A Physical Unclonable Function Based on Recyclable Polymer Nanoparticles for Enabling the Circular Economy

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Abstract

The circular economy requires that we know what a product is make of, that we can control that it is not a fake, and that we know how to recycle it. This requires a digital twin of each product, where the information is stored that we can access using a smartphone. Creating digital twins require connecting the physical and digital worlds, which in turn requires that all objects have a unique identifier that is valid in both domains. Realizing the intrinsic potential of creating digital twins of individual products, of linking physical and digital identities requires that the physical identifier is truly unique, and that it cannot be copied. Furthermore, the physical identifier should not interfere with the product lifecycle, in particular recyclability, as this would be a potential blocker in a circular economy. Here, we present a physical identifier based on a physical unclonable function (PUF) made from polymer nanoparticles and imbedded in a polymer laminate, where all components can be made from the same recyclable material. Such as materials commonly used for packaging in the fragrance and cosmetics industry. The development of the PUF system is presented, and each PUF was validated by using a proprietary smartphone application with a QR code as a common pattern defining PUF canvas. The functionality of the PUF was demonstrated across product types as diverse as lateral flow assays and construction materials. We conclude that polymer nanoparticle PUFs are a viable and directly recyclable solution for documenting authenticity and inform recyclability of polymers in the circular economy.

Introduction

The approach to providing a digital ID for consumer goods that we present here is equivalent to creating 'biometrics for things'. A direct link between a digital ID and a secure physical ID allows for an irrefutable link between the physical and digital world, enabling the creation of genuine digital twins for all consumer products. This pairing is presented as part of an integrated solution for object validation, authentication, and identification, where the immediate application lies as a solution to the problem of counterfeit goods.[1-4] Combatting counterfeiting is an old issue,[5] but it is increasingly important due to the sheer scale of the problem,[6] and the detrimental consequences to general human health and safety.[7-11] But an irrefutable optical authentication system directly available on portable devices allows for much more disruptive solution, from smart contracts bound to the physical object to direct transfer of ownership between consumer wallets.[12-15] Here we propose that secure physical IDs is also critical for realizing a circular economy.

Our proposed solution for a unique physical identifier is to use physical unclonable functions or PUFs.[4,16,17] PUFs are unique physical objects that contain vast amounts of information that can be recorded and compared.[18-23] Examples range from human fingerprints to the scattering of the random surface structure of paper.[22,24-34] While using PUFs is just emerging in optical authentication of goods, their application has been investigated in detail in electronics.[19,21,24,35-39] A solution that is to be implemented as an authentication and identification system for consumer goods requires that each PUF is cheap (far less than 0.10 USD per physical identifier), and if the solution is to be used as a driver of consumer behavior in a circular economy, the PUF must be an integral part of the product packaging to the extent that it is recycled with the product.[40] Plastics are the main material used for containers and packaging because they are economical, functional and lightweight. The most commonly used plastic materials are polyethylene (PE), polypropylene (PP), polystyrene (PS), polyesters (such as PET), polyamides (PA), and polyvinyl chloride (PVC).

The development of recyclable PUFs is a major challenge that can be overcome using advanced multifunctional polymer chemistry supported by nanotechnology. [41-43] In this paper we focus on polyesters used in many fields of industry such as beverage bottles, food packaging, pharmaceutical containers and cosmetic packaging.[44] We present the use of polyester polymer nanoparticles to design novel PUFs based optical identification system aimed to enable the circular economy.

Figure 1 shows the functionality enabled by the direct link between the physical product and a digital twin a PUF creates when considering the circular economy. The full carbon cost is imprinted on the digital twin in the production and can be documented with the PUF along the product life cycle all the way to product recycling, where the carbon-ledger is sealed with proper disposal.[45] For the consumer the PUF acts as a contract with the manufacturer, both documenting authenticity and environmental impact of the product. In the potential second life of the product, irrefutable provenance is added to the digital twin as part of the PUF. At end of life, the PUF can be coupled to correct recycling and thus mediate both corporate and individual carbon footprints. Many waste streams are readily sorted by the consumer, plastics—in particular on esthetic products—are not as readily recognized. As the current linear plastic economy is not sustainable,[46-49] and even though we are improving,[50] the complexity of polymer products hampers further progress.[46,50] One route to a circular plastic economy is new chemistry,[48,49] our suggestion is to use nanotechnology to enable—and maybe reward—consumer to perform the required manual sorting. Here, we propose an ecosystem for the brand and the user in which a PUF enables incentivize the recycling of packaging and at the same time t guarantee that the product is real. Incentivizing sorting/recycling is known e.g., beverage containers carry a fee that is returned when the container is recycled. We add to the general advantages of PUF marked products, by demonstrating a fully recycle PUFs and suggesting that the PUF is used to incentivize recycling.

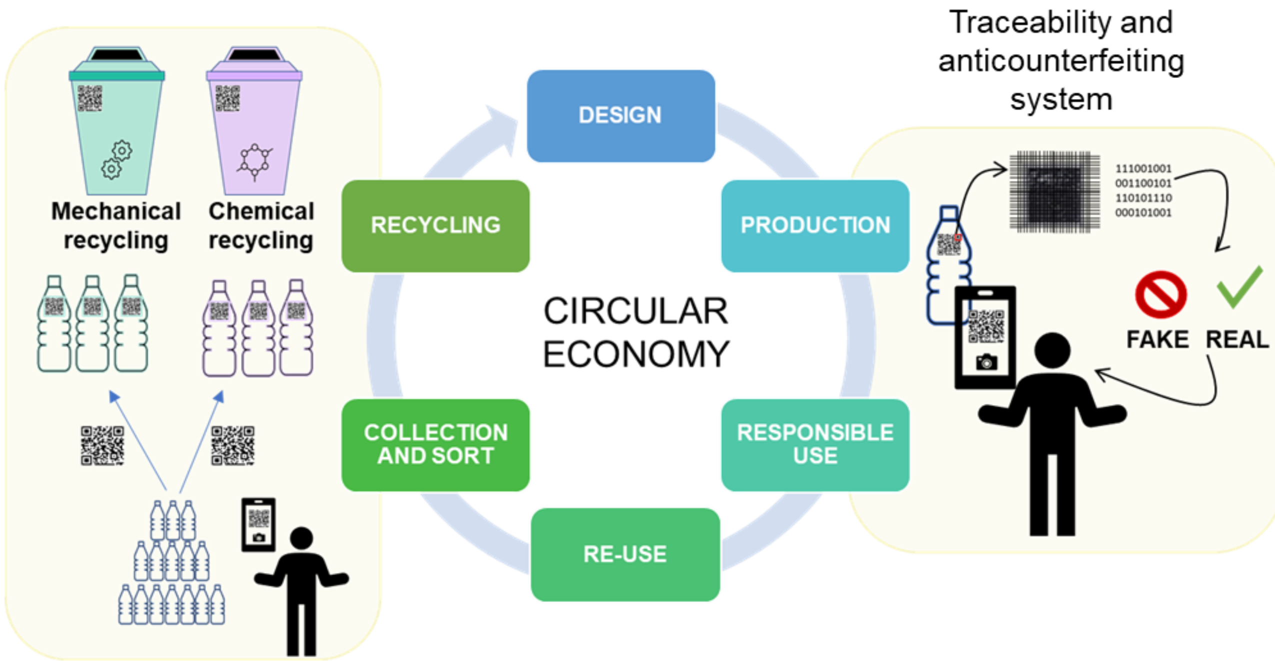

Figure 1. *The need for individual object authentication in the circular economy, the steps are: 1) the design includes PUF placement and required PUF functionality. 2) the production incorporates PUF patterns in (parts) of the product and define PUF canvas via printing or product structure, product carbon ledger and product information is burned into the digital twin of each product. 3) Product authenticity, care guides and carbon cost ensure responsible use. 4) Re-use and secondary use of products are ensured as ownership and carbon ledger can be transferred to other parties via the PUF ID. 5) Sorting by the end-user can be incentivized and quality controlled via the PUF on each product and e.g. a PUF on the recycling containers. 6) Recycling is readily done with only one materials in each recycling stream.*

Experimental procedures

The ink used for drop casting or knife coating was created by suspending polyester nanoparticles synthesized according to our previous work.[51] Briefly, the nanoparticles were synthesized by nanoprecipitation. First, polyester pellets were dissolved in DMSO at high temperature. Then, the solution was added drop wise to a poor solvent under vigorous stirring. The nanoparticles were isolated as the precipitate were collected by filtration, washed with methanol, and dried Figure 2 show the patented process and examples. The full details are included in the reference 52. Table 1 includes the matrix of experimental parameters tested in order to prepare a PUF ink based on the patented nanoparticles. To produce the all-polymer PUFs, two PUF inks was created: the nanoparticles were either suspended at 1% w/w in EtOH or in PVA/water (10 % w/w Sigma Aldrich).

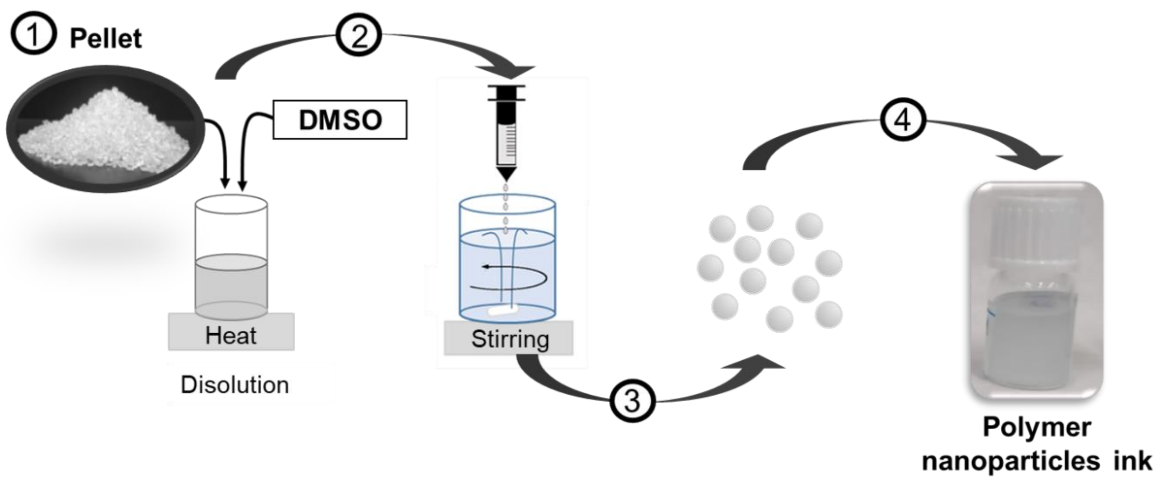

Figure 2. *A) Synthesis of the polymer nanoparticles: Pellets were dissolved in DMSO at high temperature (1). Once dissolved, is the DMSO solution was added dropwise in poor solvent with stirring (2). The formed mixture was filtered, and the isolated precipitate was washed and dried and the polymer nanoparticles was obtained (3). Finally, is the nanoparticles were dispersed to create the PUF ink (4).*[52]

Table 1. *Experimental parameters tested for the creation of a PUF ink*

Nanoparticles used	Solvents Tested	Polyester starting Concentration (g/l)
Nanoparticles formed by precipitates stored in metanol suspension	Methanol	0.1 0.5 1
	Ethanol	
	Petrol ether (60-80)	
	Diethyl eter	
	n-pentane	
	PVA	

QR codes were printed on different substrates such as recycled paper, polyethylene terephthalate (PET), low-density polyethylene (LDPE) using a conventional 1200 dpi laser printer.

Different multilayer laminates of PET (Optically Clear Adhesive Tape LUCIACS® CS986 Series), LDPE (ISTAD, resealable bag) and polypropylene (PP, transparent folders for A4) were created by depositing nanoparticles within the layers. The deposition method was either manual drop casting or knife coating. Table 2 show the different combination of samples created by drop casting or knife coating of the two inks on the four substrates.

Table 2. *Combinations of materials used to make all-polymer PUFs by applying a PUF ink on a substrata using different deposition methods*

Substrate		PET	LDPE	PP	Paper
PUF ink	Particles	Polyester			
	Carrier liquid	PVA or Methanol			
Deposition method		Knife coating or Drop Casting			
Contrast type		Scattering			

Procedure used to fabricate 1st generation all-polymer PUFs

Drop casting was performed on different thin films of commercial polymers such as PET, PE and PP. For this purpose, solutions of the polymeric nanoparticles were prepared in ethanol at 0.5 g/l. The suspensions were homogenized by sonication at room temperature. Afterwards depositing the ethanol drop on the film, the substrate was dried at ambient condition, care was taken to ensure complete evaporation of the solvent. Then, the assembly was carried out by joining both layers of a laminate, with the help of a simple glue. Thus, multilayer films were obtained with the nanoparticles fixed inside.

Procedure used to fabricate 2nd generation all-polymer PUFs

For the second generation of PUFs, we decided to increase the concentration of the polymer in the ink to 1 g/l in order to obtain better random patterns. Furthermore, the QR codes were printed directly on a PET film, and then the nanoparticles were deposited by drop casting directly on the PUF canvas of the QR code (Figure 3). After deposition, the solvent was allowed to evaporate, and the sandwich morphology was assembled using glue, thereby creating multilayer films with PUFs on the printed QR code in the form of polymer-embedded nanoparticles.

PUF Evaluation

Each fabricated PUF was first evaluated by visual inspection (apparent randomness) and then by testing using the Pufin ID smartphone application. The app operates on a neural network trained on the QR based PUF we previously reported,[17] and in developer mode the app allows for registration of new PUFs as well as validation of exciting PUFs. Further, the developer mode reports a match score derived from an undisclosed distance measure in the embedding space scaled by the thresholds set for this particular PUF system. The neural network was trained on this match score is 99+% for validation of PUF against the same PUF, and the match score is >5% for validation of a PUF against a different PUF. The app only reports the match score for the five PUF in the embedding space most similar to the PUF being validated. When a PUF falls outside of the five most similar PUFs in a validation, we report a match score of zero.

The commercial version of the application can be downloaded for Android and iOS in the Google Play store and the Apple App store.

Results and Discussion

The design concept for the recyclable optical physical unclonable function is shown in Figure 3. There are two significant differences between this design concept and previous reports of optical PUFs:[4,17,40] 1) the PUF is created as an integral part of the polymer laminate, and 2) high contrast reflective or luminescent materials are not used to create the random pattern. In particular, the latter was initially of great concern.

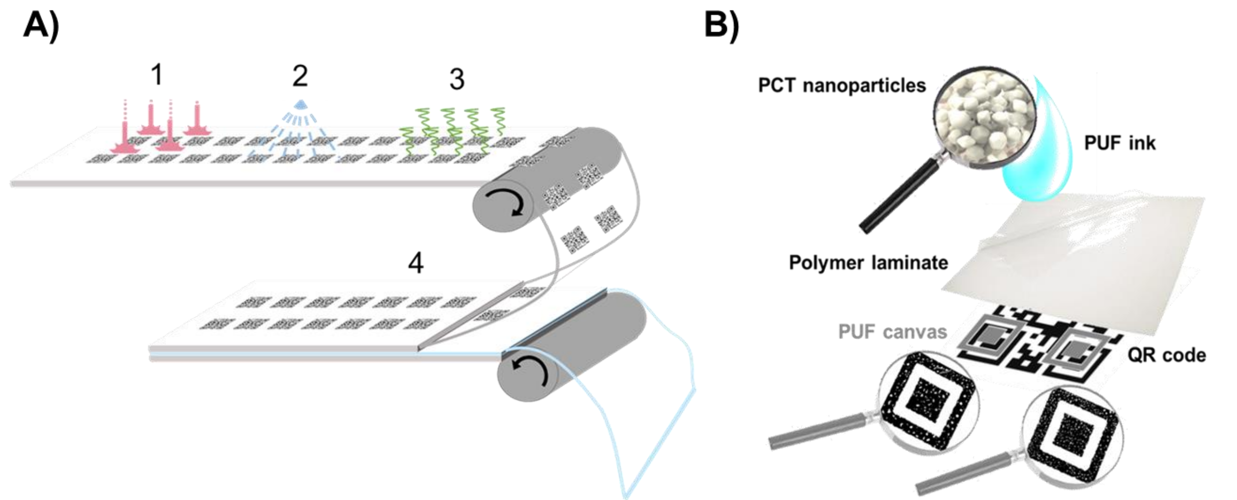

Figure 3. *Design concept for all polymer PUFs. A. The steps to follow to create all polymer-PUFs: Printing of the QR codes on the different substrates (1), drop-casting deposition of the nanoparticles (2), evaporation of the solvent (3), adhesion of both layers of laminates. B. The nanoparticle containing PUF ink is deposited in a polymer laminate on a QR code printed on a separate sheet of paper or the polymer bottom layer of the laminate. The corners of the QR code acts as PUF canvas, defining the regions where the PUFs—the random patters formed by the nanoparticles—are read.*

Figure 3 shows how the PUF is made, and how the area containing the stochastic pattern is defined as a PUF canvas. Here, the PUF canvas is the corners of a QR code, where the black square is used to define the area of the random pattern that constitute the PUF. A PUF is in principle any random pattern, but to be operational it has to be read and associated to a unique ID. Used in an authentication system each PUF should only associate to one ID, and all others will give rise to a different ID. If a PUF is not registered, no ID will be returned. The algorithm used in this work validates all three corners of the QR code in parallel. In this manner, each QR code contains three independent PUFs.

An all-polymer PUF

The nanoparticles used were synthesized according to a patented procedure.[49] These polyester nanoparticles show a spherical morphology with sizes between 160-200 nm as studied by SEM. However, it was demonstrated by DLS that when in solution, they agglomerate, reaching micrometer sizes, see also **Figure 4**. Thus, visible light scattering can be obtained, yet presumably with an efficiency inferior to common opacifiers like titanium dioxide and zirconia. Therefore, careful control of the agglomerate size must be achieved to the contrast needed to form an all-polymer PUF. By experimenting with various alcohols, pure solvents, mixtures and poly-ols as aqueous solution, and by changing the concentration of nanoparticles the optimal preparations of the PUF ink was discovered. The goal of maximum contrast and sufficient randomness in the patterns formed was achieved by drop casting from a suspension of nanoparticles in pure ethanol. To demonstrate the feasibility of the concept we first created physical unclonable functions by drop casting on PET, LDPE and PP films, which we placed on top of a QR code printed on paper. The first generation PUFs are shown in **Figure 4**.

All the four PUFs in **Figure 4** can be registered in the system and validate correctly using the Pufin ID smartphone application. While the true positive match scores lie in the 99 % range, the true negative significantly exceeds the 5% that is indicative of a good PUF design for this particular neural network. Rather than training a new neural network, which is labor intensive, we decided to investigate and optimize the PUF fabrications.

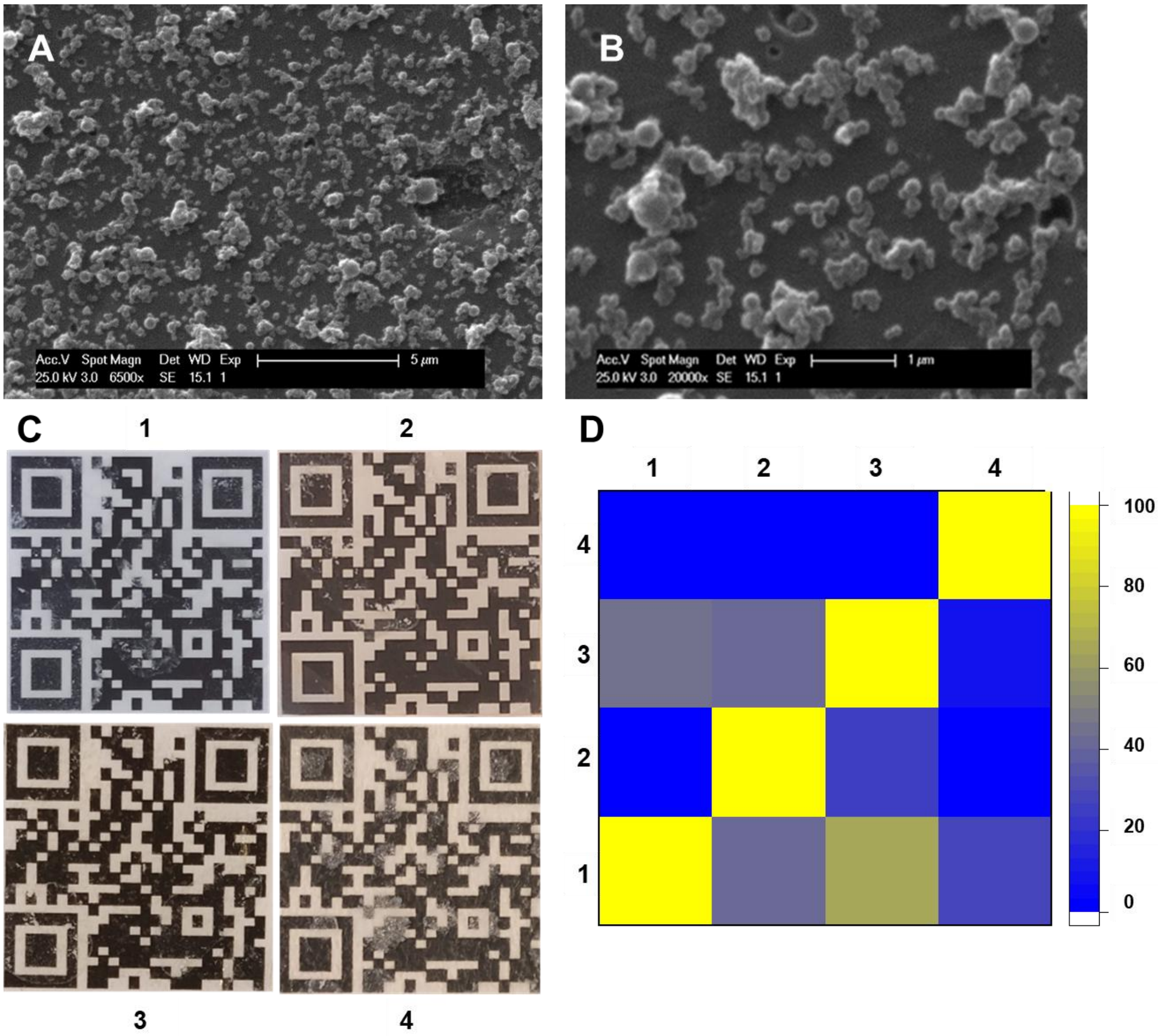

Figure 4. A. *and B. SEM image of 0.5g/l PCT/EtOH nanoparticles over PET-film. C. The four first-generation PUFs created on QR codes printed on paper. The polymer laminates are made from PET(1), LDPE(2), PP(3,4). D. The match score (0-100 %) matrix using the Pufin ID app to first register and then validate these four PUFs*.

Figure 5 shows 120 2nd generation all-polymer PUFs that was created by increasing the nanoparticle concentration and tweaking the drop casting procedure in order to ensure that significant contrast was achieved on all PUF canvas. In this fabrication, the PUF canvas was created by printing the QR code directly on the polymer laminate.

Four of these PUFs were selected and registered in the authentication system. Subsequently, from the remaining 116, 40 QR codes were chosen at random and validated to ensure that none were recognized as the same as those previously registered in the PUF database. The result of this experiment was evaluated using the match score reported by the Pufin ID app, where the match score of the true positive match and the match score for the closest five true negative matches are reported. All the match scores were recorded and compared. The result of this analysis is shown in Figure 6.

Cursory inspection of Figure 6 reveals that this second generation of fully recyclable all-polymer PUFs still show inferior performance using the smartphone application, when compared to the titanium dioxide based PUFs.[16,17] It should be noted that the smartphone application runs a neural network trained on PUFs made on paper using highly scattering titanium or zinc oxide.[16,17] Thus, there are two possible reasons why the all-polymer PUF have inferior performances: 1) the PUFs created may be too similar that is the encoding capacity is too low,[16] or 2) the neural network does not pick up the randomness in the all-polymer PUFs as it too different from the samples it was trained on.

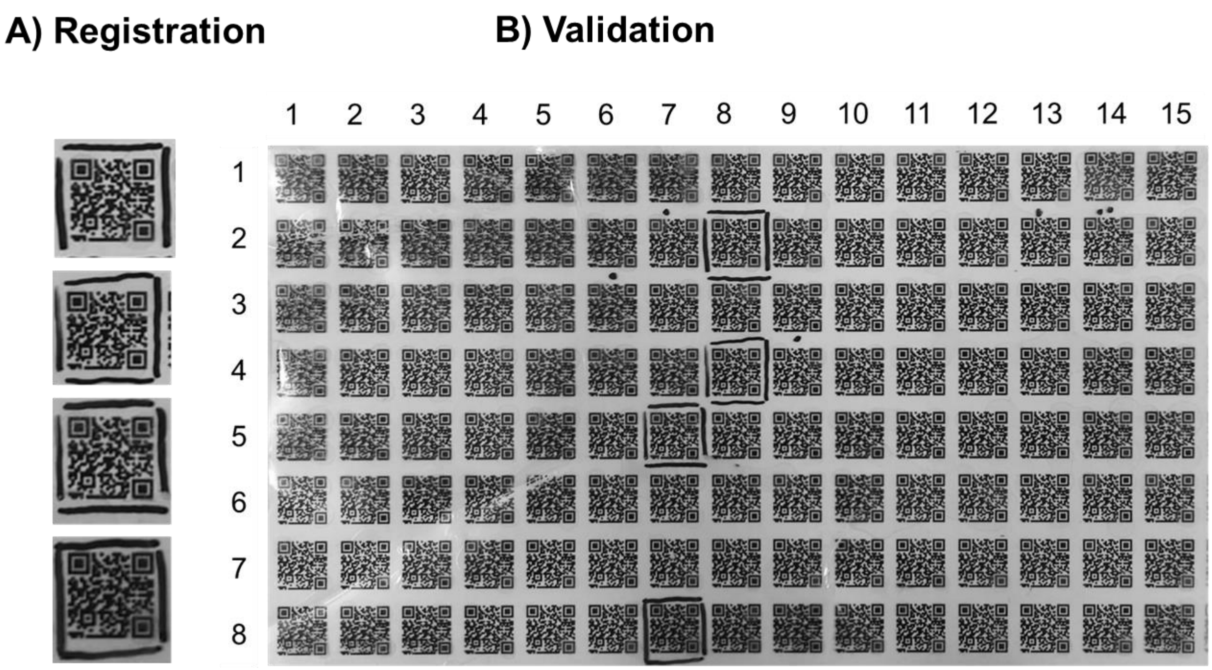

Figure 5. *120 QR codes printed on PET film (Optically Clear Adhesive Tape LUCIACS® CS986 Series) on which the nanoparticles have been deposited (NP 1% w/w in EtOH), creating 120 new PUFs. A. 4 QR codes are selected and registered in the authentication system. B. The 4 QR codes are validated against 40 different QR codes*.

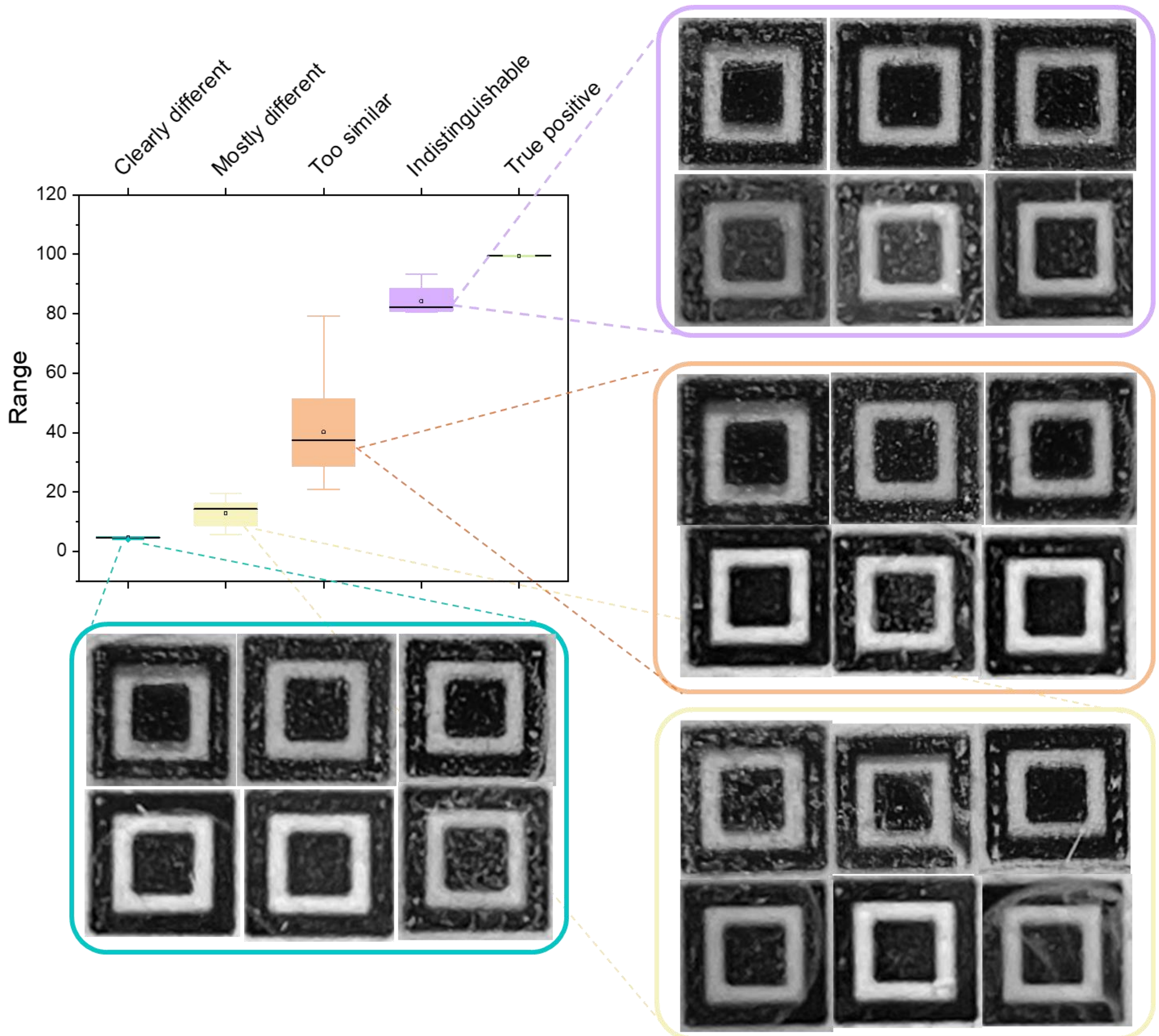

Figure 6. *Box plot showing the result of validating 40 all-polymer PUFs and representative PUF canvas from each group of match scores. The box plot show match scores grouped by criteria taken from the distance measure in the embedding space of the neural network, each is represented by the actual pictures of PUFs giving rise to this match score. Note that the human eye would not assign any two PUFs shown as identical.*

To visualize the origin of the poor performance we included the images of groups of PUFs that give rise to poor match scores in **Figure 6**. It is worth noting that the neural network only performs a task of comparison similar to what we are capable of using our own eyes. The neural network has to be trained, and is parameterized based on this training data. Therefore, it can only operate perfectly on similar samples. The question of poor performance of the all-polymer PUF using the Pufin ID optical authentication system can thus be decided by visual inspection of the PUFs. Considering the PUFs shown in **Figure 6** they all are clearly random and contain significant amounts of information.[16,17] Therefore, the failure is not in the PUF. This conclusion is further supported by the fact that the comparison 8/7 and 2/8 by human eye shows that each PUF is clearly unique and readily differentiated from the others. This is the case for all the 120 PUFs produced. There are two routes to a commercial grade optical authentication system from this point: 1) optimizing the means of fabrication to eliminate the 'too similar' and 'indistinguishable' groups in Figure 5, or 2) creating 10.000 PUFs and re-training the neural network.

Practical application of an all-polymer PUF

The reason for moving from the oxide containing PUF system to an all-polymer PUF is that it can either be grafted on polymer products or be made an integral part of the product. An example: In the cosmetics and fragrance sectors, there are currently a wide variety of packaging available. The most commonly used containers are of the aromatic polyester family due to their high chemical and mechanical resistance. These materials allow for containers of different shapes and finishes. In order to protect these containers from counterfeiting, and to provide traceability of the product, polymeric nanoparticles of the same polymer as the containers can be used by incorporating them into the container. To do this, the multilayer technique is used. To implement a PUF technology, sheets and nanoparticles of the same polymer are used with an adhesive binder, thus creating a multilayer film. This film is then formed and welded to create the packaging, which is finished by printing and coating. The latter processes are as important as the former, as these will form the canvas of the PUF. The particles in the laminate carries the random pattern, the printed PUF canvas defines where it is read. The advantage of this implementation (approach) is that the PUF particles can be all over the packaging, can be used for product traceability, and can even inform about recyclability and thereby facilitate recycling. No additives, fluorescent dyes, fillers, oxides or any material that differ from the composition of the packaging have to be used. All is made from just a single polymer.

To illustrate the point, **Figure 7** shows four examples of PUFs grafted onto polymer products with existing QR codes. Each PUF was registered in the optical authentication system and can be validated with a return of a unique ID for each product. We consider this proof of concept, and it remains to the manufacturers to decide how to implement this unique ID in their Environmental, Societal, Governance (ESG) structure. We believe it is the only way to keep a true carbon-ledger. The PUF provides a unique ID for the physical product, but all advanced functionality relies on the digital platform that build on the digital ID, which in turn is linked to the physical ID. The investment in this digital platform may very well exceed the investment in production equipment needed to move from the demonstration in Figure 7 to mass produced PUFs.

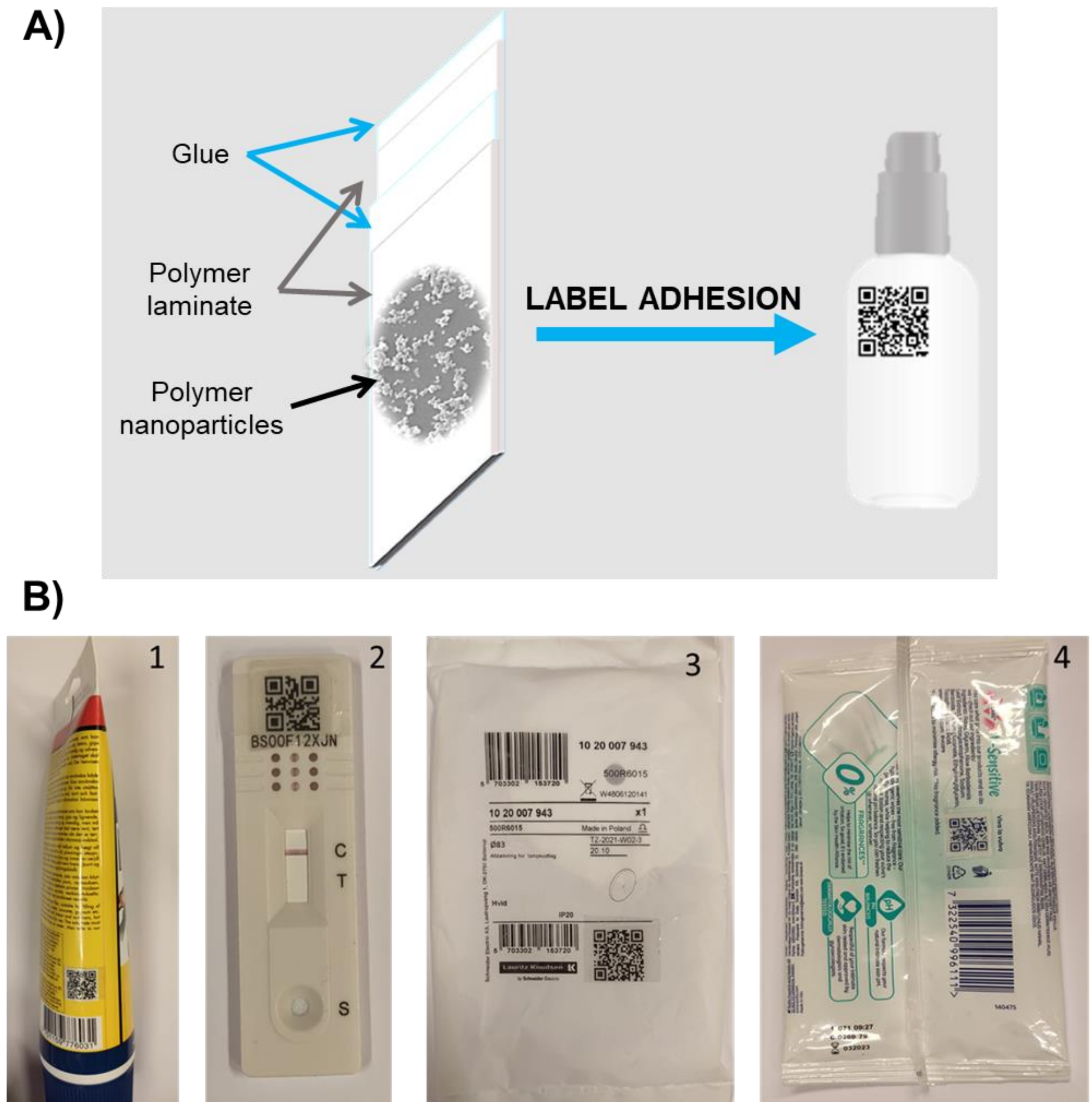

Figure 7. *A. Adhesion of the all-polymer PUF label to commercial packaging already bearing a QR code. B. Commercial products examples made with different polymers (plastics) that have QR codes for proof of concept. DANA LIM filler extra 619 250 ml (1), Antigen test (2), Electronic product packaging from Lauritz Knudsen by Schneider Electrics (3) and Libresse product (4).*

Conclusions

We have shown that by using agglomerated nanoparticles it is possible to create an all-polymer physical unclonable function or PUF. Thus, we can use the same material (polymer) as the packaging to provide security and traceability to different products. In addition, the PUF facilitates the recycling since it provides information on how to recycle it, and the product can be recycled in its entirety, as it does not contain any additives.

We used the polymer nanoparticles to create random patterns on different polymer substrates. QR codes were used to define areas of the random patterns, creating distinct smartphone-readable PUFs. We demonstrated that the all-polymer PUF can be used in a smartphone based authentication system, creating a unique physical ID for products. The true potential of these physical IDs lies in linking them to a digital ID, creating digital twins, which in the right framework removes all barriers for tracking of individual products. While the transition to a circular economy still relies on human behavior, it can be enabled through personal and corporate carbon ledgers documented by PUFs.

As a transformative technology, the ability to link physical product to digital twins requires digital maturity of the producing corporation, and the actual ability to produce the PUF enabled physical product. We all have smartphones, so from the end user perspective we are ready. The natural next steps in commercializing PUF technology lies in integration in the industrial supply chain. Taking producers of products like perfume and cosmetics as an example. Step 1 is that PUF nanoparticles must be integrated into raw materials used to form the containers. Upon quality control using vision systems (Step 2), the material is formed into containers. These are coated, printing added to define the PUF canvas (Step 3), and the container is finished. The AI must be re-trained on the specific PUF design (Step 4). At the perfume and cosmetics supplier, the PUF on the container is registered using a dedicated vision system prior to filling (Step 5), and after filling the PUF is activated using a second vision system (Step 6) and a dedicated local server (Step 7) interfaced with cloud based software (Step 8). The activation of the PUF creates a digital twin of the product, and the physical product can now be authenticated. A commercial PUF systems will require significant engineering and software development, but no further research. Using the PUF based digital twin as outlined here, requires further development of carbon ledgers, and enterprise software solutions for recycling carbon accounting.

ASSOCIATED CONTENT

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Notes

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TOC

All-polymer physical unclonable functions (PUFs) can be made using polyester nanoparticles. PUFs on polyester packaging creating a recyclable, smartphone readable unique identifier on every product. We suggest that the unique ID is used to inform and incentivize recycling as part of the circular economy.