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Addressing forensic science challenges with nuclear analytical techniques – A review

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ABSTRACT

We review the application of Nuclear Analytical Techniques (NATs) to forensic problems for the first time. NATs include neutron activation analysis (NAA), carried out in nuclear reactors for elemental analysis; accelerator-based techniques, mainly Ion Beam Analysis (IBA) for elemental and molecular analysis; and Accelerator Mass Spectrometry (AMS) for dating of traces of forensic interest by “radiocarbon dating” and other related methods. Applications include analysis of drugs of abuse, food fraud, counterfeit medicine, gunshot residue, glass fragments, forgery of art objects and documents, and human material. In some applications only the NATs are able to provide relevant information for forensic purposes. This review not only includes a wide collection of forensic applications, but also illustrates the wide availability worldwide of NATs, opening up opportunities for an increased use of NATs in routine forensic casework.

1. Introduction

With the continuous evolution of nuclear science and technology a number of techniques with analytical and imaging capabilities have become accessible. These are known collectively as “Nuclear Analytical Techniques” (**NATs**) and include both ion and neutron beam methods [1,2]. NATs rely on the interaction of a beam of ions or neutrons with the atoms (or atomic nuclei) of the specimen probed. Based on such interactions, specific elements and isotopes can be identified and quantified in a specimen or an object. The physics of all of these techniques is very well known, and NATs may be used in a fully quantitative way, that is, first principles quantification is available as is indicated for each method in the appropriate place.

NATs can be made available by a single provider or through buying in services from multiple external providers, and de Kinder & Pirée have recently given a valuable overview of the need for a coherent organisation of providers of forensic services together with a discussion of the range of issues involved [3]. Such quality control measures are in place in some research reactor and accelerator facilities, nevertheless further developments would facilitate many possible applications of NATs in forensic science.

There are different models and applications of forensic science, having in common the definition of “trace” as the most elementary information that results from a crime (see for example Pierre Margot’s “*Traceology*” chapter in the *Routledge Handbook*, 2017 [4]). Therefore, forensic science could be defined as “*the scientific study of the properties and application of traces*” [5]. But the characterisation of traces is not always straightforward, in which case it cannot usually be carried out entirely in a forensics laboratory: sometimes the analytical capabilities of external infrastructures are needed.

The oldest NAT, Neutron Activation Analysis (**NAA**), has been an important technique over the last half-century for the characterization of archaeological materials by elemental analysis (see the 2007 review by Speakman & Glascock [6], and the early interest and support of the IAEA which published the final report of a Co-ordinated Research Program on forensic and archaeological applications of NAA in 1977 [7]). The first three cases in USA courts depending on NAA data took place in 1964 and are described by

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Guinn [8]. Conclusions based on NAA had been already accepted as evidence in USA courts in at least 50 trials by 1968, reported by Karjala [9], who makes it clear that the use of such technical evidence must be presented in an appropriate (non-trivial) legal framework.

Research is also developing very powerful new forensic applications for accelerator-based techniques, mainly Ion Beam Analysis (**IBA**) for elemental and molecular analysis [10] and Accelerator Mass Spectrometry (**AMS**) for dating of traces of forensic interest by the "radiocarbon dating" and other related methods [11]. AMS is well known to be able to distinguish with high sensitivity among various isotopes and radiocarbon dating has recently been reviewed by Hajdas et al. [12].

Use of these NATs is well known and widespread in the fields of materials science, environmental science, cultural and natural heritage etc. These NATs are mature, they are applied routinely in research and occasionally in industrial applications, and they have been applied in several forensic areas. Nevertheless, NATs are not routinely used in forensics applications.

One of the obstacles is that NATs do not allow portable (in situ) analysis but are carried out in big research infrastructures, usually requiring prior booking of beamtime, and generally involving time consuming collection, evaluation and interpretation of the analytical data. On the other hand, such investigations can provide extremely valuable analytical information not available from other techniques. Furthermore, sometimes these techniques require tailoring for the forensic applications to be available as a tool to be applied beyond a laboratory level, possibly including accreditation of the involved laboratories.

Despite the long history of applications of NATs in forensic science, we could not find any comprehensive scientific review of relevant forensics applications. Based on our research of the scientific literature it has become clear that improving the effectiveness of crime prevention and control is likely to be facilitated by nuclear analytical methods and that a review of these methods will be useful.

The aim of this paper is therefore to provide for the first time a review, both for forensic and law enforcement professionals making use of analytical results and also for the analysts themselves, of the multiplicity of NATs capabilities now available for forensic applications, discussing their strengths and weaknesses, and reviewing the most recent opportunities regarding forensic applications of NATs. These opportunities include, among others, crime investigation, food safety and health-related issues, detecting fake artefacts, illicit trade of products, wildlife, and environmental forensics. This paper will focus on how NATs can support forensic scientific investigations in a synergistic manner, with examples and case studies being shown in which nuclear analytical techniques provide added value and clear benefit beyond the routine, non-nuclear analytical techniques.

2. Capabilities and potential of NATs

The NATs reviewed in this article are listed in Table 1, while their analytical capabilities are summarised in Table 2, together with other common analytical techniques. The comparison of analytical performances will support forensic scientist already using non-NATs to evaluate the inclusion of NATs in their forensic procedures. A *Glossary* summarising these NATs is given in the **Annex**. Broadly speaking, there are three categories: (i) neutron methods, (ii) Ion Beam Analysis (**IBA**) methods, and (iii) Accelerator Mass Spectrometry (**AMS**) methods:

1. Neutron methods include activation, imaging, and scattering. They are bulk methods that can handle from milligram-sized to large samples in air. They are usually performed at research reactor facilities.

Table 1
List of selected analytical techniques.

Class	Technique	Acronym
Research reactors	Nuclear Activation Analysis	NAA
	Neutron Depth profiling	NDP
	Neutron Imaging	N-imaging
	Neutron reflectometry	NR
	Neutron Scattering	N-scattering
Accelerator (Ion Beam Analysis)	Elastic (non-Rutherford) Backscattering	EBS
	Elastic Recoil Detection	ERD
	Heavy Ion ERD	HI-ERD
	High Resolution PIXE	HR-PIXE
	(Inelastic) Nuclear Reaction Analysis	NRA,PIGE
	Particle Induced X-ray Emission	PIXE
	Rutherford Backscattering Spectrometry	RBS
	High Energy Secondary Ion Mass Spectrometry	HE-SIMS
	Total-Ion Beam Analysis	Total-IBA
Accelerator (AMS)	Accelerator Mass Spectrometry	AMS
Laboratory	Grazing Incidence X-Ray Fluorescence	GI-XRF
	X-Ray Reflectometry	XRR
	Auger Electron Spectrometry	AES
	Electron Probe Micro Analysis	EPMA
	Glow-discharge optical emission spectroscopy	GD-OES
	Laser Ablation ICP-MS	LA-ICP-MS
	Inductively Coupled Plasma Mass Spectrometry	ICP-MS
	Low Energy Ion Scattering	LEIS
	Scanning Auger Microscopy	SAM
	Spectroscopic Ellipsometry	SE
	Scanning Electron Microscopy	SEM
	Secondary-ion Mass Spectrometry	SIMS
	X-ray Photoelectron Spectroscopy	XPS
	Cross Section Transmission Electron Microscopy	XTEM

2. IBA is usually done in a vacuum but can be done in air. The information depth rarely exceeds 10 µm, and IBA is particularly powerful for near-surface elemental depth profiling [13]. IBA requires a megavolt ion beam accelerator. MeV ion beams can be focussed to deep submicron sizes, enabling direct imaging.
3. AMS (typically Radiocarbon) detects very low concentrations of natural isotopic abundances (in range between 10^{-12} and 10^{-16} in typical applications), with an extraordinary sensitivity. The sample may be a small as a few milligrams by providing a few 10 µg carbon, and counting times per sample are typically a few minutes. AMS uses the same (or similar) accelerator facilities as IBA, configured differently.

Neutron Activation Analysis (**NAA**) relies on the activation of the sample by neutrons, resulting in radioactive samples that emit characteristic gamma radiation. Based on the emitted radiation it is possible to identify the elements and isotopes present. A schematic representation of NAA steps and illustration of the neutron capture process is given in Fig. 1.

NAA is a mature technique with the main developments having occurred in the 1960s and 1970s. It can detect up to 70 elements in samples and entire objects in the milligram to kilogram range, with sensitivity up to parts-per-million and better in some cases. New breakthrough developments are not expected. Its status was summarised by the IAEA in 2001 [14]. Instrumental NAA is now recognised by the Consultative Committee for Amount of Substance: Metrology in Chemistry and Biology (CCQM) as being a reference technique of metrology for the "amount of substance", that can be a primary method of measurement [15,16].

Table 2
Summary of characteristics of selected analytical techniques.

Technique[1]	Measures	Sensitivity[2]	Test portion may be returned[9]	Depth profiling	Quantitative[3]	Information depth	Imaging	Depth resolution[2]
NAA	up to 70 elements	10^{-6}	Yes[10]	No	Reference[5]	bulk	No[8]	-
NDP	light isotopes	10^{-3}	Yes	Yes	Standards[6]	10 mm	No	20 nm
N-imaging	structure	10^{-2}	Yes	Radiography	No[7]	bulk	Yes	10 µm
NR	structure	10^{-3}	Yes	Interference	Reference	10 mm	No	5 nm
N-scattering	structure	10^{-2}	Yes	Interference	Reference	bulk	No	5 nm
EBS	all elements	10^{-3}	Yes	Yes	Total-IBA[6]	10 mm	Yes	5 nm
ERD	all elements	10^{-3}	Yes	Yes	Total-IBA[6]	500 nm	Yes	5 nm
HI-ERD	all elements	10^{-3}	No[4]	Yes	Standards	100 nm	No	2 nm
HR-PIXE	all elements	10^{-6}	Yes	Model	Total-IBA[6]	20 mm	Yes	50 nm
NRA,PIGE	light isotopes	10^{-4}	Yes	Yes	Total-IBA[6]	500 nm	Yes	5 nm
PIXE	all elements	10^{-6}	Yes	Model	Total-IBA[6]	20 mm	Yes	50 nm
RBS	all elements	10^{-3}	Yes	Yes	Reference	10 mm	Yes	2 nm
HE-SIMS	molecules	10^{-5}	No	No	No	100 nm	Yes	1 µm
Total-IBA	all elements	10^{-6}	Yes	Yes	Total-IBA	10 mm	Yes	2 nm
AMS	^{14}C and other isotopes		No	No	Relative isotope ratios	N/A	Rare	N/A
GI-XRF	structure	10^{-3}	Yes	Interference	Reference	50 mm	Yes	2 nm
XRR	structure	10^{-1}	Yes	Interference	Reference	1 mm	No	2 nm
AES	all elements	10^{-3}	Yes	Sputter	Standards	50 nm	No	2 nm
EPMA	all elements	10^{-4}	Yes	Model	Standards	2 mm	Yes	20 nm
GD-OES	all elements	10^{-7}	No	Yes	Reference	50 mm	No	50 nm
LA-ICP-MS	all elements	10^{-7}	No	Yes	Reference	10 mm	Yes	50 nm
ICP-MS	all elements	10^{-7}	No	Yes	Reference	10 mm	Yes	50 nm
SAM	all elements	10^{-3}	Yes	Sputter	Standards	50 nm	Yes	2 nm
SE	structure	10^{-3}	Yes	Model	Reference	10 mm	No	1 nm
SEM	topography	10^{-3}	Yes	Model	Standards	2 mm	Yes	100 nm
SIMS	elements, molecules	10^{-7}	No	Sputter	Standards	100 nm	Yes	2 nm
XPS	all elements	10^{-3}	Yes	Sputter	Standards	50 nm	No	0.5 nm
XTEM	all elements	10^{-2}	No	Xsection	Standards	500 nm	Yes	0.1 nm

[1] The acronyms are expanded in Table 1.
[2] Typical values.
[3] Standards: requiring use of a standard sample for comparison. Reference refers to techniques that can be used as primary direct reference methods. Total-IBA: see the annex.
[4] Depending on sample. HI-ERD is not intentionally destructive but can lead to erosion of the surface layers under analysis.
[5] Depending on NAA method used. Certified reference materials are often used.
[6] Quantification based on first principles plus use of experimentally determined physical quantities such as reaction probabilities.
[7] Quantification possible in some cases, such as hydrogen concentration.
[8] Prompt gamma NAA is used in imaging mode in a very limited number of facilities.
[9] For the purposes of this table, "No" indicates that the tested portion of the sample is consumed or destroyed during the analysis. "Yes" means that although the sample may be modified you do get the sample back after the analysis, and in many cases the changes in it may not be noticeable.
[10] After a waiting period to allow the induced activity to decay.

Quality assurance and quality control practices are well established in NAA [17–20]. For instance, NAA can be validated by replicate analysis of certified reference materials or other appropriate materials, in some cases done over decades in multi-user, multi-spectrometer facilities [21], and uncertainty budgets can be readily made [22].

NAA does not change the sample's chemical composition. It also does not require chemical destruction or dissolution of the sample with the inherent risks of elemental loss or contamination. Samples with mass a fraction of a gram can be measured. Entire whole objects can also be analysed, which is useful if the object cannot be sampled. If the object needs to be returned, for instance for further analysis, one must be aware that the samples are activated. The

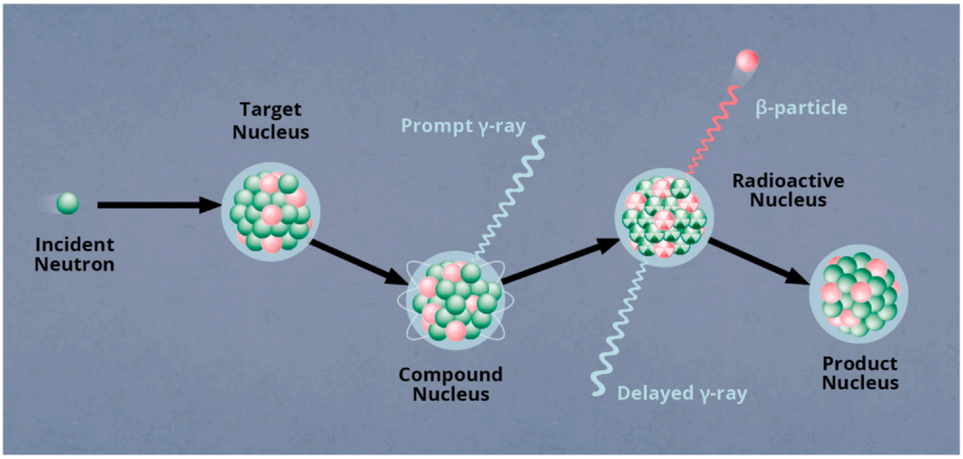

Fig. 1. Schematic representation of Neutron Activation Analysis steps and illustration of the neutron capture process.

induced radioactivity is in general very small and often decays quickly below the exemption levels as recommended by the IAEA [23] (and largely followed as in the USA [24] for example, although the approaches may vary in different countries). Once that happens, the sample can be returned to the originator, or be analysed in a forensics laboratory without restrictions. In typical environmental samples (e.g. soil), this is often immediately after analysis. In typical biological samples, this is often one to two months after analysis. In bullet fragments, this can be one month (e.g. for the conditions and bullet composition described in [25]), or more if large fragments cannot be split and need to be analysed intact. However, in the presence of some elements the waiting time can be much longer, and the sample can remain radioactive for years. When confronted with samples that need to be returned or made available for further measurements, NAA laboratories can make a very short test irradiation, that will not lead to significant activation, to determine whether such elements are present in significant amounts.

Neutrons can also be used for scattering and imaging, in a way complementary to the corresponding X-ray techniques since neutrons are strongly sensitive to light elements in a way X-rays are not. For instance, neutrons can probe non-destructively the inside of metallic objects, such as a hand grenade, and inspect their interior. Neutron imaging has been used in identification of artefacts [26], forensic anthropology [27] and forensic engineering [28]. The neutron doses involved in scattering and imaging are orders of magnitude lower than in NAA, and the objects analysed do not generally become activated, except if they contain elements such as uranium. Except in such cases, they can be released and handled immediately after the analysis.

IBA techniques, taken together synergistically in a way that is now feasible, can identify and quantify all elements, from matrix elements to trace elements at parts-per-million levels (or even lower in particular cases). Composition of different layers, elemental 2D distribution maps, and even 3D maps with special techniques, can be obtained. It was initially developed in the 1970s and 1980s as a special technique with limited application but has had substantial development mostly in the 21st century that has enabled a general application [29].

There are three main groups of IBA techniques: (i) elastic scattering methods (such as RBS and ERD – for acronyms see the Glossary in the **Annex**); (ii) atomic excitation methods (PIXE); and (iii) nuclear excitation methods (PIGE, NRA). “MeV-SIMS” is a new method using ion beam accelerators usually used also for IBA and may be done in conjunction with PIXE, but has special instrumentation and applications mostly distinct from those accessible with the other IBA methods. A schematic representation of some of the main IBA techniques is given in Fig. 2.

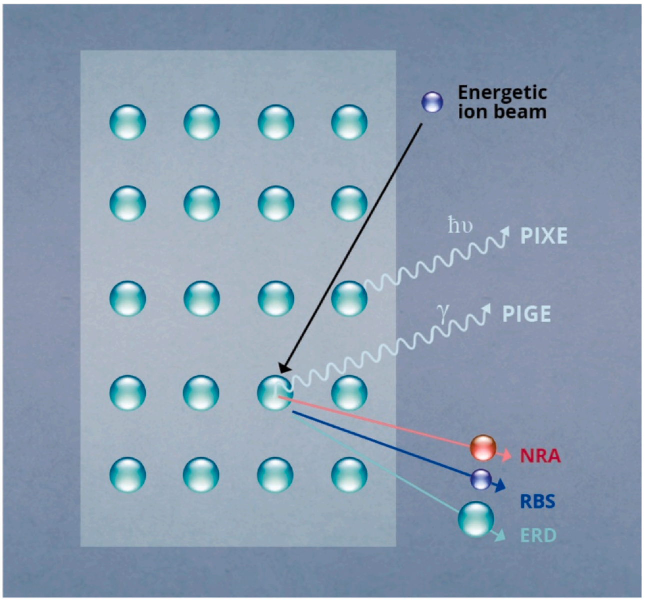

Fig. 2. Schematic representation of the main Ion Beam Analytical techniques. The nuclei are represented as spheres, the electrons are not represented. See the Annex for further information on each technique. The PIXE (Particle Induced X-ray Emission) interaction with the atomic electron shell produces a characteristic X-ray emission. The NRA (Nuclear Reaction Analysis) reaction product (red) detected is often a very energetic proton or alpha particle; when it is a gamma ray, the technique is usually called PIGE (Particle Induced Gamma ray Emission). The RBS (Rutherford Backscattered Spectrometry) detected particles (blue) are the same of the energetic ion beam. The ERD (Elastic Recoil Detection) detected particles (green) are elements present in the sample, which are lighter than the beam and recoil towards the detector.

In energetic IBA methods the beam energy is deposited in or near the sample surface and must modify the sample to at least some extent. The higher is the energy (and ion current) of the ion beam striking the surface, the more is the damage to the spot analysed, although in many cases the modification is imperceptible. The samples analysed by IBA may be subsequently re-analysed with other techniques (including the same or other IBA techniques and altogether different analytical techniques) leading to the same result of their composition (up to measurement uncertainties) that would have been obtained had the sample not been previously analysed. This is not trivial since there is significant power in an ion beam with MeV energies, which is dissipated mostly on the surface (from the nanometer to a few tens of microns in most cases). Damage-free charge and energy dissipation happens in the majority of the cases, but it is not guaranteed, especially for organic materials. There is a very large literature on how to minimise ion beam-induced damage during IBA, which was helpfully summarised by the Handbook on the subject [30], and reviewed by Bertrand et al. [31,32]. This is not new in forensic science: carbon coating before the scanning electron microscopy (SEM) examination is common in forensic analysis of gunshot residue (GSR) to increase the electrical conductivity of the specimen and to eliminate its charging [33].

Christopher et al. [10] carried out a damage test to demonstrate the non-destructive nature of IBA for GSR analysis. They focused on one GSR particle a 4.5–5 µm wide 2.5 MeV ^{1}H beam with a 3 nA current (considerably higher than what is required in routine experiments) for an 850 min long (compared to a few minutes in routine analysis) point analysis. SEM images and PIXE data after this experiment done at unrealistically extreme conditions showed no measurable compromising or degradation of the sample. The case of IBA applied forensically to the problem of authenticating paintings is also important and sketched by Jeynes [34]. The method used must also evaluate how the particular sample responds to the beam. Precisely how “zinc white” (zinc oxide) behaves under a 3 MeV proton beam is analysed by Beck et al. [35], and Zucchiatti et al. have recently provided a systematic and very useful treatment of a variety of damage mechanisms [36,37].

The IAEA has energetically supported IBA, publishing TECDOCs on the fundamentals and applications of elastic scattering [38], PIXE [39], and PIGE [40] and a technical report series [41] for improving the “Reliability and Accuracy of Heavy Ion Beam Analysis”, and promoting the use of NATs in forensic science [42]. Historically the three types of IBA have been applied to different types of samples using different types of data handling, but more recently software has been developed to exploit the synergy between the types: although this synergy has always been recognised, the necessary software has not been feasible until modern computing power became available. First was Geoff Grimes’ “OMDAQ” software [43], which is optimised for PIXE but uses the elastic scattering signal synergistically for calibration: this was used to spectacular effect recently to correct the metalloprotein database [44]. Barradas & Jeynes’ “DataFurnace” software [45] was made fully synergistic

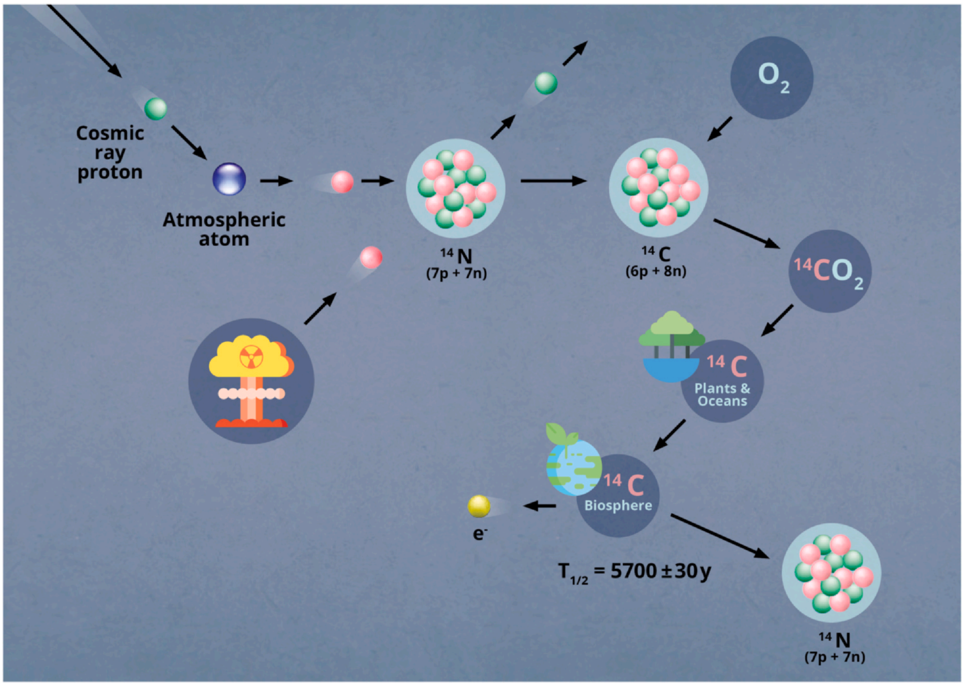

Fig. 3. Schematic representation of the Radiocarbon process. Neutrons originating from cosmic rays or nuclear weapon tests can generate this nuclear chain. Neutrons are depicted in red, protons in green.

(elastic scattering, PIXE, PIGE) in 2008 [46] with further development to support in-air IBA in 2020 [47]. The only other synergistic IBA software currently available now is Tiago Silva's powerful "MultiSIMNRA" [48], which as yet does not handle PIXE.

Accelerator mass spectrometry (AMS) enables the separation of isotopic masses and detection of natural abundance of isotopes. Radiocarbon dating is based on the detection of ^{14}C isotope (radiocarbon), which is typically carried out nowadays by Accelerator Mass Spectrometry [49]. In AMS radiocarbon dating, the ^{14}C content is directly measured relative to the two stable ^{12}C and ^{13}C carbon isotopes. This technique is especially useful to determine the age of objects including their organic components (and hence their chronology). The ^{14}C half-life of 5700 ± 30 years, makes it possible to determine the age over the past 55,000 years. A schematic representation of the Radiocarbon process is given in Fig. 3.

Radiocarbon dating is routinely used for dating of heritage objects. Its successful application for forensic cases is based on the fact that, between 1955 and 1963, nuclear weapon tests above ground level generated high concentrations of radioactive carbon (^{14}C) in the Earth's atmosphere. Cosmic ray interactions with atmospheric ^{14}N produces ^{14}C, which oxidizes to CO_2. The resulting ^{14}C content in the atmosphere remains at the natural level. However, the nuclear weapon tests above ground level during the 1950s and early 1960s produced a rapid increase in ^{14}C.

Since the Nuclear Weapon Ban Treaty came into effect in 1963, the atmospheric levels of ^{14}C have begun to fall as the radioactivity passed from the atmosphere into the oceans and the biosphere. This effect is known as "bomb-peak" [50]. As a result, all organisms that lived since the 1950s contain higher levels of ^{14}C than those that lived before that date. The rapid year-to-year changes in the atmospheric levels of ^{14}C since the 1950s, depending on the moments at which the atmospheric explosions occurred, combined with the rapid transfer of atmospheric ^{14}C, allows the determination of the age of traces that include an organic component, such as soft tissue, food or art objects [11]. This method is called bomb-pulse dating or radiocarbon dating using the ^{14}C "bomb-peak".

The precision of radiocarbon dating is determined by the minimum amount of carbon for which the isotopic ratio $^{14}C/^{12}C$ can be measured. The development of AMS allows an enormous reduction in the required amount of organic carbon in the sample, from several grams with previously employed methods (beta counting), to around 1 milligram with AMS. A further reduction into the few μg to tens of μg carbon range has recently been achieved based on instrumental developments mainly of ion sources. Hodgins showed that this one million factor enhancement in sensitivity is determinant for retrospective birth dating of human cells [11].

Radiocarbon dating requires sampling of the material and applying chemical pre-treatment process, combustion and graphitization [51]. The typical target size of carbon needed for standard ^{14}C dating with AMS is about 1 mg. But with an optimized sample preparation process, 10 μg of sample may sometimes be sufficient for the analysis [52,53].

Radiocarbon dating is a very powerful method to determine the age of a sample and thus provide information on its origin and authenticity. As the sample is fully destroyed during the chemical process, one should consider its applicability for forensics cases. The fact that only a minimum amount of sample is needed for the analysis means that the trace evidence usually remains reasonably intact after sampling. Sampling needs to be done with an extreme care not to contaminate the sample and still get a sample representative to the whole evidence.

3. Forensic applications of neutron techniques

3.1. Forensic applications of NAA

In 1954, J. R. Oppenheimer contacted Richard Dodson to propose the use of NAA to determine the provenance of archaeological materials. Following Oppenheimer's idea, Sayre and Dodson published the results of their experimental work on pottery in 1957 on the *American Journal of Archaeology* [54].

In 1964 Tuckerman et al. published an article describing the analysis of trace element in drugs [55] and two years later the first review about the application of neutron activation analysis to forensic science was published by Coleman in the *Journal of the Forensic Science Society* [56]. According to Lyon et al., the first results of activation analyses accepted as evidence in court in the U. S. were reported by Pro et al. who analyzed samples of soil to identify illicit distilleries [57]. Lyon et al. reported also interesting forensic applications of activation analysis in firearms, inks, plastics, and automobile grease, tire rubber, and paint, including car paints [58].

The First International Conference on Forensic Activation Analysis (FAA) was held in San Diego, California, in 1966 [59].

In 1971 the IAEA published the proceedings of an informal study group meeting held the year before in Bangkok, where Guinn reported about forensic application of activation analysis, including human hair; paint; drugs such as opium, marijuana, heroin; glass; oils and greases, including crude oils, fuel oils, lube oils, grease lubricants; whisky; soil; bullet lead; tapes; paper; rubber; wood; plastics; inks [60].

Some type of forensic activation analysis popular in the seventies of the past century, such as lead fragments from bullet [61,62] and gunshot residue [63], are no longer suitable in court. Other analyses, such as chemical profiling of drugs of abuse is still very valuable in forensic science. In 1977 Henke described the use of neutron activation to analyse rare-earth elements in samples of opium and cannabis [64]. NAA also has advantages relating to sampling uncertainties since large samples can be analysed [65]. More recently, it has been shown that multivariate statistical analysis can elucidate some of the underlying geochemical factors that could help to determine the provenance of Jamaican cannabis using NAA data [66].

In the last twenty years it is still possible to find publications about chemical profiling of drugs of abuse by NAA: Zhang et.al. studied illicit heroin by neutron activation analysis and grouped sixty-two analysed heroin specimens in two clusters associated to their geographical origins [67]. Ebrahimi et al. analysed opium, hashish and ecstasy pills by both NAA and PIXE [68]. Nowadays drugs of abuse are not the only chemical substances threatening public health worldwide. Illegal pharmaceutical products are trafficked by criminal organisations. The products seized must be analysed to look for toxic chemical substances, to provide evidence in court about their illegal nature and to support the inference of similarities between seized specimens [69–71]. Romolo et al. recently used NAA to analyse authentic Viagra® and illegal products containing sildenafil [72,73]. Melkegna and Jonah analysed medicinal plants in Ethiopia [74].

Another global, trans-national criminal issue is food fraud [75] or food crime [76], impacting both national economies and consumers' health and trust. Recent estimated by the European Commission indicate that the yearly cost of food fraud to the global food industry and to consumers is around 30 billion Euros [77]. Singh et al. published an article about trace elements in Indian cereals, vegetables and spices by INAA in 2006 [78] and Rahman et al. studied possible health risk due to chemical elements in common spices of Bangladesh by INAA and AAS techniques in 2018 [79]. Antoine et al. studied the determination of the growing geographic area of origin of Jamaican and international coffee by NAA [80]. Messaoudi et al. used NAA too to analyse Arabica and Robusta coffee beans samples consumed in Algeria [81]. The National Institute of Nutrition reported mineral nutrients and trace elemental compositions of Indian foods, including spices [82]. There are cases reported of spices adulterated with spurious material to reduce the cost [83]. Therefore, analysis of both essential and toxic elements not only in spices is very important and can be effectively conducted by INAA [84–86]. An example of illegal and toxic additive in spices is lead chromate, used as colouring matter in turmeric products [87]. Arpita Datta et al. demonstrated in 2020 that INAA is a suitable method for turmeric quality assurance [88].

Another important forensic trace useful in criminal investigation is glass. Fragments can be produced in many criminal activities, from burglaries to hit-and-run car accidents. Sharma et al. determined major, minor and trace concentrations of fourteen elements in five soda-lime (automobile) glass samples by conventional INAA [89]. Major elements allowed establishing the class of glass samples, whereas concentration of Al, Sc, Fe, Co, Hf, La and Ce and concentration ratios like Al/Sc, La/Sc and Ce/Sc showed promising perspectives for forensic applications [90].

NAA was used to study forensic issues involving geology, such as the theft of sand from the Coral Springs beach in Jamaica. Statistical analysis of nine elements (Al, Ca, Ce, Cr, Dy, Fe, Mn, Sc and Sr) out of the thirty-five that were measured could lead to the identification of the originating and receiving beaches of the stolen sand [91].

NAA can also be applied in forensic toxicology. Kučera et al. in 2018 reviewed the analysis of elements in hair by NAA and particle induced X-ray emission (PIXE), showing how NAA provides accurate results of toxic elements [92,93].

In the five decades since NAA started to be applied to forensic sciences, it has become an established technique worldwide. The physical principles that underpin it are well known and understood, and procedures for calibrations, sample preparation, irradiation and counting are well developed, including quality assurance and quality control methodologies. At the same time, important developments have taken place, that have expanded the range of applications for NAA.

One of these developments is Large Sample NAA, an innovative variation of NAA that, uniquely among analytical techniques, can measure the bulk composition of large samples (up to kilograms in mass) non-destructively [94]. Applications of LSNAA include large objects with irregular shapes, such as cultural heritage artefacts, rock samples, large samples of assorted ore, and finished materials and their parts [95,96].

In the last decade, strong progress in automation of the entire NAA process also took place, with the definition of a framework that brought together the different building blocks of NAA [97]. This included automation in the integrated process and data management, sample loading and changing, irradiation, counting, and data analysis. Automation not only increases the analysis capacity of a laboratory, and hence potentially the cost of analysis per sample, it also leads to shorter processing and turn-around times. Finally, the reduced human intervention in the NAA process reduces the scope for human error during the analytical process.

3.2. Forensic applications of other neutron-based techniques

Other neutron-based techniques such as neutron scattering and neutron imaging have been sporadically applied in forensic sciences.

In forensic anthropology, one outstanding issue is to analyse bone remnants that have been affected by heat, such as due to fire events, or other damage that leads to strong changes in the bone structure and composition. These changes can affect the results of techniques used to determine the circumstances and time of death based on the degradation of the bone, reducing their reliability. Neutron scattering is highly sensitive to hydrogen and can probe large fragments (several grams), and therefore is ideally suited to probe changes in hydrogen bonding patterns in representative bone samples. A recent study demonstrated the applicability of neutron scattering to the analysis of different types of burned human bone, revealing heat-induced changes in the bone structure and chemistry [98–100].

Forensic engineering is dedicated to the investigation of failures of products or their parts, that have a possible civil or criminal forensics consequence, often involving determination of liability for the failure, and also to establish corrective and preventive measures. Analysis of residual stress in failed metallic components is particularly difficult, because the stress is relieved by the fracture. It has been recently showed that neutron diffraction can be used to address this problem for brittle fractures in aluminium, and the same method can be used for other metals as well [95,101].

Neutron imaging has been extensively applied in cultural heritage studies, often directed at a better understanding of manufacturing and preservation techniques [102,103], with some potential forensic applications.

Other applications of neutron imaging are also relevant to forensic science. An innovative method to determine the post mortem interval using neutron radiography has been recently proposed, with a proof-of-concept study made using dog cadavers [104]. The study used the loss of water and therefore hydrogen content after death to correlate the increase in neutron transmission with the time after death.

Combined fast-neutron and gamma-ray radiography has been used in the interrogation of air cargo containers to produce compound images that represent both the density and composition of the contents of a container. The composition information greatly was found to be particularly advantageous in the detection of organic materials with irregular format such as concealed narcotics and explosives [105].

Techniques combining neutron and X-ray interrogation for explosives detection have been proposed since the 1950s and continue to be explored due to their potential for detection of concealed bulk explosives such as landmines, improvised explosive devices and unexploded explosive ordnance. A review of previous and current research has been recently published [106].

4. Forensic applications of IBA

IBA is important for layered objects, such as authentication of paintings and other precious objects [107,108], characterisation of documents or identification of car glasses involved in hit and run or other accidents [109,110]. IBA is often used in identification of artefacts and forgeries [111], and showed to be very powerful in gunshot residue (GSR) analysis.

IBA allows in most approaches not only elemental analysis of the surface of a specimen (and molecular analysis with SIMS or MeV-SIMS) but can also provide images with resolution down to about 1 µm, showing both morphological features of the surface using a Secondary Electron (SE) detector (the same used in the more traditional scanning electron microscopy) and elemental imaging (PIXE, PIGE, …) which, based on correlations, can also provide chemical information. It is therefore possible to group forensic applications of IBA into “bulk” analysis and chemical imaging (depending on whether lateral imaging of the sample is of interest (bearing in mind that the information depth of the technique does not usually much exceed about 10 µm) based on the major aim of the analysis.

Typical bulk applications may be the analysis of drugs, food, and spices: a mg-sized sample is dried, powdered and homogenised such that the thin near-surface region may be taken as representative of the whole. Akter et al. found potentially toxic levels of heavy metals in domestic spices by PIXE [112]. Giulian et al. analysed elemental profiles of commercial mate tea leaves in 2007 [113], dos Santos et al. used PIXE to characterize Brazilian wines [114,115], Ebrahimi Fakhar studied trace elements in opium, hashish and ecstasy pills not only by NAA but also PIXE in 2012 [63], Debastiani et al. reported a survey of coffee compositions by PIXE, with the matrix composition determined by RBS [116–118], Chytry et al. demonstrated the potential of accelerator-based techniques as an analytical tool for forensics in determining the provenance of Brazilian and Jamaican coffee using PIXE, AMS and (Fourier Transform Infrared [119], and Romolo et al. compared IBA and instrumental NAA for forensic characterisation of authentic Viagra® and of sildenafil-based illegal products [68], and Mestria et al. developed a forensic procedure based on IBA, gas chromatography–mass spectrometry and high performance liquid chromatography-high resolution mass spectrometry, to analyse products containing sildenafil or the doping agent oxandrolone [120].

Imaging capabilities are particularly welcome in forensic science for small traces such as glass fragments and gunshot residue particles (GSR). When Sen et al. published an article describing for the first time the use of PIXE to analyse GSR particles in 1982, the size of the beam spot (1 mm^2) was unsuitable for using in casework [121]. Later Niewohner and Wenz studied the internal morphology of GSR particles from different types of ammunition after cross-sectioning them by a focused ion beam (FIB) of gallium [122]. Bailey et al. in 2009 published two articles, showing how IBA can be used to further characterise GSR particles after SEM analysis. In the first article they showed images of the GSR particles of about 10 µm obtained by X-ray and backscattered particle using 2.5 MeV protons and found that the use of PIXE resulted in signals with higher intensities than the ones obtained by SEM/EDS [123]. In the second article Bailey and Jeynes demonstrated that it is possible to make quantitative analysis of a single GSR particle by RBS [124]. Romolo et al. later published a procedure to relocate and analyse by IBA some GSR particles already analysed by SEM-EDS following the forensic procedure adopted in casework in Italy. In some cases PIXE allowed the detection of elements at much lower levels than is possible using SEM-EDS and showed for the first time that B in micron size GSR particles can be identified by PIGE. This information is particularly relevant in cases when ammunition without heavy metal such as Pb, Sb and Ba in the primer are used. In a particle from CBC Magtech CleanRange SEM-EDS allowed the identification of only 5 elements (Na, K, Si, Ca and Al), PIXE allowed the identification of Fe, Ni, Cu, Zn, Ba and Cl, not detected by SEM-EDS, and PIGE identified B [125]. Another feature of IBA when used in GSR particle analysis is the improved capability of providing quantitative analysis and to study the population of GSR particle by chemometrics [9]. Duarte et al. did not limit their IBA analysis solely to GSR particles but also analysed bullets, jackets, cases, propellants and primers of three different ammunitions manufactured by Companhia Brasileira de Cartuchos (CBC) [126,127]. Romolo et al. studied the use of PIXE to overcome a typical issue in forensic analysis of GSR particle by SEM-EDS, due to the overlap of Sb and Sn peaks [128]. Lucas et al. recently reported their ion microbeam results after sectioning GSR particles by FIB and mapping the resulting internal surface by SEM–EDS [129].

Imaging capabilities are also important during forensic examination of documents. The group at Ruđer Bošković Institute in Zagreb used MeV-SIMS to characterise inks and PIXE and to determine the deposition order of inks on paper [130,131]. MeV-SIMS was utilised for the analysis of different colorants (dyes and synthetic organic pigments), and it was shown that the order in which superposing lines were written could be identified, in cases where optical methods were unable to resolve the sequence.

Calligaro and Dran published a review of art and archaeology applications of IBA, focusing of the technique’s capabilities in authentication of heritage items and presenting a comparison with other techniques such as SEM/EDX, XRF, Raman, and others [132]. They presented several examples, including how ERD could be used to determine that a human quartz skull of alleged pre-Columbian origin, conserved in the Musée du Quai Branly, Paris, France, was not pre-Columbian, but a fake produced in the late 19th or early 20th century [133]. They also present an example of large-scale chemical imaging with PIXE of Renaissance masterwork painting Madonna col Bambino by Andrea Mantegna, conserved in the Accademia Carrara in Bergamo, Italy, and show that unique information is obtained, not accessible by standard techniques (XRF in this case). Calligaro et al.

have also shown how a combination of IBA techniques, including EBS, PIXE and MeV-SIMS with AMS and Optical Photothermal Infrared Spectroscopy imaging allowed them to reconstruct the history of an historical painting [134].

Finally PIXE provided useful information for forensic toxicology by spatially resolved analysis of elements in hair [135].

5. Forensic applications of AMS

Forensic applications of AMS utilize its unique capability to determine the age of a sample, and consequently its chronology. Tuniz et al. provided an overview on the fundamental application fields of AMS in various forensic problems [136] in 2004. Since then, these application areas have become more relevant and, on several occasions, AMS has been applied in criminal cases. Some care is required in interpretation of the data, because the ^{14}C concentration intercepts the “bomb peak” at least twice (and in some cases several times due to the yearly fluctuations in atmospheric ^{14}C concentration), Strictly speaking, the dates determined are ambiguous, and usually additional information needs to be taken into account to reach final conclusions. Nevertheless, AMS has proved essential to solve several forensic cases.

Application of AMS for forensic anthropology is based on the fact that ^{14}C is incorporated into biological material through the metabolism of living beings, including humans, animals and plants. This incorporation stops when the organism dies. Therefore, the ^{14}C content within the tissue, reflects the ^{14}C level that existed in the atmosphere before and at the time of death. It is possible to establish a correlation between the ^{14}C concentration of tree rings and human body samples formed in the same time intervals, allowing the determination of calendar year of e.g. birth or death. For such cases from the near past radiocarbon dating based on the “bomb-peak” is applied.

This technique has been developed and explored over the years for the age determination of a person alive or post mortem. Usually small samples of human tissues, bones, tooth or hair are analysed. Nakamura et al. applied radiocarbon dating in criminal investigations of a murder case [137]. Crucially, the ^{14}C concentration in hair and tooth could be measured. They were different, because hair reflects the ^{14}C atmospheric concentration in the year of death, while tooth has the ^{14}C concentration corresponding to the time at which the tooth was fully formed. In this way, both the time of death and the age of the victim were identified. The measurements were consistent with the ones confessed by the murderers.

The same technique was applied by Marzaioli et al. for an unsolved case investigated by the Rome prosecutor office [138]. Radiocarbon dating was also used for the identification of a missing person by Wild et al. [139]. In other cases, the death and the year of birth of unidentified human remains as victims of war were determined by Hong et al. [140]. Ubelaker et al. [141] and Solis et al. [142] determined the age of a person based on a ^{14}C content in human teeth. Both enamel and collagen were used to measure the ^{14}C atmospheric concentration during its formation allowing the calculation of the actual year of birth with high precision. Kjeldsen et al. [143] developed a method to measure the formation age of human eye-lens crystal-lines, based on the fact that the radiocarbon ages reflect the time of birth. With this method, the date of birth of deceased humans can be determined from the eye-lenses with an accuracy of a few years. Brock et al. [144] reviewed forensic radiocarbon dating of human remains, and Calcagnile et al. [145] discussed the applicability of the analysis of various parts of a human skeletal remains for forensics medicine.

Wildlife trafficking is a serious environmental crime. It involves all stages of the supply chain from poaching protected or restricted animals or plants from their habitats, to smuggling, trading, importing, exporting, etc. Although estimates are difficult, according to Europol the profit from trafficking of endangered species could be more than 4 billion EUR globally in 2011 [146]. Quarta et al. used AMS for ivory dating in [147,148] and discussed its potential to identify illegal trade of samples obtained from endangered animals.

It is possible to detect food counterfeiting with AMS. Honey is among the most commonly forged foods [149]. Varga et al. investigated acacia honey with the radiocarbon and other analytical methods. They found that acacia honey preserves the original carbon isotopic and elemental abundance fingerprint that it had when produced. It can hence be used as an environmental indicator to identify urban pollutants or in precipitation, and emissions from industrial and agricultural activities in the vicinity [150], which all are very useful for environmental forensics, too. In addition, radiocarbon-based age determination coupled with elemental analysis can be used in biological, dietary, archaeological or other multidisciplinary studies as well. Zoppi et al. accurately determined wine vintages and could demonstrate the presence of other chemicals of natural and synthetic origin, not naturally present in wine [45]. In the same publication they also reported the application of AMS to detect drugs of abuse. Based on the ^{14}C bomb-pulse method, they established the time of harvest of illicit drugs such as heroin and opium.

A common type of fraud in art is forgery. Absolute dating of paintings and other art objects is crucial to detect forged artefacts. Authentication of paintings traditionally rely on expert knowledge and understanding of art history, individual painter styles and their evolution, chromatic palette, type of canvas and frame, etc. For many decades it also strongly relies on a range of analytical techniques that can contribute significant information to the investigation. Of these techniques, radiocarbon dating can uniquely provide an absolute time scale, but so far it had been limited to the analysis of organic materials and components in the paintings, such as the canvas, wooden frame, organic binder, etc, while many paints are based on inorganic material. Beck et al. [151] recently developed a novel method to radiocarbon date paintings containing inorganic pigments. Based on the analysis of lead white pigments, they demonstrated the capability to date 14th-16th century wall paintings. Since lead white was extensively used by the greatest artists, it is anticipated that this method can be widely used for the authentication of paintings for museums and the art market.

The very first study applying the “bomb-peak” method to date paintings was carried out by Caforio et al. on a forgery of a Fernand Leger painting [152]. Hendriks et al. [153] identified various modern painting forgeries by analysing the organic component of the binder. They showed that the oil used as binder for the pigments in the forgeries had an excess amount of ^{14}C, which is a fingerprint for the 20th century nuclear bomb tests. One of the most significant conclusions of this study was that ^{14}C dating of the paint layer is a powerful way to identify modern (post-1950) forgeries even when recycled older canvas supports were used.

Radiocarbon dating of the canvas gives a time frame of when the raw fiber material was harvested for the canvas. Brock et al. investigated contemporary artwork [154] and found that generally there is an offset of a few years with the actual completion of the works of art. In addition, radiocarbon dating provided information on the individual artist’s mode of working. It is possible to get information on the canvases, including on their reuse (where the painting on an old canvas is removed and a new forged painting is applied), extensive reworking, their long-term storage, and when a number of related paintings are available, to determine a chronology for their origin.

Under a police investigation authentication of two paintings alleged to be by Impressionist and Pointillist artists were conducted by radiocarbon dating by Beck et al. [155]. The ^{14}C content of the fibres demonstrated that the canvases were manufactured from material originated either in 1956–1957 or after 2000. As the artists that were supposed to have mastered the paintings has passed away in the 1940s, it was demonstrated that these paintings were forgeries.

Radiocarbon dating was applied also to authenticate the sculpture of the Flora wax bust in the Bode Museum, Berlin. This work of art was attributed to Leonardo da Vinci because Flora's face resembles several Leonardo portraits. However, this attribution has been subject to intense debates since the sculpture's acquisition in 1909. Radiocarbon dating analysis of the bust's wax by Reiche et al. [156] concluded that the bust was not made in the Renaissance but in the 19th century.

Nowadays, the antiquities market regularly uses radiocarbon (^{14}C) dating to screen for forged objects. In response to this development, Hajdas et al. proposed that radiocarbon laboratories follow due diligence and best practice approaches for the radiocarbon dating of cultural heritage objects requested by such clients, which are of a different nature than typical research activities [157,158].

6. Access to ion beam accelerator and neutron facilities

Many common non-nuclear analytical techniques use table-top, nearly push-button instruments with fairly low cost of acquisition and cost of ownership, which makes them suitable for wide general use, including in forensic laboratories. In contrast, NATs are implemented through ion beam accelerator [159] or research reactor facilities [160] that are usually operated as national or regional facilities. These are expensive to build and to operate, and further considerations apply to research reactors owing to their being nuclear installations, such as safety and security, and safeguards of nuclear materials. Many of these facilities have infrastructure programmes for access by national users and, in some cases, by international users as well, to whom both analytical and financial support is given. Others provide analytical services to external users, too, either on a joint research or commercial basis. Business models of the analytical services very much depend on the ownership of the facility. In case of governmental institutions these services are usually provided free of charge for other governmental laboratories.

One can conclude that, although neutron sources and accelerator laboratories are not likely to be owned or operated by police laboratories, they are in fact widely available, and their operating organizations are often willing to take on opportunities for new areas of activity such as forensic science. Their highly skilled personnel are capable of providing analytical services (including further extending the application areas), and research and development in the field of forensic science.

Concerns with chain of custody [161] and, in some cases, restrictions on evidence being sent abroad, may restrict use of NATs. However, one needs to keep in mind that research reactors are facilities where control of access is routinely implemented. In many cases, procedures already in place for handling of samples can be easily adapted to guarantee control of the chain of custody. On the other hand, ion beam accelerator facilities, that are commonly used for research (typically with less stringent access control and procedures), may require additional quality assurance to forensic laboratory standards.

7. Conclusions

In this paper, an overview is provided of the capabilities, potential and applications of nuclear analytical techniques to forensic sciences. For the first time, we review forensic applications of research reactor based techniques such as NAA and ion beam accelerator based techniques such as AMS and IBA (where the NATs were essential to provide sufficient information for forensic purposes), also describing some forensic applications and showing some case studies. In this paper we described forensic cases when NATs can provide superior analytical information, such as drugs of abuse; food fraud; counterfeit medicine; illicit trade; gunshot residue; glass fragments; forgery of art objects, other artefacts and documents; wildlife and human material.

It is clear that several common, non-nuclear analytical techniques may have similar (or better) general properties: sensitivity, accessible information depth, depth resolution, and imaging capability. However, in most cases those techniques require sample dissolution or sputtering if a depth profile is required, and usually the evidence object has to be sampled. Many non-nuclear analytical techniques allow the analysis of only very small amounts of material, posing huge problems of representativeness. NAA and neutron imaging can probe entire bulk objects, determining their average concentration and internal structure, respectively. IBA techniques are sensitive for near-surface analysis including sub-layer/embedded layer analysis, and can also be used in a large-area (few mm^2) imaging mode. The possibility of focussing the ion beam down to a micrometer size and scanning it to give a high spatial resolution analysis with mapping can be extremely useful especially for authentication of art objects and documents or (for example) for the characterisation of a single GSR particle. For selected areas of whole objects such as paintings and other works of art, documents and other evidence IBA (including fully synergistic Total-IBA) may be performed with an external beam (in-air). NAA and neutron imaging do not require the samples to be in vacuum, and analysis of entire objects in air is also common.

Summarizing, NATs have the following characteristics that can be an advantage in specific niches of application in forensic sciences:

- the samples remain intact and can be returned and analysed with other techniques (in NAA, after a period that is usually a few months or less);
- no need for sampling (except AMS);
- first principles quantification from well-understood physical phenomena;
- all elements and isotopes accessible;
- depth and lateral profiling (IBA);
- high resolution imaging and point measurement (IBA)
- simultaneous analysis with several techniques (IBA).

With the exception of AMS, the samples are not deliberately destroyed by NATs, although all the NATs are high energy techniques with the potential to modify the sample to at least some extent. In most cases of interest, the damage can be limited to be negligible and subsequent analyses are possible as usually the evidence integrity is kept.

These properties make NATs (apart from AMS) particularly important when the objects to be analysed are 1) unique or precious; 2) cannot be sampled; 3) cannot be destroyed, (for instance if re-analysis or analysis with other techniques are required); and 4) inhomogeneous (in which case applying point/line and 2D scans the analytical data can be defined with a high spatial resolution using IBA). AMS requires increasingly smaller sample mass and has been providing unique dating information in forensics cases.

We have summarised the features of NATs, and reviewed the wide collection of forensic application. Together with the wide availability worldwide of these techniques, their established uniqueness and advantages as well as continuing developments in their capabilities clearly opens up opportunities for an increased use of NATs for routine application in a variety of important forensic problems.

CRediT authorship contribution statement

Aliz Simon and Nuno Pessoa Barradas coordinated the preparation of the manuscript. All the authors contributed to the manuscript. All the authors read and approved the final manuscript.

Declaration of Competing Interest

The authors declare they have no conflicts of interest.

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Annex. Glossary

This appendix provides a summary description of nuclear analytical techniques common in ion beam accelerator laboratories or research reactors (with informative references, mostly not related to forensic applications). It is becoming increasingly clear that a synergistic combination of techniques may be very valuable, as for Total-IBA (*q.v.*).

AMS (Accelerator Mass Spectrometry) enables separation of isotopic masses and detection of extremely low concentration. In case of the radiocarbon method, it is applied to the measurement of $^{14}C/^{12}C$ ratios of order 10^{-12} to 10^{-15} and has also been used in criminal forensics including murder cases [11,162,163]. Heavy ion AMS is especially useful for nuclear forensics, cultural heritage and medicinal applications. AMS is destructive: the samples are vaporised, ionised and accelerated and the accelerator is used as an ultra-high-sensitivity isotope selector.

EBS (Elastic Backscattering Spectrometry) is elastic (non-Rutherford) scattering where the scattering cross-sections no longer follow the analytical Rutherford formula (see **RBS**). It is often used to enhance sensitivity to light elements. See **IBA** and **Total-IBA**.

ERD (Elastic Recoil Detection) with a He ion beam is typically used for H isotope profiling. It relies on the He ion recoiling the lighter target atom, which is directly detected. Depth profiling is limited to around 1 micrometer depth. See **IBA** and **Total-IBA**.

ERDA (Elastic Recoil Detection Analysis) is another commonly used acronym for **ERD**.

HI-ERD (Heavy-Ion ERD) is **ERD** (*q.v.*) with an energetic incident beam heavier than ^4He ($^{37}Cl^{5+}$ for example) and all lighter elements can be detected with good mass resolution, now usually in a ToF (Time of Flight) detector. See **IBA** and **Total-IBA**.

HI-PIXE (Heavy-Ion PIXE) is **PIXE** (*q.v.*) with a heavy-ion beam, usually at much higher energies than standard PIXE. May be used synergistically with **HI-SIMS** (*q.v.*).

HI-SIMS (Heavy-Ion Secondary Ion Mass Spectrometry) is a new technique for the determination and mapping of molecular composition [164]. High energy heavy ions encourage the release from the surface of large molecules. By contrast, standard **SIMS** (with keV-energy ion beams) result in more highly fragmented molecular species. May be used synergistically with **HI-PIXE** (*q.v.*).

IBA (Ion Beam Analysis). Includes any or all of **PIXE**, **HI-PIXE**, **RBS**, **EBS**, **ERD**, **HI-ERD**, **NRA**, **PIGE**, **HI-SIMS** (*q.v.*). All these techniques use an energetic ion beam usually from a linear accelerator with terminal voltage ~1 MV or more. Heavy-ion beams (often with larger accelerators) may also be used. Multiple IBA techniques used synergistically (either concurrently or consecutively) are called **Total-IBA** (*q.v.*).

NAA (Neutron Activation Analysis) uses a neutron beam to activate the sample, with the characteristic (delayed) reaction products subsequently giving the analytical information.

NDP (Neutron Depth Profiling) is capable of depth profiling specific isotopes, with the depth scale from the energy loss of MeV particles escaping from the reaction sites. It is very sensitive to lithium, which is extensively used in the study of lithium-based solid-state-electrolyte high-energy batteries, investigated with operando methods [165].

N-imaging (Neutron Imaging) relies on the absorption of neutrons in the sample similarly to X-ray radiography and computed tomography. However, neutrons are very sensitive to light elements, so the images reveal different features from X-ray methods. As neutrons can also scatter, images based on scattering properties can also be obtained in advanced methods. Stress and internal magnetic fields can be observed in this way.

NRA (Nuclear Reaction Analysis) is an ion beam method with an *inelastic* reaction yielding a characteristic reaction product, often a very energetic proton or alpha particle. It is very sensitive to many light isotopes, and depth profiles can be obtained. See **IBA** and **Total-IBA**.

N-scattering (Neutron Scattering) methods are various. It can be used to probe the structure of matter, in a way complementary to X ray and photon scattering based methods. It has been used, for instance, to study the axe of Ötzi, the 5,300-year-old Tyrolean Iceman natural mummy discovered in the Italian Alps in 1991 [166].

PIXE (Particle Induced X-ray Emission) arises from the same physics as XRF, but where the primary ionising beam consists of ions instead of X-rays. PIXE and XRF spectra are similar, but PIXE often has lower background and higher sensitivity. PIXE has been used at the "alpha particle X-ray spectrometer" on board the Mars Rover to obtain compositions of extra-terrestrial minerals [167]. See **IBA** and **Total-IBA**.

PIGE (Particle Induced Gamma ray Emission) is **NRA** (*q.v.*) where γ-rays are detected. See **IBA** and **Total-IBA**.

RBS (Rutherford Backscattering Spectrometry) relies on an energetic ion beam scattering from a sample. RBS is an energy loss method, so that the backscattered ions provide quantitative information on sample composition as a function of depth. RBS is traceably accurate in principle since the scattering cross-section is analytical (from the approximation of point charges scattering in a Coulomb field). RBS can therefore be used as a primary direct reference method [168] in some cases. See **EBS**.

Total-IBA (Total Ion Beam Analysis) is the synergistic technique [47,169] for non-destructive model-free fully-quantitative thin film elemental depth profiling [170] using any or all of the various **IBA** (*q.v.*) techniques either simultaneously or sequentially. Total-IBA inherits the accuracy of its most accurate technique [171].

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