



## King's Research Portal

DOI:

[10.1016/j.trac.2016.08.003](https://doi.org/10.1016/j.trac.2016.08.003)

*Document Version*

Peer reviewed version

[Link to publication record in King's Research Portal](#)

*Citation for published version (APA):*

Gooch, J., Daniel, B., Abbate, V., & Frascione, N. (2016). Taggant materials in forensic science: A review. *TRENDS IN ANALYTICAL CHEMISTRY*, 83(B), 49–54. <https://doi.org/10.1016/j.trac.2016.08.003>

### **Citing this paper**

Please note that where the full-text provided on King's Research Portal is the Author Accepted Manuscript or Post-Print version this may differ from the final Published version. If citing, it is advised that you check and use the publisher's definitive version for pagination, volume/issue, and date of publication details. And where the final published version is provided on the Research Portal, if citing you are again advised to check the publisher's website for any subsequent corrections.

### **General rights**

Copyright and moral rights for the publications made accessible in the Research Portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognize and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the Research Portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the Research Portal

### **Take down policy**

If you believe that this document breaches copyright please contact [librarypure@kcl.ac.uk](mailto:librarypure@kcl.ac.uk) providing details, and we will remove access to the work immediately and investigate your claim.

# Accepted Manuscript

Title: Taggant materials in forensic science: A review

Author: James Gooch, Barbara Daniel, Vincenzo Abbate, Nunzianda Frascione

PII: S0165-9936(16)30173-X

DOI: <http://dx.doi.org/doi: 10.1016/j.trac.2016.08.003>

Reference: TRAC 14808

To appear in: *Trends in Analytical Chemistry*



Please cite this article as: James Gooch, Barbara Daniel, Vincenzo Abbate, Nunzianda Frascione, Taggant materials in forensic science: A review, *Trends in Analytical Chemistry* (2016), <http://dx.doi.org/doi: 10.1016/j.trac.2016.08.003>.

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

# Taggant materials in forensic science: A review

James Gooch <sup>a</sup>, Barbara Daniel <sup>a</sup>, Vincenzo Abbate <sup>b</sup>, and Nunzianda Frascione <sup>a,\*</sup>

<sup>a</sup> Analytical and Environmental Sciences Division, King's College London, 150 Stamford Street, London, U.K., SE1 9NH

<sup>b</sup> Institute of Pharmaceutical Science, King's College London, 150 Stamford Street, London, U.K., SE1 9NH

\* Corresponding author. Tel.: +44 20 7848 4978

E-mail address: nunzianda.frascione@kcl.ac.uk (N. Frascione).

## Highlights

- The use of forensic taggant technologies has become considerably more widespread
- The application of taggant materials for forensic purposes are described
- The different coding methods used in commercial marking materials to infer identity are explored
- Recent developments in the field are discussed along with future perspectives

## Abstract

The use of taggant technology to mark objects for the purpose of identification is becoming a significant part of national crime reduction strategies. Taggants may be able to prevent or monitor criminal offences by associating an object with a specific piece of information. While the material properties of a taggant will largely vary between application purposes, a specific 'coding' element will generally be incorporated to infer marker uniqueness. With the speed, simplicity and accuracy of coding component analysis largely determining the overall efficacy of taggants, continuing advances in portable in-field analysis, nanotechnology and material science should have allowed for the development of new and improved forensic marking agents. However, the limited amount of recent research in this area suggests that this is not the case. This critical review therefore examines the current state of presently available taggants before attempting to offer insight into the future direction of forensic marking technology.

## Keywords

Taggant; Forensic; Identification; Security; Fluorescence; DNA; Peptide; Mass Spectrometry

## 1. Introduction

Costing approximately £24 billion a year, the social and economic effect of organised criminal activity in the UK is substantial. Offences related to the trafficking of illicit drugs (£10.7 billion), organised fraud (£8.9 billion) and acquisitive crime (£1.8 billion) are reported to be the largest sources of criminal revenue [1]. With actual costs likely to be even greater (as such estimates are only based on data from recorded crimes), developing strategies that are able to prevent or aid in the investigation of these offences is a significant priority of the forensic science community.

Among the number of products made commercially available for the purpose of reducing criminal activity, forensic taggants may be seen as one of the most successfully employed. Taggant materials have been widely accepted for use as evidence within the UK court system since 2008, with certain marking agents having aided in the conviction of over one thousand criminal offenders since that time. Based on statistics published by the Metropolitan Police Service, household burglary across London

was decreased by nearly 30% in areas where forensic taggants were applied (i.e. SmartWater®). Taggants are a class of materials that can be applied to or incorporated within an object in order to make it identifiable [2]. This is achieved by producing each 'batch' of taggant in an entirely unique formulation, allowing the particular molecular composition to be registered against a specific piece of information. Once recovered at a later date, this composition can be analysed in order to reveal the identity of the taggant and thus, the object it is marking [3].

A forensic taggant displaying entirely ideal characteristics should be of low cost to produce, have high coding capacity, be non-toxic to individuals and the surrounding environment, be simplistic and inexpensive to detect and analyse via non-destructive means and be of a complex enough nature to prevent duplication. The majority of taggant manufacturers claim that their products are versatile enough to be applied in a broad range of circumstances. However, forensic marking materials are most extensively utilised for four distinct purposes (Fig. 1):

- Property marking – Taggants may be applied upon valuable items such as vehicles and electronics in order to associate them with a specific owner. If that property becomes subject to theft but is later recovered, it may be returned to the original purchaser [4, 5]. Property taggants are usually invisible to the naked eye in order to prevent spoiling the item's aesthetic or to avoid detection by offenders.
- Anti-counterfeiting – Industries at risk of counterfeiting activities (pharmaceuticals, clothing, currency, etc.) may choose to incorporate taggants into their products to distinguish them from potential forgeries [6]. Marking agents used for this application are typically non-visible and have replaced traditional barcoding and hologram anti-counterfeit technologies, which are more easily duplicated [7].
- Tracking – Materials used in the manufacture of hazardous or potentially illegal products may be tagged by law enforcement agencies to aid detection or track initial origin. Such marking agents are predominantly applied during the production of industrial explosives that are susceptible to illicit use [8]. Robust taggants able to survive detonation may be detected to reveal information regarding type of explosive, supplier or batch ID. Taggants may also be covertly added to bulk volumes of illegal narcotics, allowing their distribution to be monitored [9].
- Monitoring – Taggants may be placed on exterior surfaces of houses, commercial properties or objects of value. As a result of criminal activity (such as trespassing or out-of-hours burglary), these taggants may be transferred to an individual, providing strong physical evidence to associate them with that offence [10]. Additional information of the suspect's subsequent activity can also be gained if the taggant undergoes a secondary object-to-person-to-object transfer.

The physical characteristics of a taggant will depend largely upon which of these four purposes it is being used for (Fig. 2). For example, markers utilised to stain currency stolen during a CViT (Cash-and-valuables-in-transit) robbery should remain permanent after application and should not be easily removed [5]. Conversely, a forensic coating applied to firearms and cartridge casings in an attempt to establish handling, should be designed to be as transferrable as possible [11]. The range of taggant formulations is therefore extensive. Markers may be dispersed within a medium such as grease, paint or ink, sprayed via aerosol, applied directly as a powder or embedded into materials during manufacturing processes [12].

Taggants designed to be covertly applied (in order to avoid destruction or discovery by offenders) may also include an additional component to allow detection at a later time point. Being invisible to the naked eye, these taggants require localization in order to be successfully recovered for analysis. This is most often achieved by adding fluorescent compounds to the taggant, which will then emit light when excited at certain wavelengths of the UV or visible spectrum [13]. Other detection methods involve the inclusion of volatile chemicals for vapour identification [14] or radioactive isotopes [3].

While these detection constituents may not always be required within a taggant, all forensic markers will include a main 'coding' element in order to infer a required statistical uniqueness. By creating a formulation that is unable to be duplicated, a taggant may effectively 'store' information regarding the object it is marking [13].

As the success of a taggant will be largely influenced by the ability of this code to be recovered and analysed, all marker coding methods produced within the UK are subject to validation by the British Standards Institution (BSI) under Publicly Available Specification 820:2012 (PAS 820:2012). In this process, taggants are exposed to a number of accelerated UV light, temperature and humidity conditions in order to simulate the longevity of coding materials within a variety of circumstances.

Despite the extensive commercialisation of taggant technology (Table 1), detailed information regarding the synthesis and manufacturing procedures of such marking agents is extremely limited within academic literature. Whilst publishing such protocols within the public domain may result in a decrease in taggant efficiency (by allowing offenders to become familiar with such materials), it is still pertinent to conduct a thorough exploration into the different encoding methods used in commercially established forensic marking agents (Fig. 3). This critical review therefore attempts to examine these coding mechanisms, as well as explore recent research that may indicate how forensic taggant technology is likely to progress within the near future.

## 2. Current technology

### 2.1 Physical taggants

Renowned for being one of the earliest existing forensic marking materials; physical taggants possess coding systems that are based on the simple morphological properties of their components. These taggants are generally made unique by exploiting solid particles of a specific size, appearance or structural arrangement [29]. Such encoding mechanisms may also be described as 'graphical', as analysis is usually achieved by basic visual methods such as low-power microscopy [13].

One coding element commonly used in physical taggants is the microdot, a small polymer disc between 2-1000  $\mu\text{m}$  in size containing minute photographic information. Text or images etched onto microdots are usually too small to be observed by the naked eye alone but may be revealed upon optical magnification [30]. Although initially developed as a covert means of transferring data during World War II, companies such as DataDot and Microtrace have taken advantage of microdot technology by including them in a number of ink and varnish based suspensions [3]. The dots within these suspensions are then able to act as simplistic tagging mechanisms by being imprinted with a unique numeric code, which is then registered against a particular owner on an electronic database [16].

Another physical tagging approach pioneered in the 1970's by the US company 3M, focuses on the use of small plastic particles, constructed using a number of different proprietary coloured materials. These materials are combined in a sequence of layers, the order of which will be specific to each formulation of taggant produced [17]. A visual inspection of the layers is then reveals the identity of

the marker and the batch from which it originated. The robust nature of the materials used in layer construction has resulted in these particles becoming a predominant tagging agent for the identification of post-detonation explosive materials [31].

However, **in a number of circumstances** the utilisation of physical type taggants may be considered disadvantageous. Marking agents based on graphical identification strategies will generally have very little 'space' in which coding can take place [2]. This consequently limits the amount of statistically unique particles (and thus taggant formulations) that may be produced. Applications such as property marking, which necessitate a continuous stream of distinct codes, may benefit better from the use of molecular-based tagging agents, which have a higher coding capacity. Other drawbacks include the easy observability of physical taggants due to the relatively large size of incorporated solid particles, limiting their use within covert situations [12].

## 2.2 Spectroscopic taggants

In spectroscopic taggants, a number of molecules possessing differing optical qualities are combined together to create a single mixture with a spectrally unique signature. These taggants utilise multi-component encoding strategies, achieving their individuality from a precise combination of different emission wavelengths or intensities [13]. However, in certain taggant designs, these optically distinct molecules may be enclosed together within solid particles (to increase taggant stability or simplify analysis) and may thus be described as single-component systems [32]. Determining the identity of these marking agents is usually performed via the analysis of overall emission signature by simple spectrophotometry techniques.

The materials most often utilised within spectroscopic taggants are simple **non-toxic** organic dyes that fluoresce in different regions of the visible spectrum [33]. Companies such as Luminex and Spectra Systems manufacture a number of marking agents based on the integration of several cyanine, phthalocyanine or squaraine-based fluorophores [19, 21]. The low cost and wide availability of organic dyes ensures that the production of spectroscopic markers remains inexpensive, but may also result in the illegal reproduction of taggants themselves if fluorophores are recognised and acquired by counterfeiters [7]. Organic dyes are also disadvantaged by broad spectral emission overlaps, short fluorescence lifetimes and sensitivity to photobleaching [12].

Industrial and academic research has attempted to overcome these challenges by developing spectroscopic taggants that include more sophisticated optical components. Guillo *et al.* recently reported the development and commercialisation of a large array of spectroscopic coding materials based on **non-toxic** lanthanide ion complexes [34]. The unique temporal and spectral characteristics associated with these complexes are considered extremely difficult to duplicate by materials available to forgers, which may significantly deter potential attempts at taggant duplication. The narrow emission bands of rare-earth materials may also serve to increase the discriminatory power of a taggant by allowing more optically distinct components to be added to a marker without spectral overlap [13]. As the number of existing lanthanide elements is limited, varying the emission intensity ratios as well as the wavelengths of rare-earth materials is often used to further increase this statistical discrimination.

## 2.3 Chemical taggants

Much like spectroscopic markers, inorganic taggants rely on a multiplex of molecules within a specific combination to infer identity. However, instead of optical signature detection, chemical encoding is achieved from a fixed set of trace materials, which may be either present or absent from a mixture [35]. Each of these materials is tested for on an individual basis, essentially creating a form of 'binary string' data in which each position of the string represents a different compound. The presence of a

particular trace may be indicated by a "1" in that position of the string, whilst its absence may be denoted by a "0" [36]. Taggant identity is determined by the comparison of binary sequence data generated from trace analysis with a list of strings registered on a database.

The UK's largest manufacturer of forensic marking technology SmartWater, bases the majority of their commercially available reagents on this premise. In these taggants rare-earth lanthanides are also utilised but are identified by their individual mass through laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) rather than fluorescence emission [4]. A method of using isotopic materials as chemical encoding components for the authentication of fuels, consumer products and agrochemicals has also been developed by U.S. firm Authentix. These trace elements usually consist of deuterated organic or inorganic compounds that are then detected by either gas-chromatography mass spectrometry (GC-MS) or multiplex immunoassay systems [22, 37]. The increased sensitivity of chemical analysis methods over spectroscopic techniques allows coding materials to be included at much lower quantities (as little as parts per billion) than other taggant types. This in turn prevents the physical properties of a marker from being altered (unlike those containing large insoluble particles, which may appear more 'granular' in nature) and may thus lower the risk of unwanted detection.

Once again, these taggant classes are not without limitations. Binary string data can be compromised if one or more components within a mixture is removed or destroyed, or if extraneous material are added (for example, by accidental mixing of two different taggant formulations) [35]. Chemical multi-component encoding may also not be compatible with highly transferrable marking agents as the different underlying chemical structures of tracing elements may cause them to be transferred at dissimilar rates, leading to incomplete recovery and subsequent erroneous identification.

#### *2.4 DNA taggants*

Discovered in the early 1950's as the foundation of information storage for all known life forms, the immense coding ability of deoxyribonucleic acid (DNA) has been recognised for a considerably long period [38]. Since that time, a range of methods have been developed for the storage of non-genetic data within DNA molecules [13]. As a result, manufacturers such as Selectamark Security Systems [27], TraceTag [28] and Applied DNA Sciences [24] now produce forensic marking agents that can infer identity via the use of unique oligonucleotide sequences. Such sequences may be naturally occurring, entirely synthetic or a combination of both (e.g. botanical DNA shuffled into a random arrangement) [39]. The order of single nucleotide units comprising a sequence will be individual to each formulation of taggant produced, allowing separate batches to be distinguished.

The benefits of near-unlimited coding capacity, straightforward synthesis and low toxicity of genetic material have ensured that DNA taggants are one of the most prevalent forms of forensic marking agents commercially available. However, challenges regarding the application of DNA-based tagging approaches are twofold. Sequencing techniques employed for the analysis of oligonucleotides are typically more cost, reagent and labour-intensive than methods used to identify other types of taggant coding components [40]. As the genetic material recovered from a taggant also requires amplification by the polymerase chain reaction (PCR) prior to testing, this method of analysis can be especially expensive [2].

Furthermore, DNA is a relatively sensitive molecule and can degrade under the normal temperature, oxidation, radiation and chemical and enzymatic activity levels associated with ambient environmental conditions [41, 42]. While DNA damages caused by these stresses may be repaired by a number of processes within living organisms, the same cannot be said of the synthetic oligonucleotides included within forensic taggants. Concerns have therefore been raised over the general stability of DNA-based tagging materials and the subsequent compromise of genetic coding sequences [2, 13, 43].

However, encapsulation of nucleotides within silica microbeads [44] or plant materials [45] may be carried out in order to protect against this potential degradation.

### 3. Future perspectives

Each of the commercially available taggant mechanisms included within this review displays a distinct set of strengths and weaknesses, either in terms of coding capacity, covert usage, overall stability or method of analysis (Table 2). These disadvantages currently prevent a single taggant type from being used as a universal product identification system, necessitating the application of separate marking methods for different purposes [3]. However, it must also be noted that major practical innovations in the fields of nanotechnology, material science and analytical instrument portability have occurred since the scientific technologies behind these coding mechanisms were established. These advances are now being exploited by a number of research groups in an attempt to develop forensic tagging reagents that can identify any object, regardless of situation or circumstance.

Much of the research surrounding the design of globally applicable marking systems has focused on the use of nanomaterial-based encoding strategies. Nanoscale particles, wires and tubes all show great potential as next generation tagging mechanisms due to their small size (preventing detection and the physical alteration of marker properties), range of potential analysis methods and ease of formulation within traditional marking reagent media [12].

Semi-conductive quantum dot nanoparticles are currently being utilised to create spectroscopic taggants with optical qualities superior to those employing organic dyes or lanthanide ion complexes [46]. With narrow emission wavelengths, colour-tuneable signals and environmental-independent fluorescence properties, quantum dots may represent an excellent opportunity to improve the multi-component spectral encoding mechanisms presently utilised by spectroscopic taggants [13, 47]. Commercialisation of this technology may have been hindered by the relative cytotoxicity of metal ions used in the synthesis of quantum dot particles [48]. Efforts to reduce these potential damages to human health and the surrounding environment are currently underway through the development of heavy metal-free quantum dots [49].

However, there has been debate over whether fluorescent molecules should be included within tagging materials at all, as many offenders are now familiar with optical anti-counterfeit methods and are able to identify markers with obvious visible emissions [12]. In an attempt to resolve this issue, a number of taggants have recently been produced that can only be analysed via the use of Raman spectroscopy [50, 51]. Much like other forms of spectroscopic coding mechanisms, these reagents comprise compounds mixed in a specific combination to produce a unique spectral fingerprint (which in this case, is generated through the inelastic scattering of monochromatic laser light). The main advantage of this method is that Raman spectroscopy may also be used for the initial detection of tagging materials as well as identification, avoiding the requirement of additional tracing components that may result in unwanted discovery [12]. The sensitivity of such detection may also be greatly increased through the phenomenon of surface enhanced Raman scattering (SERS) by the direct conjugation of Raman-active compounds to a number of metallic nanoparticles [52].

Another coding technique designed by Duong *et al.* utilises the properties of nanomaterials beyond fluorescence involves deriving the individual melting-temperatures of a panel of solid particles within a taggant mixture to provide it with a unique thermal barcode [2]. The presence or absence of these so called 'phase-change' nanoparticles within a taggant is assessed using differential scanning calorimetry (DSC) in a linear thermal scan, which generates a specific melting-point peak for each component [7]. However, only a limited selection of nanoparticles has currently been developed for



this purpose [53]. It is likely that this number will have to be increased in order for this system to possess the level of statistical differentiation required from a universal tagging system.

Aside from nanoparticle-based encoding methods, recent developments in novel tagging technology have also been achieved by the use of synthetic polypeptide sequences [54]. The principle of inferring identity through the unique order of successive amino acids has been theorised since the early 1980's [55]. However, only within the last year have methods been realised for their practical application and analysis within forensic marking materials [56]. Fig. 4 demonstrates this process, in which an oil-based medium containing hydrophilic peptide molecules is first applied onto the surface of an object. As a result of handling during criminal activity, this reagent is transferred to an individual, associating them with the offence. Detection by added fluorescence tracers is performed to permit recovery of the taggant by targeted swabbing. Hydrophilic peptides may be then isolated from the medium by simple liquid-liquid extraction, allowing their mass (and therefore sequence) to be determined by electrospray ionisation mass spectrometry (ESI-MS).

As an alternative form of single-component biomolecular coding, peptide taggants possess a number of advantages over current commercially available DNA-based marking systems. The statistical coding capacity of polypeptide chains is significantly greater than that of oligonucleotides, owing to the 22 different natural amino acids that may be used as individual sequence units (compared to the four bases possessed by DNA) [57]. Kydd reports that a string of 10 random amino acids can code up to  $4 \times 10^{13}$  unique sequences [55]. ESI-MS methods are also quicker than DNA sequencing (with processes from detection to analysis able to be completed in less than one hour) and are becoming increasingly more portable, which could allow the point-of-care testing of peptide tagging reagents. Peptides are additionally easy to produce at low cost, are environmentally harmless and relatively inert (with stability being further increased by simple chemical modifications) [57].

#### 4. Conclusions

By authenticating objects, deterring theft and monitoring illegal activity, forensic taggants continue to play a pivotal role in the reduction of criminal enterprise. This review has attempted to highlight the significant abilities of tagging materials to infer identity in an enormous range of products, as well as document the most prominent commercially available coding mechanisms used for achieving this aim.

Whilst it is quite clear that none of the commercial marking methods available currently possess every single one of the qualities demanded of a universally applicable forensic taggant, the recent research covered within this article demonstrates that concerted efforts towards taggant improvement are occurring.

The myriad of surfaces, environments and time-frames in which a taggant may be applied makes the technical innovation and validation of forensic marking materials relatively challenging. Only through continuing advances in material science, nanotechnology and instrumental analysis, can steps be made towards the development of a single versatile platform for object identification. It is hoped that this review will encourage researchers working within the field of analytical chemistry to take interest in taggant technology and engage with this development in order to make universal marking systems a reality.

#### References

- [1] H. Mills, S. Skodbo, P. Blyth, Understanding organised crime: Estimating the scale and the social and economic costs, 2013.
- [2] B. Duong, H. Liu, L. Ma, M. Su, Covert thermal barcodes based on phase change nanoparticles, Sci. Rep., 4 (2014) 5170.

- [3] J.J. Plimmer, Choosing the correct forensic marker(s) in currency, document, and product protection, *Proc. SPIE*, 6075 (2006) 60750-60759.
- [4] S.H. Wise, J.R. Almirall, Chemical taggant detection and analysis by laser-induced breakdown spectroscopy, *Appl. Opt.*, 47 (2008) G15-G20.
- [5] B.K. Mead, J. Hayward, B. Liang, M. Wan, T. Benson, J. Karp, The Role of DNA-Stained Currency in Gang Robberies within the United Kingdom, *J. Forensic Sci.*, 59 (2014) 264-267.
- [6] H.J. Bae, S. Bae, C. Park, S. Han, J. Kim, L.N. Kim, K. Kim, S.-H. Song, W. Park, S. Kwon, Biomimetic Microfingerprints for Anti-Counterfeiting Strategies, *Adv. Mater.*, 27 (2015) 2083-2089.
- [7] B. Duong, H. Liu, C. Li, W. Deng, L. Ma, M. Su, Printed Multilayer Microtaggants with Phase Change Nanoparticles for Enhanced Labeling Security, *ACS Appl. Mater. Interfaces*, 6 (2014) 8909-8912.
- [8] J.W. Grate, R.G. Ewing, D.A. Atkinson, Vapor-generation methods for explosives-detection research, *Trends Anal. Chem.; TrAC*, 41 (2012) 1-14.
- [9] N. Kaish, J. Fraser, V. Otugen, S. Popovic, Method for remote detection of volatile taggant, Google Patents, 2000.
- [10] C. Smith, S. Strauss, L. DeFrancesco, DNA goes to court, *Nat. Biotech.*, 30 (2012) 1047-1053.
- [11] P.A. Sermon, M.P. Worsley, Y. Cheng, L. Courtney, V. Shinar-Bush, O. Ruzimuradov, A.J. Hopwood, M.R. Edwards, B. Gashi, D. Harrison, Y. Xu, Deterring gun crime materially using forensic coatings, *Forensic Sci. Int.*, 221 (2012) 131-136.
- [12] M. Wang, B. Duong, H. Fenniri, M. Su, Nanomaterial-based barcodes, *Nanoscale*, 7 (2015) 11240-11247.
- [13] D. Paunescu, W.J. Stark, R.N. Grass, Particles with an identity: Tracking and tracing in commodity products, *Powder Technol.*, 291 (2016) 344-350.
- [14] J.M. Perr, K.G. Furton, J.R. Almirall, Solid phase microextraction ion mobility spectrometer interface for explosive and taggant detection, *J. Sep. Sci.*, 28 (2005) 177-183.
- [15] C.D. Loving, Composite microdot and method of forming the same, Google Patents, 1994.
- [16] G.F. Swiegers, B.W. Bootle, G.M. George, Method and system for identifying items, Google Patents, 2012.
- [17] P.K. Lee, Method of tagging with color-coded microparticles, Google Patents, 1977.
- [18] C. Outwater, Product authentication system, Google Patents, 2002.
- [19] D.J. Chandler, B.A. Lambert, J.J. Reber, S.L. Phipps, Precision fluorescently dyed particles and methods of making and using same, Google Patents, 2003.
- [20] W.J. Kerns, B. Brogger, J.L. Esterberg, Identification particles and system and method for retrospective identification using spectral codes, Google Patents, 2006.
- [21] N. Lawandy, A. Smuk, L. Olson, C. Zepp, Multi wavelength excitation/emission authentication and detection scheme, Google Patents, 2014.
- [22] T.G. Wilkinson, E. Dorland, Marking fuel for authentication, Google Patents, 2010.
- [23] T. Baque, Method of authenticating and/or identifying an article, Google Patents, 2013.
- [24] L. Jung, J.A. Hayward, M.B. Liang, DNA marking of previously undistinguished items for traceability, Google Patents, 2014.
- [25] G.M. George, Article Marking System, Google Patents, 2009.
- [26] C.L. Butland, B. Baggot, Labeling technique for countering product diversion and product counterfeiting, Google Patents, 2000.
- [27] J. Brown, B. Reichert, Compositions for use in security marking, Google Patents, 2012.
- [28] R. Sleat, G. Van Lint, A marking apparatus for nucleic acid marking of items, Google Patents, 2012.
- [29] A.K. Deisingh, Pharmaceutical counterfeiting, *Analyst*, 130 (2005) 271-279.
- [30] M.R. Whitehead, R. Peakall, Microdot technology for individual marking of small arthropods, *Agric. Forest Entomol.*, 14 (2012) 171-175.
- [31] L. Husáková, J. Šrámková, J. Staňková, P. Němec, M. Večeřa, A. Krejčová, M. Štancl, Z. Akštein, Characterization of industrial explosives based on the determination of metal oxides in the identification particles by microwave digestion and atomic absorption spectrometry method, *Forensic Sci. Int.*, 178 (2008) 146-152.
- [32] H.H. Pham, I. Gourevich, J.E. Jonkman, E. Kumacheva, Polymer nanostructured material for the recording of biometric features, *J. Mater. Chem.*, 17 (2007) 523-526.
- [33] N.H. Finkel, X. Lou, C. Wang, L. He, Barcoding the Microworld, *Anal. Chem.*, 76 (2004) 352-359.
- [34] O. Guillou, C. Daignebonne, G. Calvez, K. Bernot, A Long Journey in Lanthanide Chemistry: From Fundamental Crystallogenesis Studies to Commercial Anticounterfeiting Taggants, *Acc. Chem. Res.*, 49 (2016) 844-856.

- [35] P. Przybylowicz, *Black and Smokeless Powders, Technologies for Finding Bombs and the Bomb Makers*, National Academy Press, Washington DC, 1998.
- [36] M. Cleary, *Method of identifying a surface*, Google Patents, 1998.
- [37] D.W. Britton, M.J. Wraith, *Detection of chemicals by immunoassay*, Google Patents, 1999.
- [38] L. Hood, D. Galas, *The digital code of DNA*, *Nature*, 421 (2003) 444-448.
- [39] J.A. Hayward, J. Meraglia, *DNA to safeguard electrical components and protect against counterfeiting and diversion*, *Proc. ISTFA*, 4 (2011) 238-241.
- [40] J.P.L. Cox, *Long-term data storage in DNA*, *Trends. Biotechnol.*, 19 (2001) 247-250.
- [41] D. Paunescu, M. Puddu, J.O.B. Soellner, P.R. Stoessel, R.N. Grass, *Reversible DNA encapsulation in silica to produce ROS-resistant and heat-resistant synthetic DNA 'fossils'*, *Nat. Protocols*, 8 (2013) 2440-2448.
- [42] T. Lindahl, *Instability and decay of the primary structure of DNA*, *Nature*, 362 (1993) 709-715.
- [43] A. Glover, N. Aziz, J. Pillmoor, D.W.J. McCallien, V.B. Croud, *Evaluation of DNA as a taggant for fuels*, *Fuel*, 90 (2011) 2142-2146.
- [44] M. Puddu, D. Paunescu, W.J. Stark, R.N. Grass, *Magnetically Recoverable, Thermostable, Hydrophobic DNA/Silica Encapsulates and Their Application as Invisible Oil Tags*, *ACS Nano*, 8 (2014) 2677-2685.
- [45] P. Reep, M.H. Liang, J.J. Sheu, *System and method for authenticating multiple components associated with a particular product*, Google Patents, 2006.
- [46] M. Zhou, S. Chang, C.P. Grover, *Cryptography based on the absorption/emission features of multicolor semiconductor nanocrystal quantum dots*, *Opt. Express*, 12 (2004) 2925-2931.
- [47] G.M. Williams Jr, T. Allen, C. Dupuy, T. Novet, D. Schut, *Optically coded nanocrystal taggants and optical frequency IDs*, *Proc. SPIE*, 7673 (2010) 76730-76744.
- [48] R. Hardman, *A Toxicologic Review of Quantum Dots: Toxicity Depends on Physicochemical and Environmental Factors*, *Environ. Health Perspect.*, 114 (2006) 165-172.
- [49] H. Li, W.Y. Shih, W.-H. Shih, *Non-heavy-metal ZnS quantum dots with bright blue photoluminescence by a one-step aqueous synthesis*, *Nanotechnology*, 18 (2007) 205604.
- [50] R. Li, Y. Zhang, J. Tan, J. Wan, J. Guo, C. Wang, *Dual-Mode Encoded Magnetic Composite Microsphere Based on Fluorescence Reporters and Raman Probes as Covert Tag for Anticounterfeiting Applications*, *ACS Appl. Mater. Interfaces*, 8 (2016) 9384-9394.
- [51] K.D. Osberg, M. Rycenga, G.R. Bourret, K.A. Brown, C.A. Mirkin, *Dispersible Surface-Enhanced Raman Scattering Nanosheets*, *Adv. Mater.*, 24 (2012) 6065-6070.
- [52] M. Natan, S. Norton, R. Freeman, S. Penn, I. Walton, *Nanoparticles As Covert Taggants In Currency, Bank Notes, And Related Documents*, Google Patents, 2007.
- [53] D.C. Hyun, N.S. Levinson, U. Jeong, Y. Xia, *Emerging Applications of Phase-Change Materials (PCMs): Teaching an Old Dog New Tricks*, *Angew. Chem. Int. Ed.*, 53 (2014) 3780-3795.
- [54] J. Gooch, C. Koh, B. Daniel, V. Abbate, N. Frascione, *Establishing evidence of contact transfer in criminal investigation by a novel 'peptide coding' reagent*, *Talanta*, 144 (2015) 1065-1069.
- [55] P.H. Kydd, *Polypeptides as chemical tagging materials*, Google Patents, 1982.
- [56] J. Gooch, H. Goh, B. Daniel, V. Abbate, N. Frascione, *Monitoring Criminal Activity through Invisible Fluorescent "Peptide Coding" Taggants*, *Anal. Chem.*, 88 (2016) 4456-4460.
- [57] G. Dollinger, *Nonbiological Applications*, in: K. Mullis, F. Ferre, R. Gibbs (Eds.) *The Polymerase chain reaction*, Springer, Boston, USA, 1994 pp. 265-274.



Fig. 1. The four main applications utilising forensic taggants.

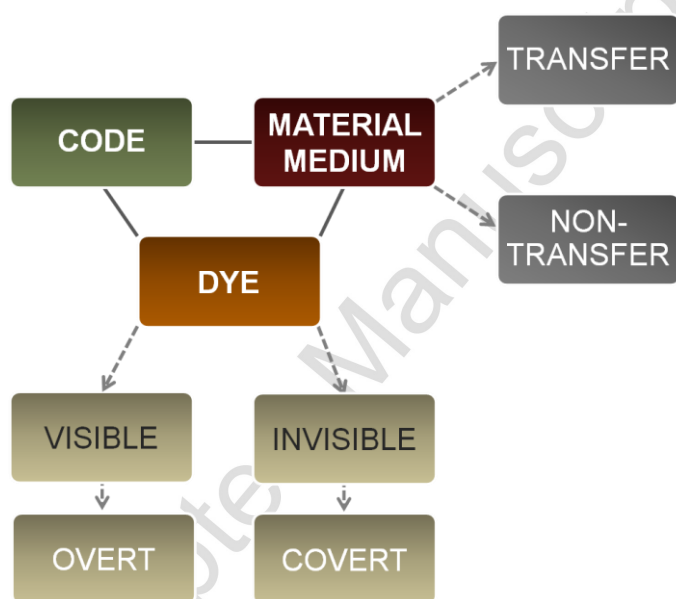
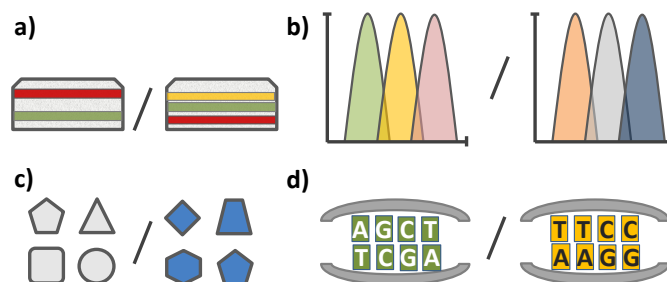
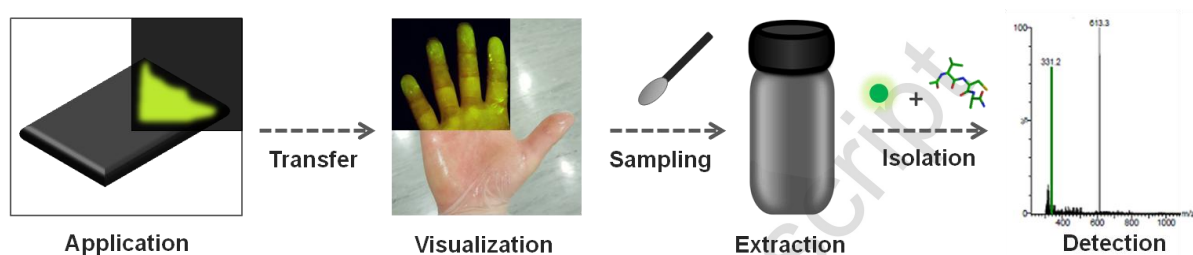


Fig. 2. Forensic taggants may include different components (i.e. code, dye and material/medium), which can determine its physical characteristics.



**Fig. 3.** Code uniqueness provided by differences in: a) physical characteristics (i.e. multiple coloured layer particles); b) optical spectra; c) chemical composition; d) DNA sequences.



**Fig. 4.** Schematic of peptide-based taggant application and analysis. Adapted from [56].

**Table 1.** Commercially available forensic taggants.

Coding Type	Manufacturers	Taggants Produced	Analysis Method	Uses
Physical	Alpha•Dot DataDot Microtrace	Alpha•Dot [15] DataDots® [16] Microtaggant® [17]	Optical microscopy Optical microscopy Optical microscopy	Property marking, vehicle marking Property marking, anti-counterfeiting Anti-counterfeiting, explosives detection
Spectroscopic	DNA Technologies Luminex Microtrace Spectra Systems	SmartDye™ [18] MagPlex® spheres [19] Spectral Taggant™ [20] SpectraFluor™ [21]	Spectrofluorometry Flow Cytometry Spectrofluorometry Spectrofluorometry	Anti-counterfeiting Anti-counterfeiting, biological labeling Currency marking, security printing Currency marking, product tracking
Chemical	Authentix SmartWater Polysecure	Authentix [22] SmartWater® [4] Brandproof® [23]	Immunoassay, GC-MS LA-ICP-MS XRF spectroscopy	Fuel Adulteration, anti-counterfeiting Property Marking, activity monitoring Anti-counterfeiting
DNA	Applied DNA Sciences DataDot DNA Technologies SelectaMark TraceTag	SigNature® [24] dDotDNA® [25] DNA Matrix™ [26] SelectaDNA® [27] CypherMark™ [28]	DNA sequencing DNA sizing analysis DNA sequencing DNA sequencing DNA sequencing	Property marking, anti-counterfeiting, CVIT Property marking, anti-counterfeiting Anti-counterfeiting Property marking, activity monitoring Property marking, anti-counterfeiting

**Table 2.** Summary of the advantages and disadvantages of commercially available forensic taggants.

Coding Type	Advantages	Disadvantages
Physical	Simplistic analysis Inexpensive	Limited coding capacity Less covert
Spectroscopic	Simplistic analysis Inexpensive	Subject to counterfeiting Limited coding capacity
Chemical	Analysis sensitivity More covert	Prone to misidentification Incomplete recovery
DNA	High coding capacity Low toxicity	Expensive analysis Possible degradation

Accepted Manuscript