ink

graphene (FLG), multilayer graphene (MLG), or a combination thereof. In one embodiment, the nanoplatelets are substantially free of functionalities. In one embodiment of this aspect, the nanoplatelets are substantially free of defects. In one embodiment of this aspect, the method further comprises exposing the platelets to one or more compatibilizing agent(s) to form coated, or functionalized nanoplatelets. In one embodiment of the above method aspect, the compatibilizing agent comprises an anhydride or an amine. In one embodiment of the above method aspect, the anhydride is trimellitic anhydride. In one embodiment of

FIGS. 1A and 1B show plots of torque vs exfoliation time, which indicate rising torque (therefore decreasing bulk [illegible]

[illegible]

and Temperature=200° C.; in FIG. 1A the starting sample [illegible] weighed 80 g, in FIG. 1B the starting sample weighed 100 g.

FIG. 2 shows representative images of exfoliated graphite samples that all started as samples of the same weight, showing the decrease in bulk density at different processing times.

FIG. 3A shows a TEM image of sheet of few layer graphene obtained by the method described herein.

FIG. 3B shows an SEM image of initial graphite.

As used herein, the term “NMP” refers to N-Methyl-2-pyrrolidone.

[illegible]

Electron Microscopy

[illegible]

As used herein, the term “SSA” refers to specific surface area.

As used herein, the term “TEM” refers to transmission electron microscopy.

As used herein, the term “TGA” refers to thermogravimetric analysis.

As used herein, the term “TME” refers to a thermomechanical exfoliation technique that is described herein, in one embodiment, the TME method is used to functionalize

[redacted]

(e.g., drum) of a melt compounding machine, heating the material to a temperature in a range of about 150 to about 250° C., processing the material until a targeted increase in volume of material is observed, and obtaining exfoliated product.

In one embodiment, this method is performed in the absence of liquid medium, in another embodiment, it is performed in the presence of liquid medium. In one embodiment, this thermomechanical method is performed in the

[redacted]

able material into the machine, a first chamber having a working volume that is suitable for holding and preparing exfoliated material, one or more shearing element located within the chamber (e.g., two or more rotors that rotate, a screw extruduer) in such a manner as to apply shear force to the material), a motor suitable for driving the shearing element, optionally a controller for controlling the rotation of the rotors, a heating element for heating the material. In one embodiment, the machine includes a rheostat for con-

[redacted]

caused by the in-plane optical vibration (degenerate zone center E2g mode) and a D peak located at around 1340 cm⁻

[illegible]

1, associated with the first-order zone boundary phonons. Increased intensity of D peaks in GNP is indicative of defects. Since there is no broadening of G peak in GNP, then the defects are edge defects resulting from lateral size reduction of graphite flakes after exfoliation. Notably, the symmetric 2D Raman band at 2600-2800 cm^{-1} is charac-

[illegible]

modified GNP, prepared using the TME method, (ii) a polypropylene composite of 28.5 wt % F-GNP prepared

[illegible]

using the CM method, and (iii) neat polypropylene (i.e., polypropylene having 2% PP-g-MA). The flexural modulus of the F-GNP composite showed an improvement of nearly 26% relative to the composite from the CM process. The flexural modulus of the F-GNP composite showed an improvement of nearly 350% relative to neat PP. The impact

[illegible]

contract) the capacity (i.e., volume) of the chamber. For

exerting pressure on a release mechanism such that when pressure is exerted, the volume of the chamber is increased

Raman studies were performed using a Jobin-Yvon/

equipped with a 632 nm He/Ne laser source, 1800 1/nm grating and an Olympus BX41 microscope system. The laser

[redacted]

physical testing were compression molded at 180° C. for 2

[redacted]

minutes in a Carver press. Flexural tests were performed on an Instron 3369 Universal tester, at a cross head speed of 1.3

[redacted]

D790 standard.

Mechanical properties of test composites, which were composites of PP with 28.5 wt % anhydride-modified F-GNP (prepared via the TME and CM methods), were compared with the control, which was neat PP). The flexural modulus of the composite with F-GNP (from the TME

[redacted]

Mechanical properties and electrical conductivity of PA composites

Sample	Electrical [redacted] (S/m)	Flexural [redacted] (MPa)	Impact [redacted] (J/m)
PA	5.00E−14	1854 ± 19	34 ± 4.2
PA/28.5 wt. % GNP	0.94	5717 ± 175	27 ± 1.5
PA/28.5 wt. % F-GNP	21.7	5084 ± 102	27 ± 1.5

12. The machine of claim 10, further comprising a pressure sensor that senses pressure within the expandable chamber and outputs a sensor signal, wherein the at least one controller uses the sensor signal to control an operation of the machine.

13. The machine of claim 12, wherein the at least one controller uses the sensor signal to modulate a volume of the expandable chamber, wherein the volume of the expandable chamber increases as the pressure increases.

14. The machine of claim 10, wherein the at least one shearing element comprises at least two rotors.

15. The machine of claim 10, wherein the at least one controller controls an operating speed of the shearing element in a range of about 50 to about 150 rpm.

16. The machine of claim 10, wherein the at least one controller controls an operating speed of the shearing element at about 100 rpm.

17. The machine of claim 10, wherein the at least one controller controls a temperature of the expandable chamber according to a selected temperature or a selected temperature range.

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