Development of Hydrophobic Fabrics Modified with Graphene: A Systematic Review

Rodrigo Denizarte de Oliveira Polkowski^{1*}, Pollyana Silva Melo¹,

Leonardo Freitas¹, Katielly Vianna Polkowski¹

¹TRL9 TECH Research and Experimental Development in Physical and Natural Sciences Ltda; Salvador, Bahia, Brazil

Pursuing fabric with hydrophobicity and self-cleaning properties has been a focal point across various applications. Graphene has emerged as a prominent contender for fabric modification to augment hydrophobicity. This study endeavors to conduct a systematic review of fabrics imbued with graphene for hydrophobic applications, concentrating on the most recent five years and employing the PRISMA method (Preferred Reporting Items for Systematic Reviews and Meta-analyses). Articles were curated from reputable databases, including Scopus, Web of Science, and Science Direct. Following meticulous inclusion/exclusion criteria, thirty-seven studies were deemed eligible for inclusion in the review. Keywords: Graphene. Fabric. Textile. Hydrophobic. Nanocomposite.

In recent years, nanotechnology has allowed the incorporation of nanoparticles into multifunctional textiles, adding new physical and chemical characteristics to fibers, threads, and fabrics. Fabrics with hydrophobic properties similar to lotus leaf surfaces have recently drawn the attention of researchers due to their significant potential for scientific and industrial applications [1-3].

Graphene has stood out recently due to its remarkable properties, such as fracture resistance, thermal conductivity, impermeability, and hydrophobicity. In addition to these applications, a wide range of graphene applications includes electrical conductivity [2-4], usage as flame retardants [5], energy storage and conversion [6], dye removal [7], and antibacterial function [8]. However, applying graphene to a polymeric matrix (fabric) remains a significant challenge for researchers, particularly when considering highscale production and low cost.

PRISMA, a reporting guideline for systematic reviews and meta-analyses, was developed to improve reporting quality. It helps plan and document review methods [10]. Systematic

J Bioeng. Tech. Health 2024;7(1):87-92 [©] 2024 by SENAI CIMATEC. All rights reserved.

reviews synthesize knowledge in afield, identifying research gaps and evaluating theories [11,12]. Our study systematically reviews graphene coated fabrics for hydrophobic applications, focusing on graphene incorporation into fibers and its uses. We consider publications from 2018 to 2023.

Materials and Methods

In this study, we performed a systematic review using the PRISMA method [10]. We searched Scopus, Web of Science, and Science Direct for articles in English published between 2018 and 2023, using keywords like graphene, fabric, textile, cloth, and hydrophobic. Initially, we found 449 studies. Two reviewers independently assessed the relevance of titles and abstracts, excluding conference papers, review studies, and duplicates (159), resulting in 290 studies for further analysis. Figure 1 depicts the study selection process following PRISMA guidelines.

After the initial selection, exclusion criteria were applied based on the absence of graphene impregnation (108), studies related to sensors (19), films (11), electronics (1), biological applications (7), and electronic devices (10), resulting in a total of 55 studies deemed relevant to the subject under analysis. The identified articles underwent independent evaluation of their titles and abstracts to assess their suitability, leading to a total of 37 articles that were included in this study.

Received on 17 September 2023; revised 15 December 2023. Address for correspondence: Rodrigo Denizarte de Oliveira Polkowski. Rua Mundo, No. 121, 409C, Bahia Technology Park, Trobogy, Salvador - BA, Brazil, Zipcode 41745-715. E-mail: rodrigo@trl9.tech.

Figure 1. Systematic review flowchart according to the PRISMA protocol.



Results and Discussion

Figure 2 presents a graph containing data obtained through the PRISMA protocol, showing an increase in studies related to fabrics coated with graphene over the last five years. Until this research, when the data was compiled, there were already 3 articles in this area, and more studies are expected. The highest number of studies was recorded in the years 2019 and 2022. In 2019, there was an increase of approximately 122% compared to the previous year, while in the period from 2020 to 2021, the number of registrations remained the same as in 2018. The most significant increase in registrations was identified in 2022, with a 155% increase compared to 2018, 2020, and 2021.

The systematic review focused on evaluating graphene synthesis methods, particularly emphasizing the chemical reduction of graphene oxide (GO) for fabric applications. Among the reviewed studies, GO was prevalent, featuring 25 studies related to hydrophobic fabrics, while other types of graphene (sheets, reduced, and quantum) were identified in 12 studies. GO is distinguished by its numerous oxygen-based functional groups on its surface, providing active sites for chemical functionalization and developing specific properties. The synthesis of GO can be achieved through three primary methods: Brodie's, Staudenmaier's, and Hummer's methods, with the latter being the most frequently cited in this study [2,4,13-18]. The studies were further categorized





Figure 3. Studies published in this systematic review by application method.



based on the method of graphene application onto the fabric surface, with the dip-coating technique emerging as the most commonly mentioned (15 studies) due to its efficacy in achieving aligned and layered structures for high-quality coatings (Figure 4).

Dip-coating is a widely utilized process for dyeing, printing, or applying various chemical finishes to textile materials such as fabrics, knits, yarns, and nonwovens. This technique has gained prominence in fabric applications owing to its simplicity, strong adhesion, easy availability, and potential for large-scale production [1,5,6,19-23]. Fang and colleagues (2019) studied the preparation of glass fibers coated with graphene and high electrical conductivity through the sol-gel and dip-



Figure 4. Studies published in this systematic review by water contact angle.

coating techniques, and they reported that the dipcoating method achieved the best properties [16].

The studies included in this systematic review employed various analyses, such as scanning electron microscopy (SEM), thermogravimetry (TGA), Fourier-transform infrared spectroscopy (FTIR), dynamic mechanical analysis (DMA), X-ray diffraction (XRD), and X-ray photoelectron spectroscopy (XPS), to evaluate the efficiency of graphene incorporation into the fabric's structure. However, these studies' primary property and objective were hydrophobicity. The "Lotus Effect," described by Oles and colleagues (2000), refers to the physical properties that characterize a superhydrophobic and selfcleaning surface. This property is typically assessed through contact angle analysis, where a contact angle less than 90° indicates a hydrophilic surface, meaning the liquid can wet it. Conversely, a contact angle greater than 90° indicates a hydrophobic surface, with a contact angle exceeding 150° termed superhydrophobic. In this systematic review, 35 studies evaluated hydrophobicity through contact angle measurements. Figure 5 illustrates the relationship between the highest contact angle values achieved after the incorporation or coating of the fabric with graphene.

Out of the total of 35 studies that evaluated the contact angle of the fabric after graphene modification, 28 studies successfully modified the fabric surface to achieve a hydrophobic classification [1,2,3,6,9,14-18,20,22,25-38].

Various resin matrices, including PET, PU, PP, and polyester, were examined in multiple studies.

The focus was on functionalizing graphene, not resin synthesis. For instance, Achagri and colleagues (2020) achieved a 148° contact angle on PET fabric using dip-coating [1]. Atighi and colleagues (2022) and Wang and colleagues (2018) also obtained contact angles exceeding 100° for PET matrices. PET exhibits partial hydrophilic properties due to its oxygen-containing groups, resulting in a 71.4° contact angle. Coating with graphene oxide increased the angle to 116.3° [36].

Among the studies in this review (37), 65% focused on developing fabrics for oil-water separationapplications. Otherapplications included flame retardancy [38], catalytic activity [21], self-cleaning [4], and anticorrosive coatings [26,28,39].

Conclusion

The systematic review presented in this article provides a comprehensive overview of recent studies focused on fabric modification with graphene for hydrophobic applications. The reviewed works demonstrate a significant enhancement in fabric hydrophobicity following graphene modification in various forms. Dip-coating emerged as the predominant and favored approach among the different application methodologies explored in the reviewed studies.

Notably, graphene oxide was found to be the most commonly utilized variant of graphene, significantly surpassing other graphene forms in frequency of usage.

The integration of the dip-coating technique with graphene oxide infusion was of particular

significance. This combined approach resulted in a consistent and notable improvement in hydrophobic properties across the reviewed studies. The synergistic effects of dip-coating and graphene oxide infusion consistently yielded superior results, substantially enhancing the materials' ability to resist wetting and repel water.

Acknowledgments

The authors thank the TRL9 TECH Company and Brazilian Research Agency-CNPq for all the technical and financial support.

References

- Achagri G, Essamlali Y, Amadine O, Majdoub M, Chakir A, Zahouily M. Surface modification of highly hydrophobic polyesterfabric coated with octadecylamine functionalized graphene nanosheets. RSC Adv. 2020 Jul 1;10(42):24941- 50.
- 2. Hou M, Hong X, Tang Y, Jin Z, Zhu C, Tao C, etal. Chemically reduced graphene oxide-coated knitted fabric imparted conductivity and outstanding hydrophobicity. Textile Research Journal. 2021 Oct 1;91(19-20):2169-83.
- Wang C, Guo R, Lan J, Tan L, Jiang S, Xiang C. Preparation of multi-functional fabric via silver/reduced graphene oxide coating with poly(diallyldimethylammonium chloride) modification. Journal of Materiais Science: Materiais in Electronics 2018 May 1;29(10):8010-9.
- Li T, Zou L, Cheng K, Liu X, Shi H et al. Environmenttolerant conductive and superhydrophobic poly(mphenylene isophthalamide) fabric prepared via y-ray activation and reduced graphene oxide/nano SiO2 modification. J Appl Polym Sei 2022 Apr 20;139(16).
- Ahmed T, Mohammed Q, Subeshan B, Rahman M, Nuraje N, Asmatulu E. Improving flame retardancy and hydrophobicity of fabrics via graphene inclusion obtained from recycled batteries. Mater Today Proc.2022 Jan 1;71:78-89.
- Zhang P, Gu L, Liu W, Ge D, Yang L, Guo Y, et al. Underwater highly pressure sensitive fabric based on electric-induced alignment of graphene. Materiais 2023 Feb 1;16(4).
- Zahid M, Saeeda M, Nadeem N, Shakir HMF, El-Saoud WA, Attala OA et al. Carboxylated graphene oxide (c-GO) embedded thermoplastic polyurethane (TPU) mixed matrix membrane with improved physicochemical characteristics. Membranas (Basel) 2023 Feb 1;13(2).

- Zahid M, Khalid T, Rehan ZA, Javed T, Akram S, Rashid A et al. Fabrication and characterization of sulfonated graphene oxide (Sgo) doped pvdf nanocomposite membranes with improved anti-biofou ling performance. Membranas (Basel) 2021 Oct 1;11(10).
- 9. Jia G, PlentzJ, Dellith J, Dellith A, Wahyuono RA, Andra G. Large area graphene deposition on hydrophobic surfaces, flexible textiles, glass fibers and 3D structures. Coatings 2019;9(3).
- Shamseer L, Moher D, Clarke M, Ghersi D, Liberati A, Petticrew M, et ai. Preferred reporting items for systematic review and meta-analysis protocols (prisma-p) 2015: Elaboration and explanation. BMJ (Online). BMJ Publishing Group 2015;349.
- 11. Gopalakrishnan S, Ganeshkumar P. Systematic reviews and meta-analysis: Understanding the best evidence in primary healthcare. J Family Med Prim Care 2013;2(1):9.
- 12. Gough D, Thomas J, Oliver S. Clarifying differences between reviews within evidence ecosystems. Systematic Reviews. BioMed Central 2019;8.
- 13. Dhumal PS, Khose R V., Wadekar PH, Lokhande KD, Some S. Graphene bentonitesupported freestanding,flexible membrane with switchablewettability forselective oil-waterseparation. Sep PurifTechnol 2021 Jul 1;266.
- Zhu X, Gao Z, Li F, Miao G, Xu T, Miao X et al. Superlyophobic graphene oxide/polydopamine coating under liquid system for liquid/liquid separation, dye removal, and anti-corrosion. Carbon N Y. 2022 Apr 30;190:329-36.
- 15. Li Y, Li Z, Tian M, Qu L, Chen Z, Zhu S et al. Reduction and deposition of graphene oxide nanosheets on the multifunctional hydrophobic polyester nonwoven fabric via a one-step hydrothermal route. Mater Res Express 2019 May 17;6(8).
- 16. Fang M, Xiong X, Hao Y, Zhang T, Wang H, Cheng HM, et ai. Preparation of highly conductive graphenecoated glass fibers by sol-gel and dip-coating method. J Mater Sei Teehnol 2019 Sep 1;35(9):1989-95.
- Wang D, Li D, Lv P, Xu Y, Wei Q. Deposition of polytetrafluoroethylene nanoparticles on graphene oxide/polyester fabrics for oil adsorption. Surface Engineering 2019 May 4;35(5):426-34.
- Guo G, Liu L, Zhang Q, Pan C, Zou Q. Solutionprocessable, durable, scalable, fluorine-grafted graphene-based superhydrophobic coating for highly efficient oil/water separation under harsh environment. New Journal of Chemistry 2018;42(5):3819-27.
- 19. Abbas TM, Hussein SI. Improving the mechanical properties, roughness, thermal stability, and contact angle of the acrylic polymer by graphene and carbon fiber doping for waterproof coatings. J lnorg Organomet Polym Mater 2022 Oct 1;32(10):3788-96.

- 20. Xu C, Gou W, Wang X, Zhou J, Liu J, Chen K. Synthesisof paraffin@PS/reduced graphene oxide microcapsules via Pickering emulsion for multiprotective coatings. Colloids Surf A Physieoehem Eng Asp 2021 Mar 20;613.
- 21. Sharma K, Ali M, Singh R, Majhi S, Sharma S, Tripathi CSP et al. Silver nanoparticles decorated on graphene oxide modified polyester fabric: Catalytic reduction of 4-nitrophenol, organic dyes and SERS application. Journal of Physies and Chemistry of Solids 2022 Jun 1;165.
- 22. Galante AJ, Yates KA, Romanowski EG, Shanks RMQ, Leu PW. Coal-derived functionalized nanographene oxide for bleach washable, durable antiviral fabric coatings. ACS Appl Nano Mater 2022 Jan 28;5(1):718-28.
- 23. Ferrero G, Sandgren Bock M, Stenby EH, Hou C, Zhang J. Reduced graphene oxide-coated microfibers for oil-water separation. Environ Seil Nano.2019;6(11):3215-24.
- 24. Oles M, Nun E, Dambacher G, Schleich B. Learning from nature-polymer surfaces with a nano structure that utilise the Lotus-Effect and clean themselves. 1st Annual International IEEE-EMBS Special Topic Conference on Microtechnologies in Medicine and Biology. Proceedings (Cat. No.00EX451), Lyon, France, 2000:331-333. Doi: 10.1109/MMB.2000.893798.
- 25. Li G, Hong G, Dong D, Song W, Zhang X. Multiresponsive graphene-aerogel directed phasechange smart fibers. Advaneed Materials 2018 Jul 26;30(30).
- 26. Uzoma PC, Liu F, Xu L, Zhang Z, Han EH, Ke W et al. Superhydrophobicity, conductivity and anticorrosion of robust siloxane-acrylic coatings modified with graphene nanosheets. Prog Org Coat 2019 Feb 1;127:239-51.
- 27. Chen K, Liu H, Zhou J, Sun Y, Yu K. Polyurethane Blended with Silica Nanoparticle-Modified Graphene as a Flexible and Superhydrophobic Conductive Coating with a Self-Healing Ability for Sensing Applications. ACS Appl Nano Mater 2022 Jan 28;5(1):615-25.
- 28. Uzoma PC, Wang Q, Zhang W, Gao NJ, Liu F, Han EH. Investigation of the wettability, anticorrosion, and accelerated weathering behaviors of siloxane modified acrylic resin and functionalized graphene nanocomposite coatings on LY12 aluminum alloy. J Coat Technol Res 2021 May 1;18(3):789-806.
- 29. Liu C, Liao X, Shao W, Liu F, Ding B, Ren G et al. Hotmelt adhesive bonding of polyurethane/fluorinated polyurethane/alkylsilane-functionalized graphene nanofibrous fabrics with enhanced waterproofness,

breathability, and mechan ical properties. Polymers (Basel) 2020 Apr 1;12(4).

- 30. Tukawski D, Lisiecki F, Dudkowiak A. Coating cellulosic materials with graphene for selective absorption of oils and organic solvents from water. Fibers and Polymers 2018 Mar 1;19(3):524-30.
- 31. Zhu J, Zhang S, Wang L, Jia D, Xu M, Zhao Z, et ai. Engineering cross-linking by coal-based graphene quantum dots toward tough, flexible, and hydrophobic electrospun carbon nanofiberfabrics. Carbon N Y. 2018 Apr 1;129:54-62.
- 32. Naseem S, Wu CM, Xu TZ, Lai CC, Rwei SP. Oilwaterseparation of electrospun cellulose triacetate nanofiber membranes modified by electrophoretically deposited TiO2/graphene oxide. Polymers (Basel).2018 Ju15;10(7).
- 33. Hu Y, Chen Z, Wu C, Wang F, Cui S, Ai S et al. The Synthesis and Research of Glass Fiber Felts Coated with Graphene Oxide/PhenolicResin Binder. Fibers and Polymers 2019 Apr 1;20(4):732-8.
- 34. Zhao J, Han P, Quan Q, Shan Y, Zhang T, Wang J et al. A convenientoil-water separator from polybutylmethacrylate/graphene-deposited polyethylene terephthalate nonwoven fabricated by a facile coating method. Prog Org Coat 2018 Feb 1;115:181-7.
- 35. Atighi M, Hasanzadeh M, Sadatalhosseini AA, Azimzadeh HR. Metal-organic framework@graphene oxide composite-incorporated polyacryl on nitrile nanofibrous filters for highly efficient particulate matter removal and breath monitoring. Ind Eng Chem Res 2022 Dec 28;61(51):18613-24.
- 36. Xiang C, Guo R, Lan J, Jiang S, Wang C, Du Z et al. Self-assembling porous 3D titanium dioxide-reduced graphene oxide aerogel for the tunable absorption of oleie acid and RhodamineB dye. J Alloys Compd 2018 Feb 25;735:246-52.
- 37. Zhou J, Ou E, He Y, Fan Y, Ye Y, Tang B. Preparation of carbonized kapok fiber/reduced graphene oxide aerogel for oil-water separation. Chem Eng Technol.2020 Dec 1;43(12):2418-27.
- Ahmed T, Mohammed Q, Subeshan B, Rahman M, Nuraje N, Asmatulu E. Improving flame retardancy and hydrophobicity of fabrics via graphene inclusion obtained from recycled batteries. Mater Today Proc 2022 Jan 1;71:78-89.
- 39. Sharif NA, Ehsani M, Zaarei D, Kalaee MR, Khajavi R. Nanocomposite coatings based on modified graphene oxide and polydimethylsiloxane: characterization and thermal properties. Russian Journal of Applied Chemistry 2020 Nov 1;93(11):1765-73.