

# Graphene-Inspired Films: Capturing Nanoplastics and Blocking Their Path to Crops

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## Abstract

In 2020, Italian researchers confirmed the presence of nanoplastics (NPs), smaller than microplastics (MPs), in apples and carrots, raising concerns about the accumulation of NPs in the human body through crop consumption. Given that many MPs carry electrical charges, this study aimed to develop an innovative solution to prevent the penetration of NPs into crops using electrical energy. To confirm whether NPs of styrene acrylonitrile resin used in this study undergo electrical migration, they were exposed to an electric field using copper plates, lead wires, or graphite rods as electrodes. NPs migrated toward the anode (+), and the copper plates and lead wires were transformed after supplying electricity, but not the graphite rods. Since graphite rods have a small surface area and vulnerability to physical damage, graphite powder was coated onto cellophane tape or agar films (CTcGP or AFcGP, respectively) to mimic the graphene structure. At 9 V, graphene-mimic (graphite-based) films effectively captured NPs present in soil and reduced the penetration of NPs into crop roots by more than 88%. Therefore, incorporating solar panels for power supply could enable environmentally friendly and cost-effective applications. This research demonstrates the practicality of an electrochemical solution to address agricultural and crop contamination caused by NPs.

## Keywords

Materials Science, Nanomaterials, Graphene-Mimic Films, Nanoplastics, Agricultural Contamination

## 1. Introduction

Microplastics (MPs) refer to all types of plastic fragments smaller than 5 mm, and they are classified into two types by their origin. Primary MPs are plastic frag-

ments purposefully manufactured to be 5.0 mm or smaller, and they have been used in various common products, such as cosmetics, clothing, food packaging, and industrial processes [1] [2]. A large number of primary MPs enter ecosystems, causing pollution [3]. Secondary MPs include small plastic fragments generated from the wear and tear of nearly all plastic products humans use, such as water bottles, fishing nets, tires, and tea bags [1]. Plastic discarded into nature is broken down into small fragments by physical, biological, or chemical forces of nature, and depending on the degree of breakdown, it can be reduced to an undetectable size [4]. Nanoplastics (NPs) are plastic fragments less than 1  $\mu\text{m}$  in size [5]. Since NPs are very small, they are believed to be able to affect cellular function by crossing cellular membranes [6].

MPs or NPs take a long time to decompose, leading to their accumulation in ecosystems [6]. Since non-degradable chemicals released into nature can bioaccumulate within organisms through the food chain, MPs or NPs are highly likely to be ingested, absorbed, and accumulated in the bodies of many organisms [7] [8]. Therefore, various research is ongoing to determine how MPs or NPs accumulated in the body affect living organisms. For example, the filtration ability and immune function of bivalves were reduced by exposure to microplastics [9] [10]. This indicates that MPs or NPs accumulated in the body impair the ability to detoxify harmful substances by damaging DNA [11]. Moreover, MPs increased the rate of developmental arrest and developmental malformations and decreased the rates of fertilization [12] [13].

Recently, NPs have been detected in various crops, such as apples and carrots in Italy, making the threat of NPs more prominent [14]. It stems from the extensive use of plastics in modern farming, a prime example being plastic films used to prevent moisture evaporation from the soil. However, the problem arises afterward. Most used plastic films are not completely removed.

When the films are exposed to physical impacts such as water, wind, or sunlight (ultraviolet) for a long period of time, the interaction between film particles weakens and they break down into smaller particles, ultimately into NPs [15]. The toxicity of NPs to organisms depends on their size or surface charge [16]. Since NPs are accumulated in the body when these crops are consumed, their detection in crops is very dangerous [17]. It is known that plastic fragments are often charged or become charged by interacting with natural organic materials [7]. This information suggests that using electricity can inhibit the entry of charged NPs into crops.

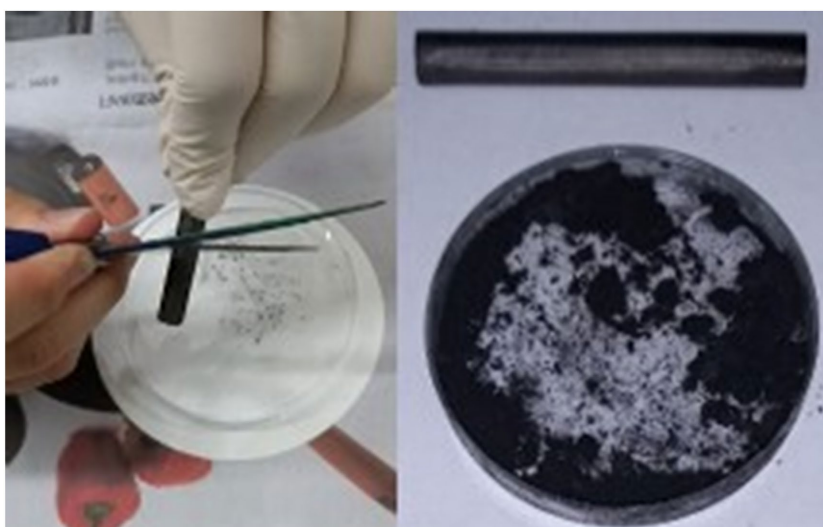
Graphite, a crystalline allotrope of carbon atoms, has high conductivity of heat and electricity. Graphite is consumed in many critical industries such as refractories, lithium-ion batteries, foundries, and lubricants [18]. Graphene was properly isolated and characterized in 2004 by pulling graphene layers from graphite with a common adhesive tape in a process called micro-mechanical cleavage [19]. Through various follow-up research on the properties and commercialization of graphene, it was revealed that graphene has exceptionally high tensile strength,

electrical conductivity, and transparency [20], and it is used in semiconductors, electronics, electric batteries, and composites [21]. Moreover, research is also being conducted continuously on how to utilize it as a material for quantum mechanics, electricity, chemistry, mechanics, optics, and magnetism.

The purpose of this study is to develop an innovative solution that prevents the penetration of NPs into crops using graphene-mimic films. To this end, it was determined whether NPs are charged, whether a graphene-mimic film composed of graphite powder is conductive, and whether the electrically charged graphene-mimic film captures NPs and suppresses the penetration of NPs into plant tissue.

## 2. Methods

### *Preparation of CTcGP or AFcGP*

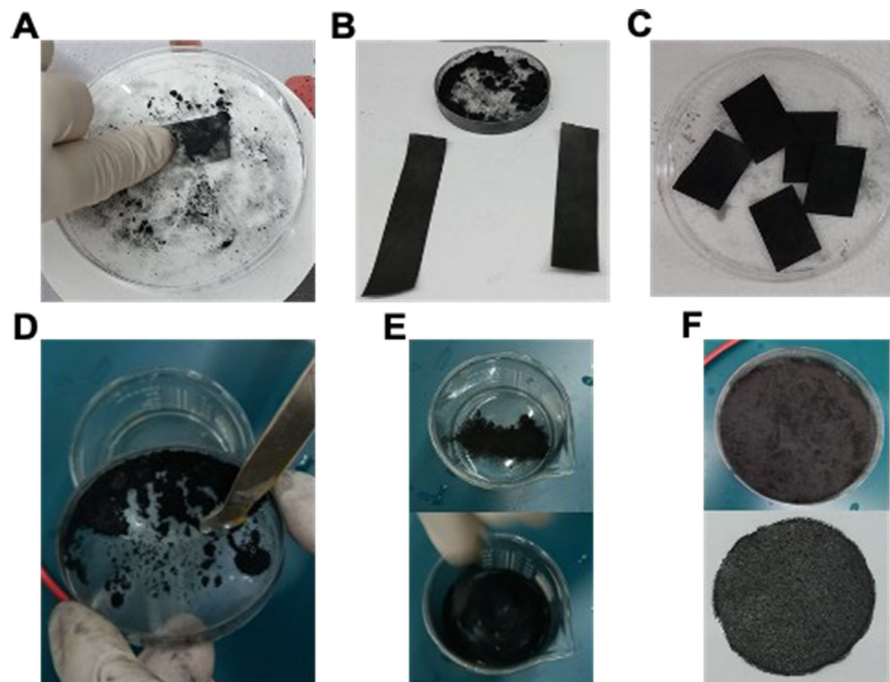


**Figure 1.** Preparation of graphite powder from graphite rods.

To prepare cellophane tape coated with graphite powder (CTcGP) or agarose film coated with graphite powder (AFcGP), graphite powder or graphite rods were purchased from DUKSAN GENERAL SCIENCE or an online shopping mall (COUPANG). Graphite rods were scraped with scissors to obtain graphite powder, and the collected graphite powder was gathered in a Petri dish (Figure 1).

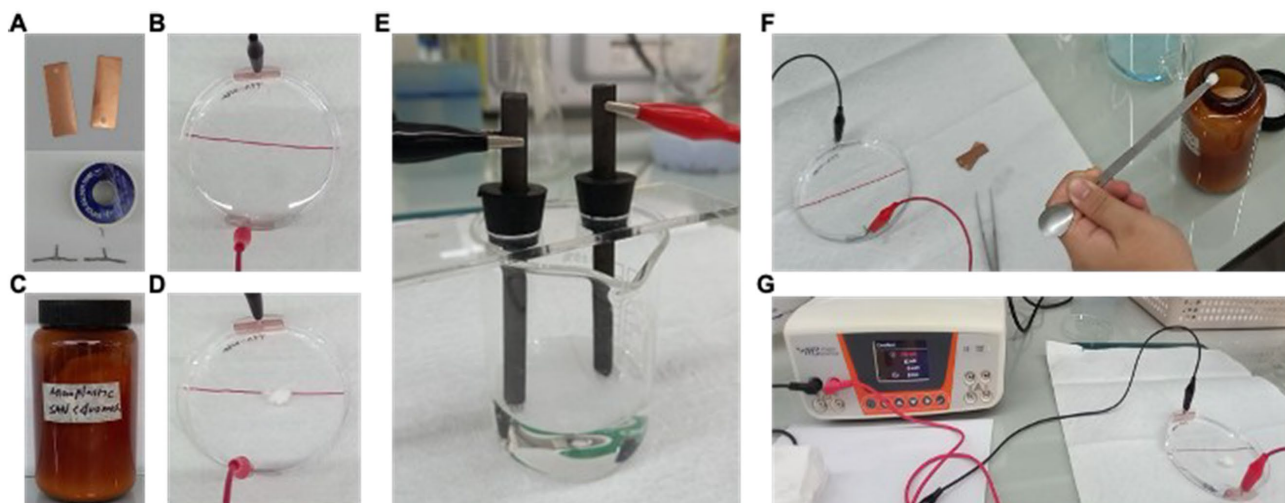
For preparing CTcGP, cellophane tape (CT) was used. The CT was cut to approximately 8 cm in length, and then the prepared graphite powder (GP) was sprinkled onto the surface of CT. The CT, with all surfaces covered in GP, was cut to approximately 2 cm in length (Figures 2(A)-(C)).

For preparing AFcGP, agarose gel was used. A total of 0.8 g of agarose powder was completely dissolved in 100 mL of 1× Tris-acetate-EDTA (TAE) buffer using a microwave. GP was added to the melted agarose solution, mixed thoroughly with a spoon, then poured into a Petri dish and dried at room temperature for 4 days (Figures 2(D)-(F)). The fully dried agarose film was then removed from the Petri dish.



**Figure 2.** Preparation of CTcGP or AFcGP ((A), Coating of graphite powder on cellophane tape (CTcGP); (B), Prepared CTcGP; (C), Cut CTcGP; (D), Addition of graphite powder to agarose solution; (E), mixing of graphite powder and agarose solution; (F), Mixture of agarose gel and graphite powder poured into a petri dish, and drying the mixture).

### *Migration of NPs by electricity*



**Figure 3.** Preparing the Device to Observe the NPs Migration ((A), copper panels and lead wire; (B), connecting the copper panels to a Petri dish filled with TAE buffer; (C), SAN NPs; (D), NPs on the 1× TAE; (E), Connecting the graphite rod to the beaker filled with TAE buffer; (F), Connecting the lead wire to a Petri dish; G, electricity supply).

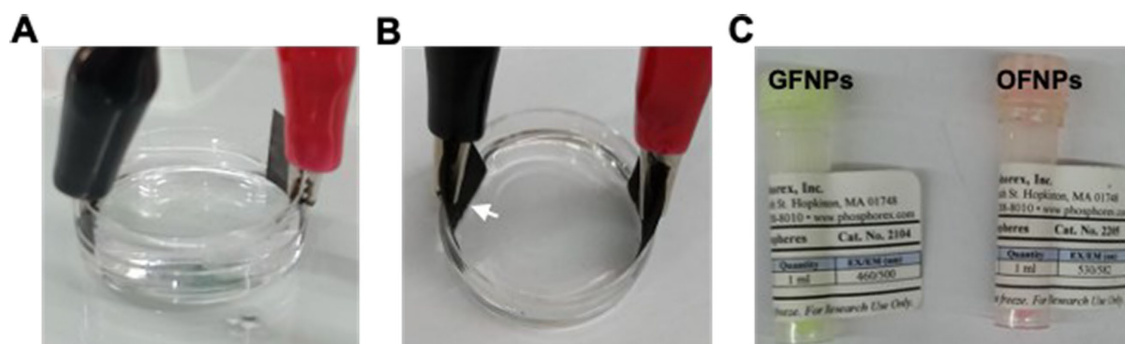
The copper plates or lead wires were secured at both ends of a Petri dish using alligator clip wires (Figures 3(A)-(B)). Ten milliliters of 1× TAE was added to the Petri dish until it was just below the level of the clips. Styrene acrylonitrile resin

(SAN) powder was placed in the center of the TAE buffer. According to the manufacturer's specifications and previous literature, the SAN resin powder used in this study has a particle size distribution predominantly below 1  $\mu\text{m}$ , which operationally classifies it as nanoplastics (NPs) (Figures 3(C)-(D)).

A total of 50 mL of 1 $\times$  TAE buffer was added to a 250 mL beaker, then two graphite rods connected with alligator clip wires were placed in the beaker, with NPs positioned between the two rods (Figures 3(E)-(F)). Electricity at 100 V was applied through the alligator clip wires, and the movement of the NPs on the surface of the TAE buffer was observed (Figure 3(G)).

#### *Migration of fluorescent NPs (FNPs)*

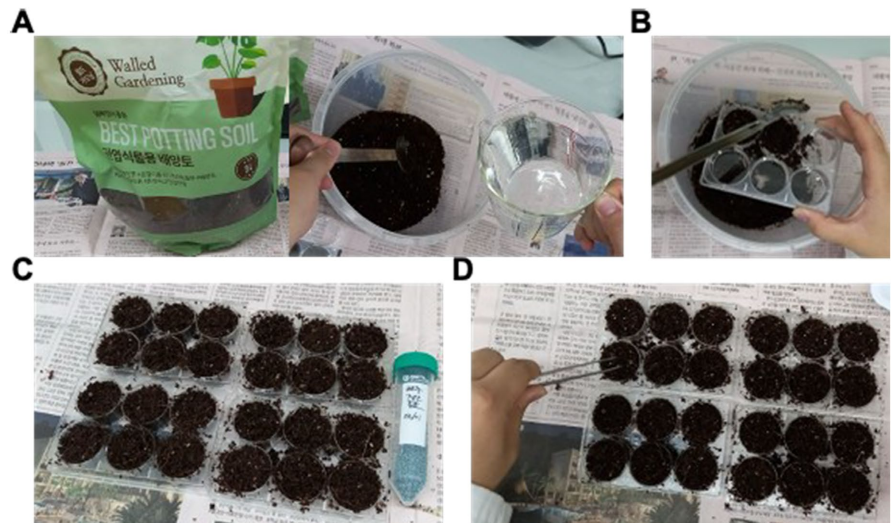
Two CTcGPs were connected to each end of a 35 mm diameter Petri dish using alligator clip wires (Figure 4(A)). After adding 3 mL of 1 $\times$  TAE buffer and applying 100 V of electricity, the electrical conductivity of the CTcGP was confirmed by the formation of bubbles at the positive electrode (white arrow in Figure 4(B)). Green or orange fluorescent-labeled nanoparticles (GFNPs or OFNPs, respectively) were diluted with distilled water to concentrations of 1/100 or 1/1,000 (Figure 4(C)). In the center of the Petri dish, 10  $\mu\text{L}$  of the 1/1,000-diluted GFNPs and OFNPs was added, and 100 V of electricity was applied. After 15 minutes, the power was turned off, the CTcGPs at both ends were removed, and the movement of NPs to the CTcGP surface was observed using a fluorescence microscope.



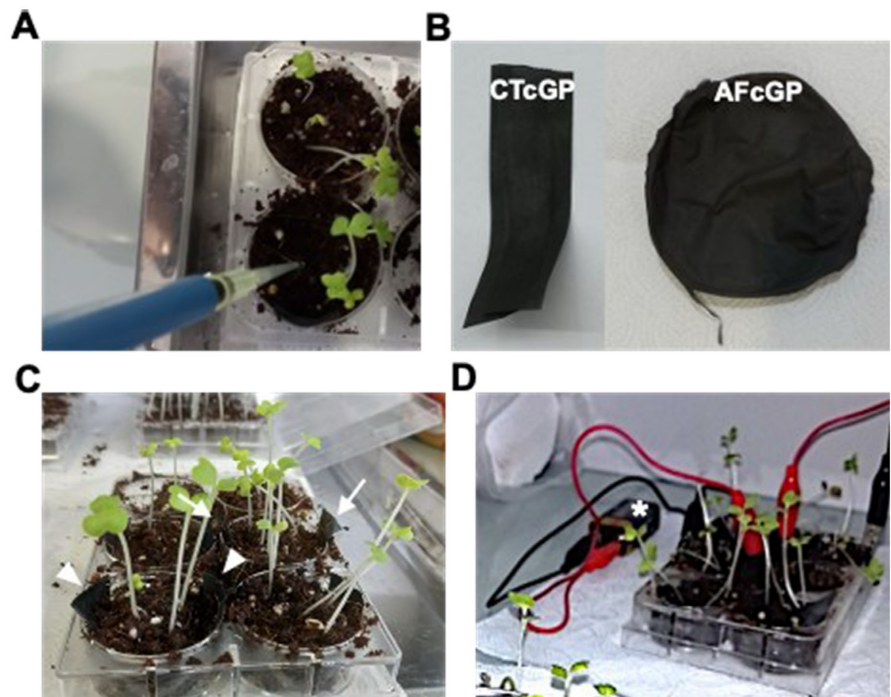
**Figure 4.** Migration test for GFNPs or OFNPs ((A), Connecting the CTcGP to a Petri dish filled with TAE buffer; (B), Bubbles generated in CTcGP connected to the (-) pole (white arrow) after power supply; (C), GFNPs or OFNPs used in this study).

#### *Planting cabbage seeds on NPs-contaminated soil*

Commercial potting soil (Walled Gardening, Organic Potting Mix with Perlite; composed primarily of peat moss, perlite, and organic compost) was purchased from an online shopping mall (COUPANG) and used consistently across all experimental groups to ensure reproducibility. The soil was transferred to a 5 L basket, and water was added to mix the soil and water thoroughly (Figure 5(A)). The moistened soil was then placed in each well of a 6-well plate (Figure 5(B)). Five cabbage seeds were placed in each well and incubated at room temperature (Figure 5(C), Figure 5(D)). A 100 V mixture of GFNPs and OFNPs diluted to 1/1,000 was applied to each seedling (Figure 6(A)).



**Figure 5.** Seeds Planting ((A), Preparation of potting soil; (B), Transfer the soil to each well of 6 well plate; (C), 6-well plates filled with soil and cabbage seeds; (D), Planting seeds).



**Figure 6.** Application of CTcGP or AFcGP on the NPs-contaminated soil (A, Add GFNPs to soil; B, Preparation of CTcGP or AFcGP; C, Application of CTcGP (white arrows) or AFcGP [ph57] [ph58] (white [ph59] [ph60] arrowheads) to soil; D, Power supply via 9V battery).

#### *Application of CTcGP or AFcGP on soil*

Prepared CTcGP or AFcGP pieces were cut to 1.5 cm by 3 cm and placed at each end of the well in the 6-well plate containing soil and growing cabbage seedlings (Figure 6(B), Figure 6(C)). These pieces were connected to the positive and negative terminals of a 9 V battery using alligator clip wires.

Electricity at 9 V was supplied through the alligator clip wires for 4 or 7 days. After removing CTcGP or AFcGP, the GFNPs or OFNPs on their surfaces were observed using a fluorescence microscope.

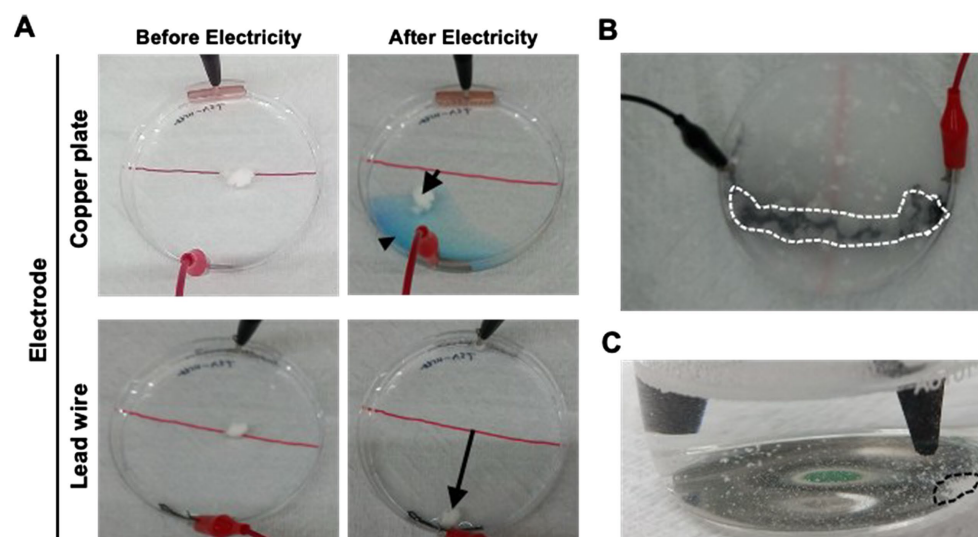
### 3. Results and Discussion

#### *Migration of NPs by Electricity*

It should be clarified that the graphene-mimic films used in this study function primarily as flexible graphite-based electrodes. They do not possess the quantum or electronic properties of monolayer graphene, but instead leverage the electrical conductivity and layered structure of graphite powder.

To prevent the penetration of NPs into plants using electricity, it was first determined whether NPs migrate in an electric field. For this purpose, copper plates, lead wires, or graphite rods were tested as electrodes.

When electricity was supplied to the copper plates, lead wires, or graphite rods, the NPs placed in the center of the Petri dish gradually moved toward the (+) electrode (**Figure 7(A)**, **Figure 7(C)**). This indicates that the NPs carry a negative charge.



**Figure 7.** Electric mobility of NPs by electrode composed of copper panel, lead wire, or graphite rod ((A), Migration of NPs by electricity; (B), Deformed lead wire after power supply; (C), NPs migrated to the (+) pole in a beaker using a graphite rod as an electrode).

Although direct DLS or SEM characterization was not performed in this study, the observed electrical migration behavior is consistent with that of previously reported nanoplastic-scale particles.

To determine the optimal electrode for the electric migration of NPs, the appearance of the copper plates, lead wires, or graphite rods was observed after the electric migration of NPs was completed. A blue solution was observed on the copper plate connected to the (+) electrode (**Figure 7(A)**). Negatively charged oxygen ions move toward the (+) electrode, and copper combined with oxygen ions

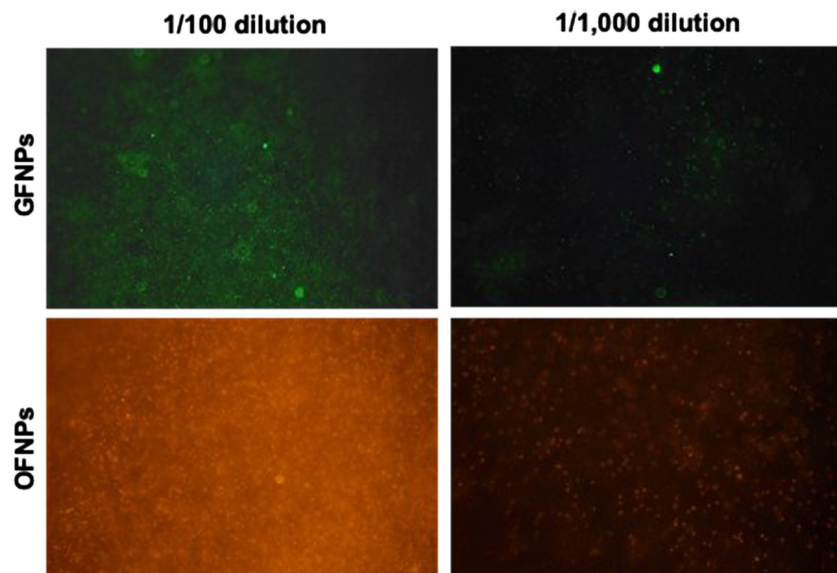
is oxidized into blue copper oxide [22]. Therefore, this indicates that the blue substance formed after the electric migration of NPs is copper oxide.

The lead electrode showed no change in the initial stage of electric supply; however, as the electric supply time increased, a black substance was generated from lead accumulated on both lead electrodes (Figure 7(A), Figure 7(B)). Considering that lead in water exhibits neurotoxicity [23], the use of lead as an electrode is deemed unsuitable. However, no changes were observed in the graphite rods used as electrodes, indicating that graphite is the optimal material for electrodes in the electric migration of NPs in soil.

#### ***Observation of Fluorescently Labeled NPs***

NPs are too small to be visible to the naked eye. Therefore, to visualize the NPs, green or orange fluorescent NPs (GFNPs or OFNPs) were used in this study. GFNPs or OFNPs were diluted to 1/100 or 1/1000, and then each diluted sample was observed under a fluorescence microscope.

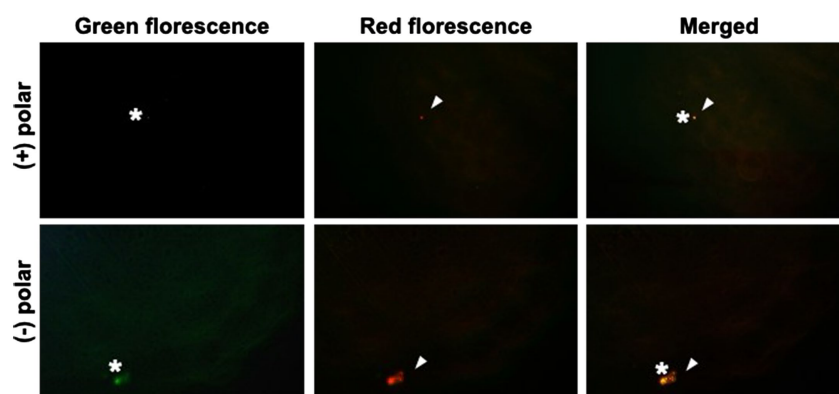
GFNPs or OFNPs diluted to 1/100 were shown as countless fluorescent particles, resulting in an overall bright green or orange image, respectively. Since FNPs diluted to 1/1000 showed fewer fluorescing particles than 1/100, the images showed a darker background and the FNPs were distinctly discernible. Therefore, it indicates that the particles of the FNPs can be detected under a fluorescence microscope (Figure 8).



**Figure 8.** Observation of GFNPs or OFNPs under a fluorescence microscope.

#### ***Migration of Fluorescent NPs Using CTcGP***

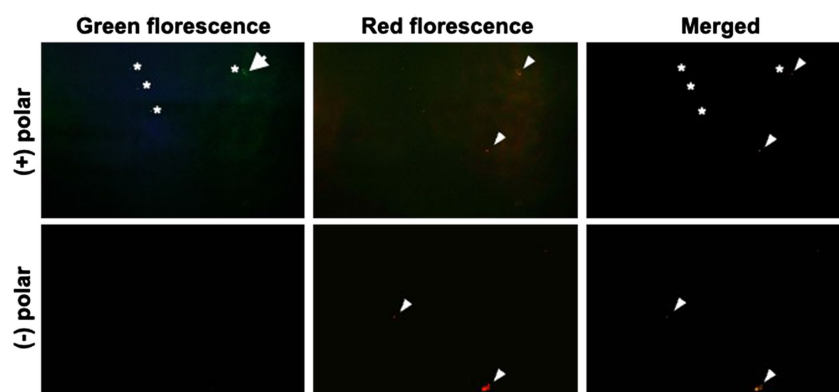
Although the graphite rod was determined to be the optimal electrode for the electric migration of NPs, the graphite rod does not have a large surface area, making it unsuitable for application in wide soil areas. Graphite consists of a structure where carbon atoms are arranged in thin layers stacked on top of each other, and a single layer of carbon atoms arranged in this manner is called graphene [21].



**Figure 9.** GFNPs or OFNPs on the surface of CTcGP before electrical supply.

Since graphite powder electrode has exceptional rigidity and electrical conductivity, a graphene-mimic structure was selected for the electrical mobility of NPs. However, because pure graphene was not easily obtainable, a graphene-mimic film was prepared by applying graphite powder onto cellophane tape (CTcGP). To determine whether CTcGP with an applied electric current can electrically migrate NPs, GFNPs and OFNPs were subjected to electromobility.

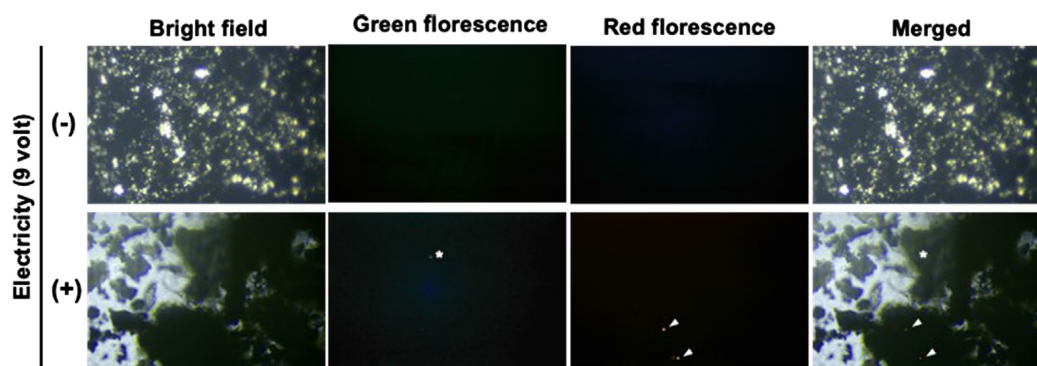
CTcGP was connected with alligator clip wires to a power supply in a Petri dish filled with TAE buffer, and a GFNPs and OFNPs mixture was dropped in the center of the dish. The presence of FNPs on the surface of CTcGP were confirmed using a fluorescence microscope. Before the power was supplied, FNPs were rarely detected on the surface of the CTcGP connected to either the (-) or (+) terminal (**Figure 9**). After supplying electricity for 10 minutes, four GFNP signals and two OFNP signals were detected on the surface of CTcGP connected to the (-) terminal, and only two OFNP signals were detected on the surface of CTcGP connected to the (+) terminal (**Figure 10**).



**Figure 10.** GFNPs or OFNPs on the surface of CTcGP after electrical supply.

#### *Capture of NPs in Soil by CTcGP or AFcGP*

To verify whether NPs present in soil are captured by CTcGP or AFcGP under an electric current, a 9 V electric current was applied to CTcGP or AFcGP electrodes inserted into soil containing a mixture of GFNPs and OFNPs.



**Figure 11.** Capture of GFNPs or OFNPs on soil by CTcGP electrodes.

After 4 days, the CTcGP or AFcGP was removed from the soil and their surfaces were observed under a fluorescence microscope. No FNPs were detected on the CTcGP surface connected to the (-) electrode, but one GFNP and two OFNPs were detected on the CTcGP surface connected to the (+) electrode (Figure 11). The result, where a small number of GFNPs and OFNPs were detected on the CTcGP surface connected to the (+) electrode, may be due to the easy detachment of GP from the cellophane tape surface, resulting in minimal retained NPs.

In contrast, when AFcGP was used as the electrode, no GFNPs were detected on either the (-) or (+) electrodes, but four and six OFNPs were detected on the AFcGP surfaces connected to the (-) and (+) electrodes, respectively (Figure 12). These results suggest that continuous electric current applied to CTcGP or AFcGP in the soil allows for the capture of polar NPs present in the soil.



**Figure 12.** Capture of GFNPs or OFNPs on soil by AFcGP electrodes.

### *Inhibition of NPs Penetration into Crop Roots*

Recent studies indicate that crops grown in soil contaminated with NPs have shown detectable levels of NPs [15] [17] [24]. Therefore, it was examined whether the penetration of NPs from soil to crops is inhibited by current-supplied CTcGP or AFcGP. For this purpose, cabbage seeds were germinated in soil contaminated with GFNPs and OFNPs, and an electric current was applied to CTcGP or AFcGP inserted in the soil. After 7 days, each seedling was removed from the soil, and the roots were washed with water. The presence of GFNPs or OFNPs within the roots

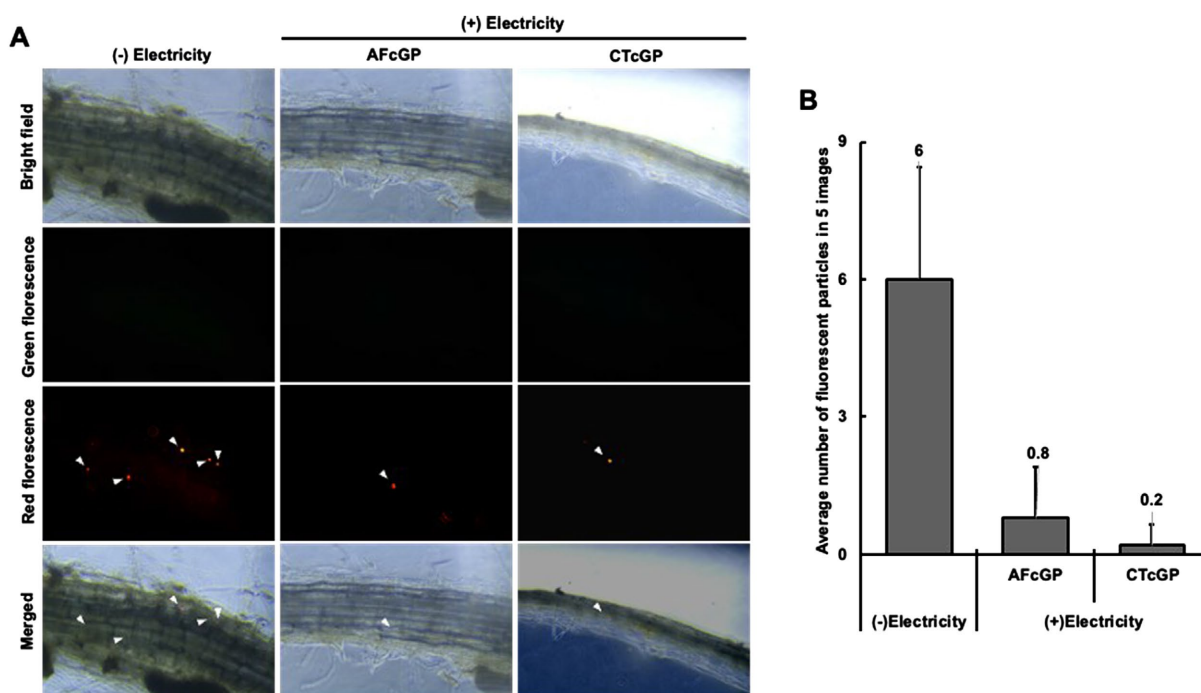
was then determined by observation under a fluorescence microscope.

In the roots of cabbage seedlings grown in soil without an electric current, no GFNPs were detected, but OFNP signals were detected (Figure 13). This suggests that NPs in soil penetrate crops through their roots.

For seedlings grown in soil with an electric current applied through AFcGP or CTcGP, no GFNP signals were observed in the roots. However, OFNP signals were rarely observed in the roots. To quantitatively assess their detection, the average number of detected OFNPs was calculated from five images. On average, six OFNPs were detected in the roots of seedlings grown in soil without an electric current, while an average of 0.8 OFNPs was observed in seedlings grown in soil with an electric current applied via AFcGP, and 0.2 OFNPs in those grown with an electric current applied via CTcGP. The result indicates that an electric current via AFcGP or CTcGP significantly reduces the penetration of NPs into crop roots (Figure 13).

Continuous application of a 9 V electric current in soil may induce electrochemical side effects, including local pH changes or electrolysis-related byproducts. These factors could potentially influence plant health independent of nanoplastic uptake. However, no visible differences in seed germination or root growth were observed during the experimental period. Future studies should quantitatively assess soil pH and plant physiological responses to isolate these effects.

Statistical analysis was performed using a two-tailed Student's t-test to compare the number of detected OFNPs between the control and electrically treated groups. The reduction in nanoplastic penetration was statistically significant ( $p < 0.05$ ).



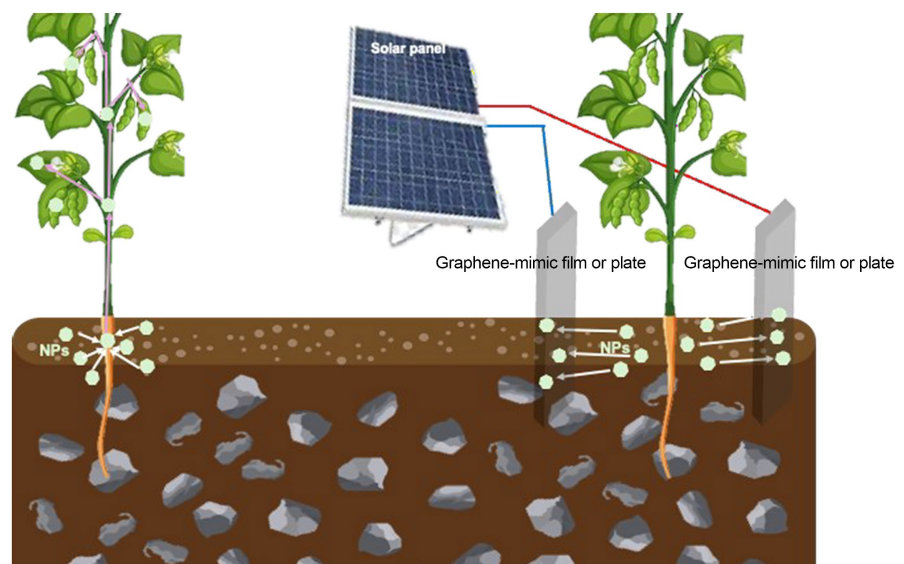
**Figure 13.** FNPs in roots of cabbage grown in FNPs-contaminated soil (A, FNPs in roots; B, average number of FNPs in root).

## 4. Conclusions

Recently, NPs have been detected in crops such as apples in Italy, raising concerns about the accumulation of NPs in the human body through the food chain. This issue is closely linked to the widespread use of plastic products in agriculture, such as plastic films and pesticide containers. The purpose of this study was to inhibit the penetration of NPs from soil into crops by electrically capturing NPs.

Using copper plates, lead wires, or graphite rods as electrodes, it was visually evaluated whether NPs are electrically migrated. While the migration of NPs was observed with copper plates or lead wire electrodes, significant issues arose: the copper plates oxidized, and the lead wires degraded. In contrast, the graphite rods showed no changes after the electrical supply was completed, indicating that they are a suitable material for facilitating the electric migration of NPs. However, graphite rods have small surface areas and are prone to breakage under minor impacts, making them unsuitable for direct application in soil. This highlights the need for a flexible graphite structure with excellent electrical conductivity that can withstand practical use.

Graphite is composed of a structure in which carbon atoms are connected in planes of graphene stacked layer upon layer and has excellent electrical conductivity and non-toxic properties, making it an advantageous material for environmental applications. Since obtaining pure graphene in large quantities is difficult, a graphene-mimicking structure was developed by coating graphite powder on cellophane tape or agarose film (CTcGP or AFcGP, respectively). CTcGP and AFcGP exhibited electrical conductivity and successfully captured fluorescently labeled NPs in the soil. Moreover, while NPs in the soil penetrated plant roots, their penetration was strongly inhibited by CTcGP or AFcGP when electricity was supplied.



**Figure 14.** Application example of graphene-mimic films or panels for capturing NPs in soil and for preventing the penetration of NPs from soil into crops.

The CTcGP or AFcGP developed in this study effectively operated at a low voltage of 9 V, meaning that they could function efficiently with electricity generated from solar panels without additional devices, such as electrical transformers, making them highly cost-efficient (Figure 14). However, cellophane tape, which is made from plastic, and agarose film are easily degraded by moisture, posing the risk of captured NPs being released back into the soil. Therefore, further research is required to explore methods for coating graphite powder onto materials, such as glass, that resist decomposition or deformation by moisture or electricity and remain non-toxic to living organisms.

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## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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