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Perspective

Developing Wound Moisture Sensors: Opportunities and Challenges for Laser-Induced Graphene-Based Materials

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Abstract: Recent advances in polymer composites have led to new, multifunctional wound dressings that can greatly improve healing processes, but assessing the moisture status of the underlying wound site still requires frequent visual inspection. Moisture is a key mediator in tissue regeneration and it has long been recognised that there is an opportunity for smart systems to provide quantitative information such that dressing selection can be optimised and nursing time prioritised. Composite technologies have a rich history in the development of moisture/humidity sensors but the challenges presented within the clinical context have been considerable. This review aims to train a spotlight on existing barriers and highlight how laser-induced graphene could lead to emerging material design strategies that could allow clinically acceptable systems to emerge.

Keywords: wounds; community nursing; sensors; moisture; RFID; laser-induced graphene; LIG; polymers

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1. Introduction

The quantitative measurement of moisture level is a core requisite within the agri-food sector and countless industrial processes, and it is little surprise that a wealth of strategies have been employed to facilitate rapid, real-time monitoring. Moisture level also plays a critical role within the healthcare sector and especially in wound management, but, as yet the translation of those technologies that are routinely applied within industrial processes have yet to make substantive headway at the front line of patient care. In many cases, the composition of the sensing systems would be far from appropriate when considering their application in clinical contexts, but there have been considerable advances in composite technologies within the emerging wearable sector to suggest that this may be beginning to change. Wound care has long been a recipient of major advances in polymer technologies, with many of the current dressings being composed of multi-layer multi-functional materials designed to protect the wound site, encourage healing and minimise damage upon redressing [1–4]. The evolution of smart dressings has been underway for some time, but it is only recently that manufacturers have started to explore the development of dressings with diagnostic capabilities [5–10].

The key device requirements are for the low-cost manufacture of conductive elements whose size, shape and mechanical flexibility will not impede the use of existing wound dressings nor impact on the underlying healing processes. There is an extensive literature base on the development of moisture/humidity sensors, but a detailed discussion of the respective merits and limitations is beyond the scope of the present communication. Rather, we posit the potential suitability of one subsection towards wound management: laser-induced graphene (LIG) [11–14]. While graphene has long been promoted as a wonder material for a multitude of sensing applications, the complexities of the processes needed for both reproducible manufacture and integration within the intended sensor have traditionally been problematic. The discovery of laser-induced graphene, however, has provided a step change in the development process as it allows direct scribing onto inexpensive films through simply changing the design within an appropriate graphic design application—much in the same way as an image is prepared for a conventional desktop printer [11]. This stands in marked contrast to conventional fabrication processes such as photolithography or screen printing, where even small changes incur significant cost overheads associated with the re-tooling of masks. The ability to rapidly prototype LIG-based sensors on a variety of substrates has undoubtedly increased interest in their use as electrode substrates, but the material properties of the resulting 3D carbon framework can also provide a number of advantages in terms of wound sensing applications. The intrinsic conductivity, mechanical flexibility and rich interfacial chemistry yield a functional material that could be ideally suited to the demands of moisture sensing within wound dressings. The aim of this review has been to consider how developments in LIG-based materials could be harnessed to address the challenges presented by such application and to critically assess their appropriateness and potential impact.

2. Wound Care Context

An appreciation of the significance of moisture in wound management dates back to antiquity, but the first scientific exploration is often attributed to Winter (1962) [15]. An appropriate level of moisture in the wound has since been proven to be critical in triggering re-epithelialisation and granulation processes, with wound hydration often considered to be a key factor contributing towards normal wound healing [16]. It is important to acknowledge, however, that excess hydration, especially when combined with a high level of matrix metalloproteinases, can lead to wound maceration, which will disrupt the healing process and can be common in chronic wounds [17]. In such conditions, the wound fluid itself serves as a "wounding agent" and hence monitoring the hydration level of a wound becomes critical. At present, this is done via visual inspection by the patient, caregiver or nurse when the dressing is replaced, but it is possible that the periwound surfaces could be exposed to the action of excess wound fluid/exudate for considerable periods before the physical signs of strikethrough or saturation appear [17].

While the frequency of dressing changes will vary from one patient to another, Ousey and colleagues (2013) conducted a detailed study of dressing management and found that the majority of wound dressings are replaced twice weekly [18]. It must be noted, however, that dressing changes do not simply occur when the signs of saturation are evident but that they are often changed when it is not necessary, creating avoidable excess waste. Milne and co-workers (2016) found that, in a study of 30 patients, some 45% of 588 dressings studied were changed while the wounds were considered to be within an optimum moisture range [19]. These values are comparable to those found by Ousey et al. (2013), where 55% of patient wound redressings were due to routine care changes. Ousey's study also found that, in fact, only around 15% of changes were due to the dressing being saturated, with the other 30% of dressing changes due to maximum wear time being reached. As dressing changes are often accompanied by pain for the patient, it is easy to appreciate how the introduction of a sensing system to monitor wound moisture levels would be very advantageous in preventing premature dressing changes and would ultimately improve patient care.

It is easy to rationalise how minimising disruption at the wound–dressing interface could reduce pain. There are also clear cost-saving opportunities in relation to staff time associated with the dressing change, but it can also be anticipated that critical management of the data from a wound moisture sensor could enable a more judicious choice of dressing type to better meet the requirements of a given wound. Optimisation of the dressing type could therefore speed up healing and, in doing so, lead to substantive savings. The integration of sensing technology here could dramatically improve nurse–patient interactions, allowing more efficient prioritisation and visit scheduling.

The scale of the problem is evident when considering that, in the UK, of the 2.2 million wounds being managed by the NHS, 1.45 million are treated by community/district nursing staff [20]. The treatment of chronic wounds is a particular issue where delays in healing and the development of complications will inevitably lead to longer and more intensive treatment and can lead to hospitalisation and the need for further specialist intervention [21]. This can be problematic for those being treated in the community, where access to clinical help can be difficult, especially under the continuing COVID-19 pandemic [22]. Connected health systems (mHealth) have long been heralded as a means for enhancing community care but, while there have been tremendous advances in some aspects of decentralised sensor technologies (i.e., vital signs [23,24]), there has been much slower progress in the commercial realisation of networked wound diagnostics. In this case, the underpinning community communication network is not the prime hurdle as current estimates suggest that 96% of households in the UK have some form of internet connection [25]. As the infrastructure for both rural and urban connectivity is clearly well developed, the main impediment to connected wound moisture monitoring lies with the sensors themselves. Attempts to develop a commercial wound sensor were pioneered by Milne and colleagues through their WoundSense system [19], but the physical bulk and need for manual interrogation are limiting factors in its adoption. A more recent prototype by Mehmood attempted to address these issues through a wearable format offering wireless reporting [26,27] but, again, miniaturisation of the device footprint and the economics of the device manufacture are significant detractors.

3. Design Requirements and Sensing Methodologies

Given the frequency of dressing changes and the fact that the sensing component would need to be replaced with each new dressing to prevent contamination, there is a need for devices that are small, inexpensive and mechanically flexible. The latter is particularly important to avoid the situation where the device itself aggravates the wound and impedes the healing processes. It would be necessary to separate the sensing (disposable) component and the electronics (monitoring) component, which would mirror the approach taken with home glucose measurements, where the disposable test strip is the primary consumable with a reusable meter. Various measurement methodologies have been employed and fall within the electrical, optical and mass detection divisions. In this communication, however, attention has been focused on the electrical and, in particular, those systems which are adaptable to mass manufacturing processes. The latter will be crucial when considering the journey from lab bench to commercial reality while acknowledging the fact that a wound sensor integrated within a dressing could easily have a lifetime of less than a day [18]. This aspect stands in marked contrast to domestic or industrial moisture/humidity sensing applications, where sensors may be expected to last for months (or years). Electrical systems have traditionally been dominated by resistance-, impedance- and capacitance-based devices, though there is increasing interest in radio frequency identification (RFID) systems based on inductive–capacitive coupling.

A brief inspection of commercial moisture/humidity sensors reveals that few would be acceptable for use in a wound management context as a consequence of their physical bulk and the inflexibility of the sensor. Two examples of low-cost moisture probes typically used in precision agriculture applications are highlighted in Figure 1. The sensing components are deposited as a patterned thin film but on rigid substrates. This is purely to comply with the demands of the agricultural applications—where perpendicular insertion into the soil surface is required.

Transferring the electrodes onto soft substrates that can be unobtrusively positioned within a dressing is clearly a critical step and, as such, there is a need for the sensing component to be immobilised onto thin flexible films. Devices from two distinct sensing directions, wearables and disposables, however, have led to material advances that could help meet the requirements of wound sensors. Some of these are highlighted in Table 1 and typically fall within the polymeric (PET, PI, PDMS) and cellulosic paper domains.

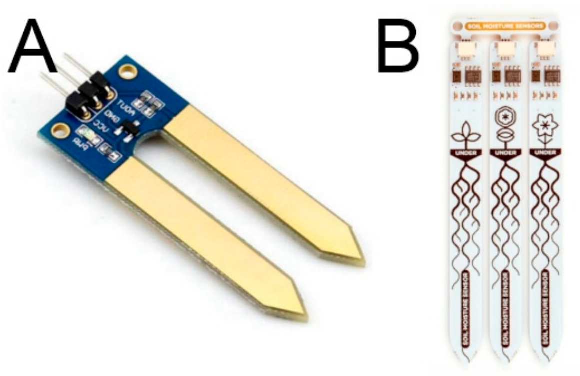

Figure 1. Planar soil moisture sensors based on resistance (**A**) and capacitance (**B**) measurement methodologies.

Table 1. Moisture sensing strategies based on interdigitated sensors deposited on thin flexible films.

Material	Method	Substrate	Ref
SWCNT/PVA	R	Fiber	[28]
PEDOT:PSS	I	Rayon/PET	[29]
Cu wire/PE/PI	C/LC	Fiber	[30]
Ag	R	PDMS	[31]
α-In2Se3 nanosheet	R/I	PET	[32]
Ag nanowire/graphene oxide	R	PDMS	[33]
PVA/graphene flowers	C/R/I	PET	[34]
Carbon nanocoil/nanotube	R	Paper	[35]
Graphene oxide/ZnO	R	Paper	[36]
Reduced graphene oxide	R	PI	[37]
MWCNT ink	C	Paper	[38]
Cellulose acetate butyrate	C	PET	[39]
Cellulose–Ag	LC	Paper	[40]
Ti/Au	C	PI/PET	[41]
Ti/Au	R	PI/PET	[42]
LIG–cellulose	R	Paper	[43]
LIG–cellulose	R	Paper	[44]
LIG/graphene oxide	C	PI/PET	[45]

Abbreviations: C = capacitance; I = impedance; R = resistance; LC = inductive–capacitive coupling; SWCNT = single-walled carbon nanotube; PVA = polyvinyl alcohol; PE = polyester; PEDOT:PSS = poly(3,4-ethylenedioxythiophene):polystyrene sulfonate; LIG = laser-induced graphene; PDMS = polydimethoxysiloxane; PI = polyimide; PET = polyethylene terephthalate.

A common design approach is to deposit a pair of interdigitated electrodes on the target substrate through either printing silver (screen or inkjet), evaporation of Ti/Au layers [41,42] or through the laser scribing of carbon tracks directly within the material [43–45], as indicated in Figure 2A. In some instances, the base substrate can facilitate ionic conduction between the electrodes upon interaction/adsorption of water and may be sufficient to enable the acquisition of a quantifiable signal and is common in the case of those employing cellulosic fibres [41–44]. The more common approach relies upon the deposition of a second, moisture-sensitive layer via drop casting, spraying, etc. [33,35,36,45]. A pertinent example is the work by Lan and colleagues (2020), where lasered graphene electrodes coated with graphene oxide were employed to facilitate measurement of stomatal moisture dynamics on leaves [45].

The detection mechanism will vary depending on the methodology being adopted (resistance, capacitance, etc.) and the material configuration, but the core requirement in each case will be the incorporation of a hygroscopic material that can interact with the moisture within the wound. As such, they rely on the water vapour undergoing chemical adsorption (chemisorption), physical adsorption (physisorption) and capillary condensation processes [46,47]. At low relative humidity (RH), chemisorption of the water occurs to the electrode material itself or the composite component (cellulose fibres, graphene oxide particles, etc.), as indicated in Figure 2B. Proton hopping via a hydronium ion (H_3O^+) serves as the main charge carrier, with conductivity increasing as subsequent layers of water are physisorbed. As the number of physisorbed layers of water increases, the interfacial layer becomes more liquid-like (capillary condensation), with conductivity greatly eased via the Grotthuss pathway ($H_2O + H_3O^+ \Leftrightarrow H_3O^+ + H_2O$) [46]. The adsorption of moisture can also markedly change the dielectric properties of the sensing layer, with the resulting change in capacitance facilitating another detection option.

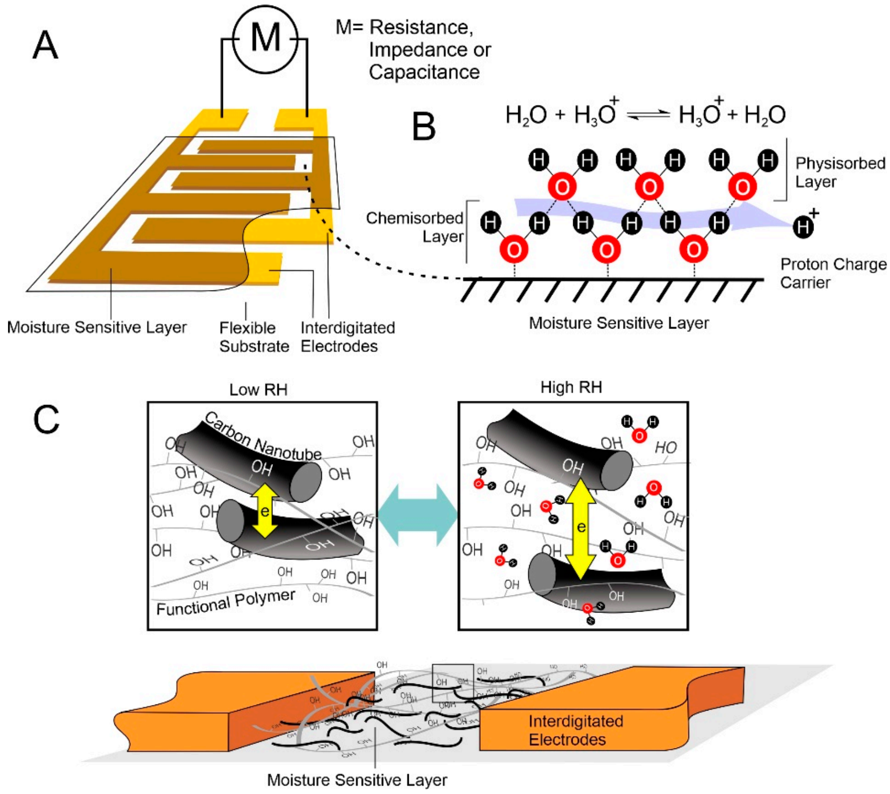

Figure 2. (**A**) Typical format of electrochemical humidity sensors and the resistive sensing methodologies associated with (**B**) ionic conduction and (**C**) physical changes brought about through swelling of the moisture sensitive film.

A second resistive approach can also be exploited where the analytical signal is influenced by physical changes in the moisture-sensitive layer. In this case, penetration of moisture within a film (PVA) or fibre (polyamide) laced with a conductive element (typically carbon nanotubes) leads to swelling of the former (Figure 2C) [28,48]. Conduction in such cases is typically though electron tunnelling between adjacent nanotubes and thus the swelling induces the displacement of the nanotubes, with the consequent increase in distance between the latter leading to an increase in resistance, which can be correlated to the moisture level.

As indicated in Table 1, a large variety of moisture-sensitive materials have been employed, but their application to wound monitoring contexts necessitates consideration of the wider design issues. A multilayer approach would be needed in which the sensing component is protected from direct contact with the wound surface and a secondary layer would be required to manage the release of exudate. A triple-layer "bandage" approach has been described by Tessarolo et al. (2021) in what is one of the few direct attempts to investigate moisture sensors within a wound environment [29]. In this case, the impedimetric sensor (PEDOT:PSS) was sandwiched between two passive gauze layers (wound contact; excess exudate capture). The sensor itself was printed on an "active" gauze layer, which served to manage the response to moisture. Critically, the team demonstrated that the distribution of the moisture across and within the active gauze layer differed depending on the composition (cotton, rayon or polyethylene terephthalate (PET)). As such, it could be envisaged that individual sensors could be optimised for particular wounds through judicious selection of the supporting textile substrate.

4. Laser-Induced Graphene (LIG)

The accessibility of LIG has increased dramatically in recent years through the increasing availability of consumer-orientated laser engravers—some costing less than GBP 100. Such systems can be used to pattern graphitic/graphene tracks on a range of substrates (polymers, wood, food, textiles) under ambient conditions, and, while they provide a simple means of rapid prototyping, the underpinning methodology is scalable and could be readily adapted for volume production either on a batch basis or through roll-to-roll processing [49–51].

The laser-induced photothermal conversion of the film results in a micro-nanoporous foam, as indicated in the electron micrographs detailed in Figure 3. The localised heat leads to degradation of the polymer (polyimide or cellulosic materials), resulting in the release of gas. This initially causes the polymer film to foam, with the subsequent cooling yielding raised carbonised structures. There is considerable morphological heterogeneity and this tends to be highly dependent on the laser configuration. The electron micrographs in Figure 3 detail the LIG features arising from two different laser settings: 7.5 W and 12.5 W [52]. It is clear that the resulting LIG structures possess a huge surface area, which is particularly suited to sensors employing a capacitive methodology [53–57].

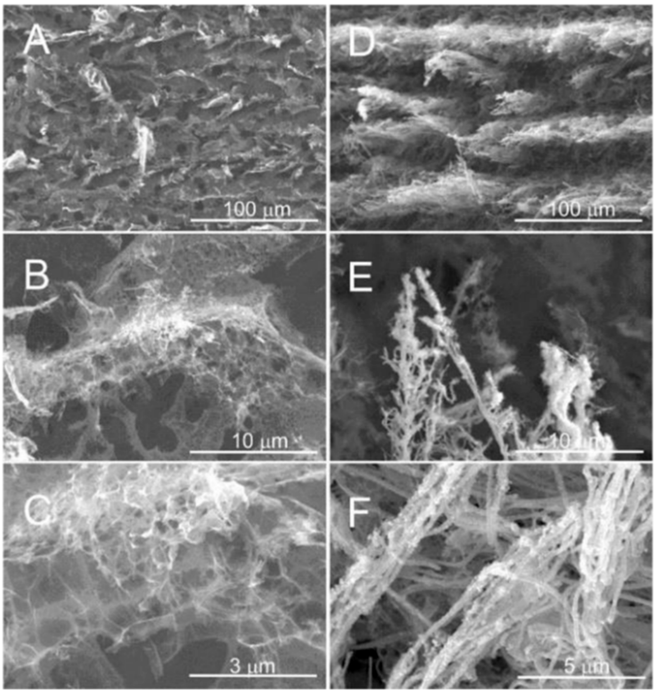

Figure 3. Scanning electron micrograph images of lasered polyimide tracks produced at a power of 7.5 W (**A**–**C**) and 12.5 W (**D**–**F**). Reproduced with permission [52].

The mechanical flexibility of the LIG-based sensors is widely touted as a major feature and, while this would certainly be advantageous when attempting translation to wound management, there are several important caveats to reconcile. It is necessary to note that simple bending of the film will lead to changes in resistance—a feature that has led to its use in strain sensing [13,58,59], but which can be considered a limitation in chemical sensing—especially in regard to those systems relying on resistive methodologies, where subtle changes in shape could lead to ambiguous responses.

In the case of the leaf-based measurements, it could be anticipated that the sensor would be applied to a flat portion of a leaf, but this option may not be available (or appropriate) in the context of wound management, where the position and morphology of the wound can be extremely variable. A prospective sensor placed on the leg (anterior aspect of the tibia) will experience markedly different forces upon movement than a similar sensor placed on the plantar aspect of the foot, which will endure pressure and shear. It could be envisaged that the borders of the foot, in particular, will experience pressure from footwear and shear from movement within a shoe/sock. While LIG scribed directly onto a substrate can undergo bending motion, stretching is much more problematic—with physical extension leading to fragmentation and circuit breaks. There have been attempts to address such limitations with serpentine designs or the transfer of the LIG from conventional PI to elastomeric substrates such as silicone or polyurethane [60–62]. It is important to note that the actual size of the sensing component itself can be very small (in the order of millimetres) and it could be envisaged that, when placed within a dressing, this component could remain relatively unperturbed by motion in the underlying skin—it is the track connections to the external monitoring device that are liable to be affected. If these could be removed, then the issue of flexibility/stretchability may be redundant.

5. Radio Frequency Identification (RFID)

The introduction of radio frequency identification (RFID) technology—such as that used on packaging/security labels—partly addresses this issue, allowing the sensor to be read wirelessly without the need for the controlling circuitry to be located next to the wound [63–65]. The latter is often neglected in the literature, where the focus is often on the bioanalytical responses for the sensing component rather than the electronics and power source that must accompany it. On one level, RFID systems could be considered to be relatively simple in terms of composition and placement (at least relative to wired sensors) but there are substantial design and material challenges that need to be addressed in order to facilitate the retrieval of real-time wound data. The conceptual system would be composed of a tag (comprising an integrated circuit and antenna), which is placed within the wound dressing, with the information held within the tag read wirelessly by a reader (mounted or handheld). The basic approach is highlighted in Figure 4 and it is clear that, while the controlling chip may be tiny, the key component, from a clinical perspective, will be the design of the antenna as it will dictate the overall dimensions.

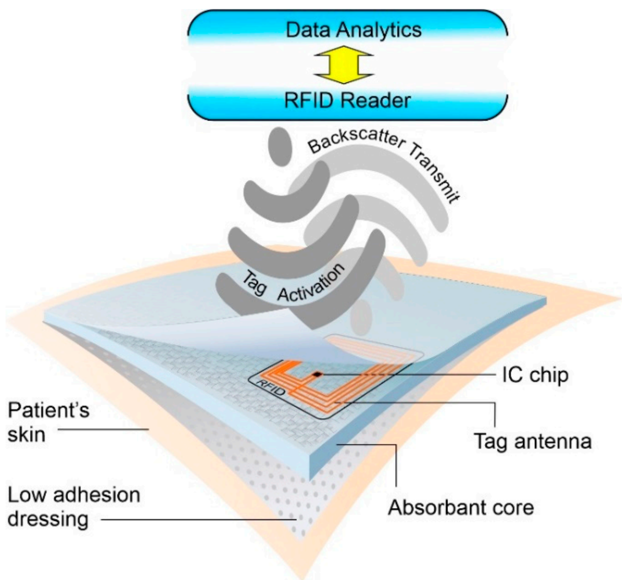

Figure 4. Conceptual implementation of a radio frequency identification (RFID) sensor within a wound dressing.

In contrast to the wired sensing systems, the RFID chips can take their power directly from the reader device. In its simplest iteration, the reader transmits an appropriate electromagnetic wave that is received by the tag antenna, which subsequently converts it to dc power and activates the chip [66]. The chip transmits the stored data as a modulated (encoded) signal using the antenna, which is then received and decoded by the reader to obtain the information from the tag. While such devices are common to modern business—enhancing logistics and tracking—it is easy to envisage how this could be adapted to clinical environs and patient care, where the information is a unique patient identifier [67,68]. While their adoption could be considered as a more robust form of barcoding or QR coding, careful manipulation of the tag components and antenna design can open up possibilities for the tag to go beyond simple data storage, and there has been considerable interest in their adaptation to the measurement of temperature and moisture, which would clearly have direct application to wound management [64,65,69–71].

The removal of ancillary electronics and power cells greatly reduces the dimensions and cost of the individual devices. The integrated circuit within the chip is the principal cost element and, for passive systems, can be of the order of several euro cents, which, even with a high frequency of dressing change, could be economically acceptable given the potential savings in nursing time. Nevertheless, there has been extensive interest in the development of chipless tags [72–74]. The latter can offer much simpler (and scalable) fabrication processes, which can significantly reduce costs. However, such systems have lower data storage capabilities but, critically, their sensing capabilities are still available despite the loss of the IC chip component. The nature of the antenna materials that comprise the antenna can be adapted such that their properties change in response to environmental stimuli (i.e., change in dielectric constant as a consequence of moisture adsorption) and thereby alter the resonant frequency of the tag [63,71]. It is here that LIG again has been found to have considerable impact through being able to rapidly prototype antenna designs, and it is perhaps little surprise that the next generation of chipless RFID tags with sensing capabilities are LIG-based [53].

The RFID tag can be read at a distance of up to several metres without the need for line of sight, which is critical given that the tag can be buried within the various layers of dressing. A common misconception is that the tag must be manually scanned by a person yielding a reader, but precision agriculture has demonstrated the capability of interrogating RFID tags at a distance and transmitting the data (via LoRaWAN) to the cloud [75]. It could be envisaged that a reader could be positioned on a bed for autonomous processing for those who are immobile or capable of being positioned within a room where the patient is liable to spend a significant period of time. The main cost is attributed to the reader, but this would be re-usable/transferrable from patient to patient and again would mirror current approaches to 24–48 hr vital signs monitoring, where the device uses replaceable electrodes but the sensing unit is returned to the clinical practice once complete. The pursuit of RFID humidity technology is clearly a promising pathway, with a concerted move away from the bulk of hardwired systems to the much more patient-acceptable wireless format [29,30].

6. Conclusions

The management of chronic wounds can require daily care, which can place considerable demands on community care nursing provision. This can be especially problematic given the increasing complexities borne by the COVID-19 pandemic and where it is widely recognised that there have been substantial reductions in nurses working within the community. There is a place for technology to assist in the prioritisation of visits and it could be envisaged that a simple monitor to periodically monitor strikethrough in moderately/highly exuding wounds could significantly improve management and provide more consistency in care. The technology is clearly available, as witnessed by the ever-expanding analytics base used in modern industry and, as hopefully indicated herein, the translation of the technology to community wound care is not unrealistic. The advances in laser graphene methods for fast tracking the development of sensors and the emergence of chipless tags that can be read wirelessly should be capable of impacting daily wound management practices.

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