

Literature Review of the Use of Zinc and Zinc Compounds in Paper-Based Microfluidic Devices

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Abstract

Zinc and its compounds, alloys and composites play an important role in the modern day world and find application in almost every aspect that can improve the quality of our lives. This ranges from supplements and pharmaceuticals that are meant to improve our health and wellbeing to additives meant to guard or reduce corrosion in metals. However, over the past several years, a new area of technology has been garnering a great deal of attention and has made use of zinc and its compounds. This is with reference to paper-based microfluidic technology that offers several advantages and that keeps expanding in the amount of applications it covers. In this paper, a review is offered for the applications that have used zinc or zinc compounds in paper-based microfluidic devices.

Keywords

Zinc, Zinc Oxide, Zinc Compounds, Paper-Based Devices, Paper Microfluidics, Low-Cost Platforms, Point-of-Care Diagnostics

1. Introduction

Paper has been used for biological and chemical applications for over a century [1] [2]. However, over the past several years, a lot of research and resources have been dedicated to developing new paper-based devices, or improving their performance, for use in a wide range of applications. Paper-based devices have been used for biomedical applications [3], food safety [4], soil analysis [5], water analysis [6] and in many other miscellaneous applications [7]. Muller *et al.* [8] may have developed the first simple paper-based microfluidic device in 1949 [9]

[10] [11]; however, it was the Whitesides group in 2007 [12] who gave a renewed view and push for the possible applications of this technology. The main advantages of paper-based microfluidic technology is that these devices are inexpensive, simple, portable and easy to use. However, the most significant advantage is that these devices depend on capillary action to flow the sample across the different sections of the device without a need for a pump, which results in device miniaturization and cost savings. Paper-based microfluidic devices have adapted several of the conventional detection techniques such as colorimetric detection, electrochemical detection, chemiluminescence, fluorescence, mass spectrometry and surface-enhanced Raman spectroscopy [13]. And since zinc and its compounds are utilized in a broad range of applications due to their inherent advantages, it was clear that they were to be used in paper-based microfluidic devices.

Zinc is an essential trace element for the human body since it plays an important role in maintaining cell health and functionality and is crucial for the function of more than 300 enzymes [14] [15]. The human body contains 2 to 3 grams of zinc and requires a daily intake of 10 to 15 mg [16]. Zinc also finds use in many different applications outside of the human body mainly due to its abundance and nontoxicity [17]. Zinc and zinc oxide have been used in the rubber industry, pharmaceutical and cosmetic industries, textile industry, electronics industry, for enhancing filtration, in making batteries, and in many other miscellaneous applications [18] [19] [20] [21]. Zinc oxide is an inorganic material with specific properties that is abundantly produced after silicon dioxide and titanium dioxide [22]. Zinc oxide nanomaterial is stable, safe to human beings and has been categorized by the Food and Drug Administration in the United States as “generally recognized as safe” [23]. It has been vastly used in environmental and medical applications due to its nontoxicity, biosafety, biocompatibility and biodegradability [24]. Textiles have been functionalized by zinc oxide to impart favorable properties such as improved antimicrobial [25], super hydrophobicity [26], self-cleaning [27], thermal insulation [28], UV-absorption [29], and flame retardancy [30]. The cotton fibers in paper-based devices have also been functionalized by zinc oxide to provide or enhance analytical or engineering purposes.

In this review, some of the paper-based microfluidic devices that have used zinc or zinc compounds will be highlighted with the different applications they were used in.

2. Application of Zinc and Zinc Compounds in Paper-Based Devices

2.1. Biomedical

Zinc and its oxide are biocompatible [31] [32] and have been used in several paper-based devices for different biomedical applications. Li *et al.* [33] used zinc oxide nanowires for the electrochemical detection of glucose in spiked phosphate-buffered saline and human serum, **Figure 1**. However, they anticipate that

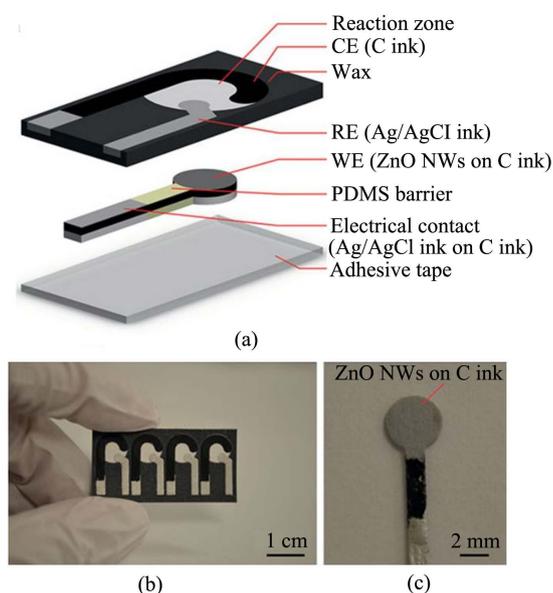


Figure 1. Paper-based device developed by Li *et al.* [33] that uses zinc oxide nanowires for the electrochemical detection of glucose.

their device can be used to detect glucose in whole blood if a filtering membrane is added on top of their device's reaction zone to filter out the blood cells. Ferreira *et al.* [34] used zinc microparticles as the reducing agent in a paper-based device to detect nitrate in human saliva.

Detection of biomarkers using paper-based devices will help in making rapid and informed decisions whether further investigation is needed to determine the health status of an individual. Tiwari *et al.* [35] functionalized paper with zinc oxide nanorods to preconcentrate myoglobin which is a biomarker for heart disease. Guo *et al.* [36] developed a paper-based device integrated with zinc oxide nanowires that can detect three cardiac biomarkers which are the human heart-type fatty acid binding protein, cardiac troponin I, and myoglobin, simultaneously. Kong *et al.* [37] constructed a paper-based device utilizing zinc oxide nanoflowers for the sensitive detection of the biomolecules, L-glutamic acid and L-cysteine. Sun *et al.* [38] constructed an ultrasensitive electrochemical immunosensor using branched zinc oxide nanorods and porous zinc oxide spheres with gold nanoparticle composite for the detection of α -fetoprotein which is a tumor marker used in the diagnosis of hepatocellular carcinoma. Sun *et al.* [39] used tetra-carboxyl naphthalocyanine zinc sensitized zinc oxide nanorods as photoactive materials in a paper-based photoelectrochemical immunosensor to detect the carcinoembryonic antigen. Liu *et al.* [40] constructed a paper-based device for the colorimetric determination of the carcinoembryonic antigen based on the intrinsic peroxidase activity of zinc ferrite (ZnFe_2O_4)-multiwalled carbon nanotubes. Wang *et al.* [41] constructed a hollow-channel paper-based device with CdS quantum dots/reduced graphene oxide/zinc oxide nanorod arrays for the photoelectrochemical detection of multiple microRNAs.

Portable, rapid and sensitive detection of diseases, particularly infectious,

would play a significant role in decreasing their spread. Li *et al.* [42] reported on the first microfluidic paper-based origami nanobiosensor that uses zinc oxide nanowires to improve the biosensing performance of detecting the human immunodeficiency virus p24 antigen. Li *et al.* [43] also used zinc oxide nanowires directly grown on the working electrode of the paper-based device to enhance the detection of the p24 antigen used as a marker for the human immunodeficiency virus and showed that their device was capable of differentiating between concentrations of CR3022, an IgG antibody to the COVID-19 coronavirus. Narang *et al.* [44] developed a paper-based device incorporating zinc-silver nanobooms that used cyclic voltammetry for the electrochemical detection of the herpes virus DNA. Kim [45] grew zinc oxide nanorods on cellulose paper and then added gold nanoparticles to these rods so as to enhance the Raman signal in detecting the presence of prenatal diseases and identifying the types of diseases from amniotic fluids.

Finally, it is very useful to have a simple and rapid method for detecting different drugs. Caglayan *et al.* [46] used a fluorescent coordinatively unsaturated metal complexes based on the zinc II ion to provide an easy-to-detect fluorescence response in the detection of the banned performance-enhancing drug Adrafinil. Narang *et al.* [47] developed a paper-based device for the detection of methylenedioxymethamphetamine which is an addictive narcotic and a potential neurotoxic agent used as a recreational drug.

2.2. Environmental Applications

Paper-based microfluidic devices generally integrate a suitable detection method such as colorimetric, electrochemical or fluorescent techniques for quantitative analysis of nutrients in air, soil, or water [11]. Zinc and its compounds have been used in microfluidic paper-based devices for environmental applications to provide rapid, inexpensive, and quantitative analysis. Nitrate is part of the nitrogen cycle but contributes to water nutrient pollution when present at elevated concentrations [48] [49]. Jayawardane *et al.* [50] and Charbaji *et al.* [51] used zinc microparticles as a reducing agent in paper-based devices meant for the detection of nitrate in water. The zinc microparticles provided the reduction of nitrate to nitrite needed for its detection by the Griess assay. **Figure 2** shows the paper-based device developed by Charbaji *et al.* for detecting nitrate in water. While Jayawardane *et al.* created a reduction channel by depositing zinc microparticles on the surface of paper, Charbaji *et al.* used a newly developed composite material made up of cellulose and zinc microparticles, that they called Zin-culose [52], **Figure 3**, to provide the required reduction step. Zhang *et al.* [53] used dual-emission manganese-doped ZnS semiconductor nanocrystals on paper-based test strips for the visual detection of diethylphosphorothioate (organophosphate) residues.

Another environmental application that made use of paper-based microfluidic devices was the detection of pentachlorophenol. Pentachlorophenol is a xenobiotic that enters the environment as a byproduct of various industrial processes

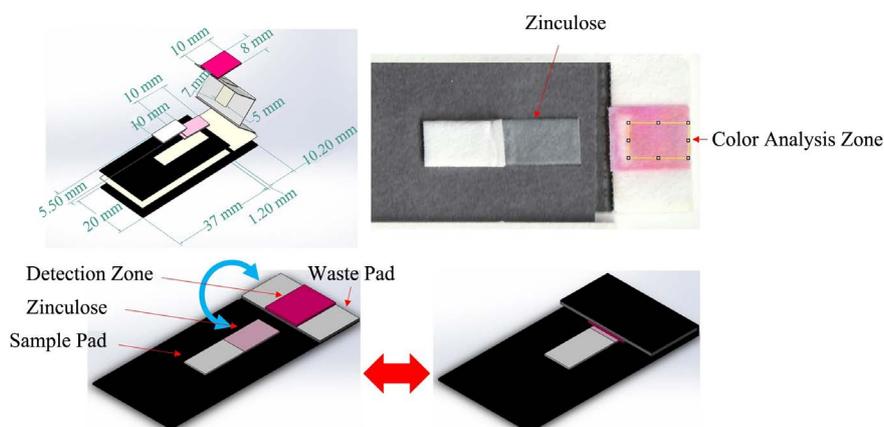


Figure 2. Paper-based device developed by Charbaji *et al.* [51] for the detection of nitrate in water.

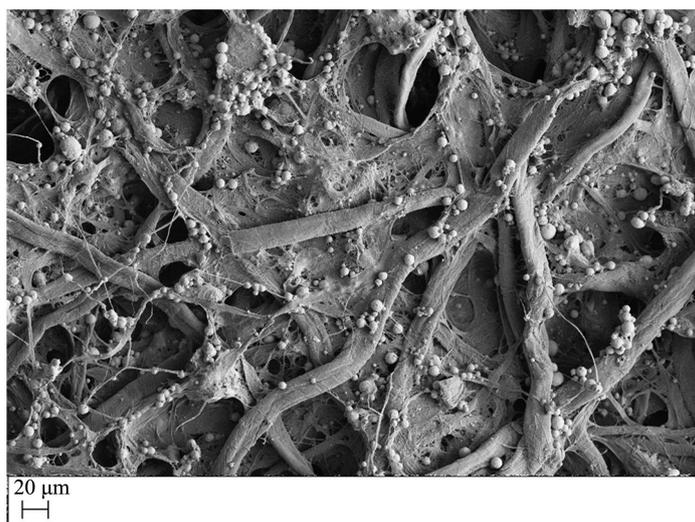


Figure 3. A strip of Zinculose observed using a scanning electron microscope at a magnification of 500× [52].

and is a cause for great environmental concern. Sun *et al.* [54] developed a disposable paper-based microfluidic origami device with functionalized zinc oxide nanospheres to detect pentachlorophenol.

Paper-based devices were also used in the detection of gases. Gimenez *et al.* [55] developed a paper-based oxygen sensor that uses the absorption and desorption of oxygen from the zinc oxide crystals deposited and dried over graphite electrodes drawn on paper for detection. Koga *et al.* [56] deposited zinc oxide on paper using a two-step paper-making process to create a molecular sensor and they used nitrogen dioxide to test its sensing performance.

2.3. Food Safety

Monitoring the quality of agricultural and food products in order to ensure its safety is of utmost importance to safeguard the health and wellbeing of consumers. Developing small-size field deployable sensing systems and combining that

with state-of-the-art communication techniques is a way to perform qualitative and quantitative multi-component analysis of food products to guarantee its safety [57]. Paper-based devices have many desirable advantages that make them ideal candidates for such an application and they have been demonstrated to be effective in testing the quality of food products [58]. Several paper-based devices utilizing zinc or its compounds have been developed to detect different analytes or toxins in food. Yukird *et al.* [59] deposited zinc oxide from a solution over 2 detection zones in a paper-based device. The first was a multiwall carbon nanotube electrode and the second was a laser desorption ionization mass spectrometric detection zone. This was meant to improve their sensitivity in detecting “bisphenol A” which is a chemical compound used in the production of food containers.

Chen *et al.* [60] developed a fluorescence paper-based sensor based on zinc 5, 10, 15, 20-tetra(4-pyridyl)-21H-23H-porphine (nano ZnTPyP) quenching CdTe quantum dots for the detection of three carbamate pesticides which are metolcarb, carbofuran, and carbaryl. Teepo *et al.* [61] and Ratnarathorn *et al.* [62] used zinc microparticles as the reducing agent in paper-based devices to detect the presence of nitrate in food samples. **Figure 4** shows the paper-based device developed by Teepo *et al.* for detecting nitrate in food samples.

Chen *et al.* [63] established a high sensitivity paper-based fluorescent sensor to detect L-theanine in tea water which is one of the markers used to evaluate the sweetness and freshness of tea. Their device used CdTe quantum dots/corn carbon dots with nano tetra pyridel-porphine zinc to provide the quenching effect. Another paper-based fluorescent sensor utilizing CdTe quantum dots and spherical nano tetra pyridel-porphine zinc was developed by Chen *et al.* [64] for the identification and quantitative analysis of caffeine.

2.4. Miscellaneous Applications

Although the above sections highlighted microfluidic paper-based devices developed for biomedical, environmental and food safety applications, it's important to mention that there are a lot of other applications that have also used zinc

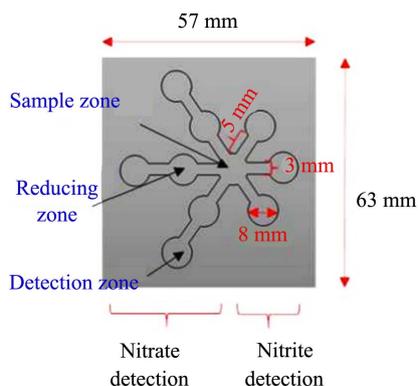


Figure 4. Paper-based device developed by Teepo *et al.* [61] for the detection of nitrate in food samples. The reducing zone contained zinc dust.

or its compounds in paper-based devices. This section will refer to some of these paper-based devices used for miscellaneous applications. Song *et al.* [65] have provided a decent review of paper-based physical sensors utilizing zinc oxide nanostructures up to the year 2017. Therefore, this review looks at papers published from 2017 onwards for paper-based physical sensors utilizing zinc compounds. Wang *et al.* [66] developed a one-axis piezoelectric accelerometer using zinc oxide nanowires that have been grown on paper, **Figure 5**. The fabrication of their device is inexpensive and doesn't require sophisticated equipment. Dubourg *et al.* [67] developed a paper-based ultraviolet sensor utilizing zinc oxide. They used a laser post-processing step to induce significant modifications to the surface morphology and structure of the zinc oxide film. This contributed to the super-hydrophobicity of the printed zinc oxide nanoparticles which reduced humidity interference while enhancing sensitivity for ultraviolet detection.

Some paper-based devices were also developed in order to provide a flexible power-source. Purohit *et al.* [68] developed microfluidic galvanic and hybrid cells on a single layer of paper. The cell had zinc and copper powder that was painted on the paper to serve as the anode and cathode. Zhang *et al.* [69] developed a zinc-air battery with zinc foil pasted on titanium foil to act as the anode. Burrola *et al.* [70] presented an alkaline nickel oxide hydroxide/zinc battery with a zinc anode cut from commercially available sheets.

An interesting paper-based device using zinc oxide was developed by Zhang *et al.* [71]. They used zinc oxide nanorods to fabricate a flexible light emitting diode on paper and used it as an excitation light source in a multiplexed photoelectrochemical immunosensor.

3. Challenges and Future Trends

Zinc, its oxide and compounds have been seeing increased use in paper-based microfluidic devices, and their usage is expected to expand further to encompass more applications. However, some challenges still need to be overcome to ensure repeatability in the preparation of zinc oxide nanoparticles to enable the surface modification of these particles with organic compounds [20]. Also, zinc oxide may need to be combined with other material such as metals, semiconductors,

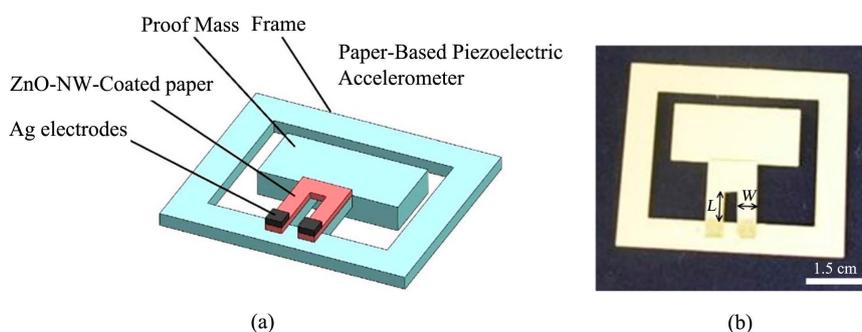


Figure 5. (a) Schematic of the paper-based one-axis piezoelectric accelerometer developed by Wang *et al.* [66]; (b) Photograph of the paper-based device.

and nanocarbons to overcome its limitations such as fast recombination of photogenerated electron-hole pairs, photocorrosion and need for UV light for activation in photocatalytic applications [72].

The development of composite materials using zinc microparticles such as Zinculose [52] will help improve the performance of paper-based devices using zinc microparticles in different applications. As paper-based electrochemical devices start to play a bigger role in the paper-based platform technology, use of zinc oxide is expected to enhance the sensitivity and selectivity of these devices [73]. Zinc oxide has demonstrated effectiveness in biomedical fluorescence assays [74] and will be instrumental when more paper-based devices are developed for fluorescence assays. It is anticipated that the future direction is to develop more paper-based devices for photonic and electronic applications which will use zinc oxide nanoparticles since these particles are ideal for use in the field of photonics, nanoscale electronics and optoelectronics [75] [76].

4. Conclusion

Paper-based microfluidic technology is a field that is still developing with frequent new applications and advancements being achieved. Zinc, its oxide and compounds have been utilized in a lot of different applications using paper-based technology. This is the case since zinc, zinc oxide and zinc compounds are inexpensive, biocompatible, non-toxic, environmentally friendly, and have their distinctive physical and chemical properties. They have been widely utilized to improve the properties and performance of cellulose substrates in paper-based analytical devices. In this review, paper-based devices that have used zinc or zinc compounds in different applications were highlighted. These devices were developed for biomedical, environmental, food safety and several other miscellaneous applications. However, it is anticipated that more devices utilizing zinc, its oxide or compounds will be developed in the future. These devices will either improve the performance of the ones that have already been designed before or will be completely novel sensors for new applications.

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Conflicts of Interest

The authors declare no conflict of interest.

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