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Data analysis software package for radionuclide standardization with a digital coincidence counting system

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ABSTRACT

The Nuclear Metrology Laboratory (LMN) – IPEN, São Paulo, Brazil – developed a Digital Coincidence System (DCS), based on the Coincidence Counting Methodology, in order to improve its capabilities in radionuclide primary-standardization. Digital process is implemented in two steps: data-acquisition (a set of measurements) and offline software data-analysis and calculation. The present work shows the basics of the data-acquisition unit (Software Coincidence System – SCS), describes the DCS' data-analysis process and the initial approaches chosen for the implementation of the software package (Coincidence Analyzing Task – CAT). ^{152}Eu standardization, performed for DCS testing, software expansion and validation, is briefly discussed.

1. Introduction

Digital techniques have been employed in Coincidence Methodology for radioactivity measurements with many advantages (see, for instance: Keightley et al., 2013). Following this trend, the Nuclear Metrology Laboratory (Laboratório de Metrologia Nuclear – LMN), IPEN, São Paulo, Brazil, developed a Digital Coincidence System (DCS), in order to improve its radionuclide standardization capabilities. DCS is composed of the data-acquisition unit, called Software Coincidence System – SCS – (Toledo et al., 2008), and the data-analysis software package, called Coincidence Analyzing Task (CAT), subject of the present work.

SCS measurements consist of the sampling and digitization of detection signals. Saved pulse data is constituted by the peak occurrence-times (time stamps) and corresponding amplitudes (pulses height). Independent data files are recorded for each detection channel, such as for beta and gamma signals from a 4πβ-γ detection assembly. The data-set includes files from the radioactive sample(s) measurements and from no-sample measurements, these last ones used for background (BKG) count-rates evaluation.

CAT package includes static-linking libraries and the analysis program, written in C++ language. Libraries provide sets of tools, mainly classes defining useful objects which encapsulate suitable data, such as pulse-info and counting, energy distributions (spectra) and final report (results). The analysis program performs:

- configuration input-file reading, in order to retrieve the paths and names of the target files (pulse input data-files and output-files for processing results) and the analysis setting values, such as dead-time, pulse amplitude discrimination (thresholds for beta pulses and energy-windows for total absorption gamma peaks), gamma pulse delay (in order to match the beta/gamma coincidence distribution centroid) and coincidence resolving time.
- pulse-info reading (time stamps and pulses height are retrieved from data-files);
- pulse selection and counting, according to the given analysis settings mentioned above;
- beta/gamma coincidence counting (within the selected resolving time);
- getting of spectra data (beta and gamma amplitude and coincidence time histograms);
- sample activity calculation by applying correction factors.

The new CAT software replaces classical electronics performing most of the functions of the conventional modules employed in a coincidence system, used for activity measurements.

1.1. Methodology

The Coincidence Counting Methodology (CCM) has been used for decades for developing radionuclide primary standards (Baerg, A. P.,

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1966), with high precision and accuracy. The techniques used in CCM include measurement and calculation procedures to determine the activity of a radioactive sample, from the counting rates of related decay events, such as beta emission and gamma transition, as well as the coincidence rate (beta/gamma event pairs detected simultaneously). Counting rates computation is influenced by some factors such as measurement dead-times, background radiation, radioactive decay, accidental coincidences, and proper corrections must be taken into account.

Linear Extrapolation Method (Baerg, A. P., 1973) is usually employed when the beta counting is influenced by some factors such as the occurrence of internal conversion (IC) electrons or secondary electrons originated from photons. Extrapolation method is accomplished by a set of measurements, in order to obtain the sample activity as a function of beta counting efficiency, ε_β, starting with the maximum possible efficiency and then reducing it progressively. There are two basic ways to reduce ε_β: by inserting polymeric or metallic thin films as low energy electron absorbers; or by increasing the beta pulse amplitude threshold level in order to cut off a larger amount of lower amplitude pulses.

The use of absorbers guarantees the rejection of lower energy electrons, which do not contribute for detection (reducing the beta count) because of their total deposition of energy in collisions inside the absorber material.

The threshold increasing method cuts off the lower amplitude beta pulses, which can be originated not only by low energy but also by high energy electrons, depending on the detector's dimensions, as explained below, considering two different cases:

- (A) For large detectors, all the emitted particles are totally absorbed inside the detection-gas volume and the produced beta pulse-heights are proportional to the particles' emission-energies; the obtained amplitude spectrum then corresponds the particles' actual emission-energies; the rejection of pulses at the beginning of the spectrum (low amplitudes) is equivalent to the use of absorbers;
- (B) For small-size detectors, the range of high energy particles will likely exceed the detector's dimensions and just a fraction of the energy is absorbed inside the detection-gas volume; most of the energy is lost in collisions against the detector's walls, with no contribution to the pulse height formation; therefore, low amplitude pulses are produced and the respective spectrum corresponds to the deposited energy; high and low energy electron pulses are mixed together and those remaining below the threshold level are rejected as well. This effect must be taken into account, and usually Monte Carlo simulations (MCs) are performed in order to find corrective factors.

In practice, the extrapolation curve plots the activity as a function of an inefficiency factor: $(1 - \varepsilon_\beta)/\varepsilon_\beta$, and the extrapolated value when this factor goes to zero, corresponds to the radioactive sample activity. DCS performs the efficiency extrapolation in subsequent programming iterations, by means of progressive increases of the beta pulse height threshold level.

Conventional measurement systems are usually made up of a 4πβ-γ radiation detection assembly (beta and gamma channels), coupled to appropriate modular electronics, for pulse processing, such as: spectroscopy amplifiers; amplitude discriminators; gate and delay units; coincidence unit; counting units. High precision and accuracy can be reached with such systems, however, the measurement process (usually including extrapolation procedures) can take several days to be accomplished, and the success of results depends on a careful previous planning and on precise system setting-up of discrimination levels, delay-times and coincidence resolving time (time-range of acceptable coincidences).

Digital and software techniques (Buckman and Ius, 1996; Havelka et al., 2002) have being used to improve CCM, performing the process in two steps: measurement (data-acquisition) and data-analysis. Data-acquisition consists on digital processing of the pulse-trains, sampling and digitizing beta and gamma detection pulses and recording relevant pulse-info into data-files. Data-analysis is then carried out offline by retrieving, via software, the pulse-info from previously recorded data-files. Since the reconstitution of original detection signals is virtually possible, the main tasks and settings of conventional systems (electronic-modules) are replaced by suitable algorithms (software-modules), in order to set-up a virtual system that performs the necessary amplitude discrimination and time analysis, followed by the determination of pulse and coincidence rates, and the sample activity. The whole system and its settings can be planned and improved, aiming at the optimization of results, for a given data-set.

2. Digital Coincidence Systems

LMN DCS development was based on two known digital systems, referenced above, the Digital Coincidence Counting – DCC– (Buckman and Ius, 1996) and the Software Coincidence Counting – SCC – (Havelka et al., 2002), briefly discussed in this section. DCS is also presented, showing its similarities and peculiarities.

2.1. Digital Coincidence Counting – DCC

DCC (Buckman and Ius, 1996) is a powerful system that performs two-channel detection signals digitalization at a 20 MS/s sampling rate. The whole nuclear pulse (voltage sampling values) is saved into data files as well as its time-stamp. In order to save disk space, irrelevant information of the base line (pulse absence) is discarded.

Data analysis software is accomplished by a virtually assembled system, where electronic modules are replaced by software functions. C++ code is compiled as Dynamic Link Libraries, DLL, which functionality is called inside the graphic user interface, GUI, compiled under the LabVIEW environment (http://www.ni.com/- last access: July

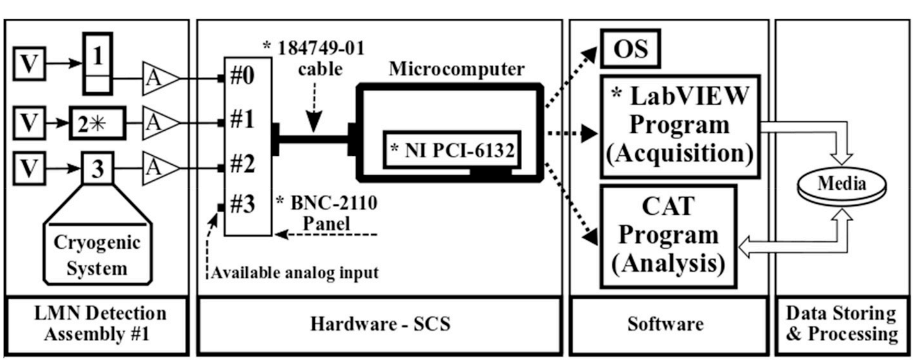

Fig. 1. Block diagram of the Software Coincidence System, SCS (DCS′ data-acquisition unit), showing the main connections, software and data-flux. The signals from LMN 4πβ-γ Detection Assembly #1 are connected, via 184749–01 cable, to BNC-2110 panel and then to the PCI-6132 card, inserted in a Desktop Computer – PCI slot.
Caption:
∗ Radioactive sample
1 NaI(Tl)/PMT gamma detector;
2 Proportional 4πβ counter;
3 HPGe γ detector;
V Detectors' bias voltages;
A Spectroscopy amplifiers;
OS Operational System;
* National Instruments devices and software.

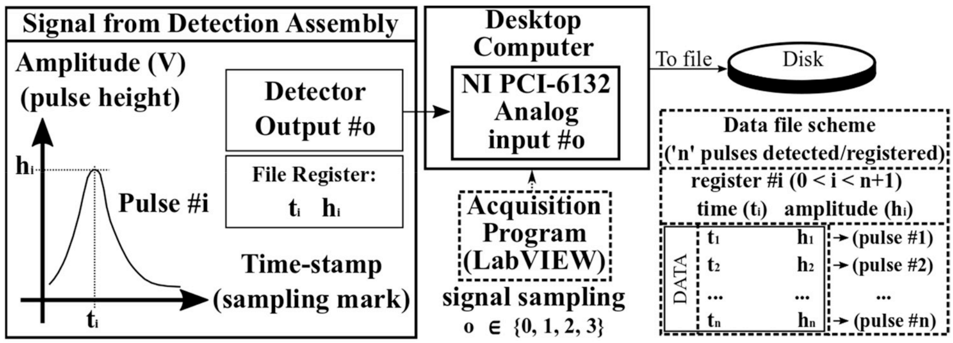

Fig. 2. Graphical representation of a nuclear pulse on the left side of figure shows the pulse-info that composes the two-field pulse-record. On the right side, the file format is schematized. Each file line corresponds to a pulse-record (time stamp and pulse height).

2019).

2.2. Software Coincidence System – SCC

SCC (Havelka et al., 2002) is a four-channel hybrid system, quite similar to conventional assemblies, made up of modular electronics – amplitude discriminators, delay, ADC, and control units. A clock unit provides the time reference and the control to the constant dead-time generation. Most of the tasks are performed by the hardware, by setting pulse amplitude ranges, dead-time (10.0 μs) and coincidence range (2.0 μs). System is connected to a PC via ISA slot; pulses (amplitudes and time stamps) and coincidences that satisfy the hardware defined settings are saved into data files. Therefore, such approach results in simpler analysis software, both for event counting and activity calculation.

2.3. LMN Digital Coincidence System – DCS

The home made DCS is composed of two units: Software Coincidence System (SCS), for data-acquisition; Coincidence Analyzing Task (CAT), the data-analysis software package.

SCS (Toledo et al., 2008) is based on the *Virtual Instrument* (LabVIEW VI) technique (http://www.ni.com/), widely employed to build compact instrumentation, control and supervisory systems.

SCS' block diagram is represented in Fig. 1, showing the employed National Instruments (NI) devices and software (http://www.ni.com/) (with asterisk marks), as listed below:

- Data acquisition card NI PCI-6132 (S Series Multifunction DAQ);
- Connection panel BNC-2110; connection cable 184749–01;
- NI LabVIEW 2010 programming platform.

PCI-6132 card features four analog inputs and, then, up to four detectors can be connected, for simultaneous signals sampling. For instance, Fig. 1 shows a representation of the LMN Detection Assembly #1 with three detectors: NaI(Tl)/gamma; Proportional 4π beta-counter; High-Purity Germanium – HPGe/gamma.

Acquisition configuration and control are achieved with a proper acquisition-program, written in the graphic language LabVIEW. Digitalization of detection pulse train signals is accomplished by internal Peak-detect (NI VI) that extracts the time-stamp and pulse-height for each pulse-signal, from the data sampling. Such pulse info is saved into the corresponding channel data-file, in a two-field pulse-records format (ASCII): time-stamp; pulse height. Time stamps are given by a time-mark obtained from signal reconstitution, in 'Sampling-unit': considering the card sampling-rate of 2.5 MS/s, pulse times in seconds are obtained by multiplying the time-mark by a 4.0×10^{-7} factor (sampling time resolution). Pulse heights are given in 'Volt' unit. Fig. 2 shows a diagram of the pulse-info that constitutes the ASCII file record. Table 1 presents a fragment from an actual data-file. The pulse-info uncertainties are 0.01% for the time marks and 0.6% for the amplitude voltages. Since the PCI-6132 internal buffer has the capacity to store 1 s of data sampling (four-channel-acquisition), data is saved into disk in 1-s blocks.

Table 1
Fragment from an actual measurement data file, obtained with SCS, showing some detection pulse records (lines). Time Stamps are in sampling units, 4×10^{-7} s (sampling frequency: 2.5×10^{6} Hz). Pulse height voltages are given in Volt, obtained from the sampling process by a Peak Detector VI (http://www.ni.com).

Time Stamp (Mark) ($\times 4.0 \times 10^{-7}$ s $\pm$ 0.01%)	Sampled Voltage (V $\pm$ 0.6%)
2640.911765	4.766909
2937.733115	4.434017
11901.047009	1.045080
43586.769373	5.098162
68787.689474	4.631227
78410.492032	4.333210
124296.002183	1.942139

Digitalization process guarantees that all pulses are registered in deadtimeless mode. Only very low amplitude pulses are suppressed for noise rejection and no other previous pulse selection is performed by means of amplitude or time criteria, as needed in conventional systems. Instead, such task is accomplished in the data-analysis step, by the properly implemented algorithms (section 3.2).

DCS requires a single sample-measurement, with the minimum possible electron energy attenuation or electron absorption. Different processing can be performed for the same data-set, by changing the software input parameters, such as amplitude discrimination levels (beta thresholds) or energy windows (total absorption gamma peaks), coincidence-time ranges and software-implemented measurement dead-times. These input values can be changed for attaining the parameterized output, or trimmed to optimize the results. By increasing the energy threshold it is possible to change the beta counting efficiency to obtain the efficiency extrapolation curve (SAD, section 3.2.1: amplitude discrimination changing).

Considering the generality of SCS configuration, pulse-info files allow any sort of time and amplitude analysis to be performed and different measurement systems can be emulated, changing or implementing new features into the analysis software. For example, an extending dead-time module is scheduled as a future goal, since CAT's current version only provides non-extending measurement dead-time module (section 3.2.2). Available analog input channels can be treated separately, such as for Pulse Height Analysis (PHA, as performed by Multi Channel Analyzers – MCA), or in groups, as needed in nuclear Coincidence Counting (α-γ, β-γ, γ-γ, etc.). Analyses of non-nuclear events are also possible.

3. Coincidence Analyzing Task package – CAT package

The software package – Coincidence Analyzing Task, CAT – is developed in C++ language under the 'Object Oriented Programming' paradigm (OOP), to perform the DCS′ data-analysis step. The following subsections present the relevant data structuring, algorithms and program operation.

3.1. CAT data structures, classes and hierarchies

Static-linking libraries provide useful functionality and software tools, including data structures and algorithms. Data structures (objects defined by classes' hierarchies) provide suitable encapsulation for the variables, from nuclear pulse information to program outputs and results. Such structures were thought to have strict relationship with the data base set under development, from which common information will be shared by other applications, such as nuclear data, sample catalog, measurement and analysis configurations and result reports.

Simplified graphical representations of the main classes are presented, using the (Unified Modelling Language – UML) – (http://www.uml.org/- last access: July 2019). Some types, templates and containers, such as '**QDateTime**', '**QString**' and '**QVector**' are defined inside Qt software-development environment (https://www.qt.io/- last access: July 2019).

3.1.1. Physical Quantities library

The 'Physical Quantities' general purpose library defines a set of quantities composed of a value, its uncertainty (standard-deviation), unit and prefix, according to recommendations of the International System of Units – SI) – (http://www.bipm.org/en/si/- last access: July 2019). Math operators and functions defined in 'cmath' C++ header' are overloaded in order to include uncertainty propagation functionality. Class hierarchy includes the base type, **Quantity** that defines the general quantity (value & uncertainty), and specializations, as follows: **ExactQuantity** defines an exact quantity object, with null uncertainty; **PoissonQuantity** establishes a non-negative Poisson-count, which uncertainty is the square root of the count-value (Poisson standard-deviation). **Unit** and **Prefix** classes define the unit and a prefix (SI), associated to quantity-objects.

3.1.2. System Structures library

'System Structures' library is a general purpose tool set to describe an entire system. Class **Descriptor** encapsulates alphanumeric strings (identifier numbers or codes, manufacturer's name, model number, etc.) and date objects (fabrication, assembling or purchasing dates; etc.) to characterize the system's elements or components. **Element** instances point to their respective descriptors and to their parent elements, establishing the parent/children tree or system hierarchy. Derived class **System** allows 'in-software' system assembling, by means of a vector of pointers to **Element** objects. Such structuring tools were thought for in-software systems assembling and describing, for the generation of the output-report files. Detection assemblies and its elements (detectors and electronics) can be described as systems and, together with other elements, such as radioactive samples and software-modules, constitute a larger system layout, such as for DCS description: SCS and CAT configurations and parameters.

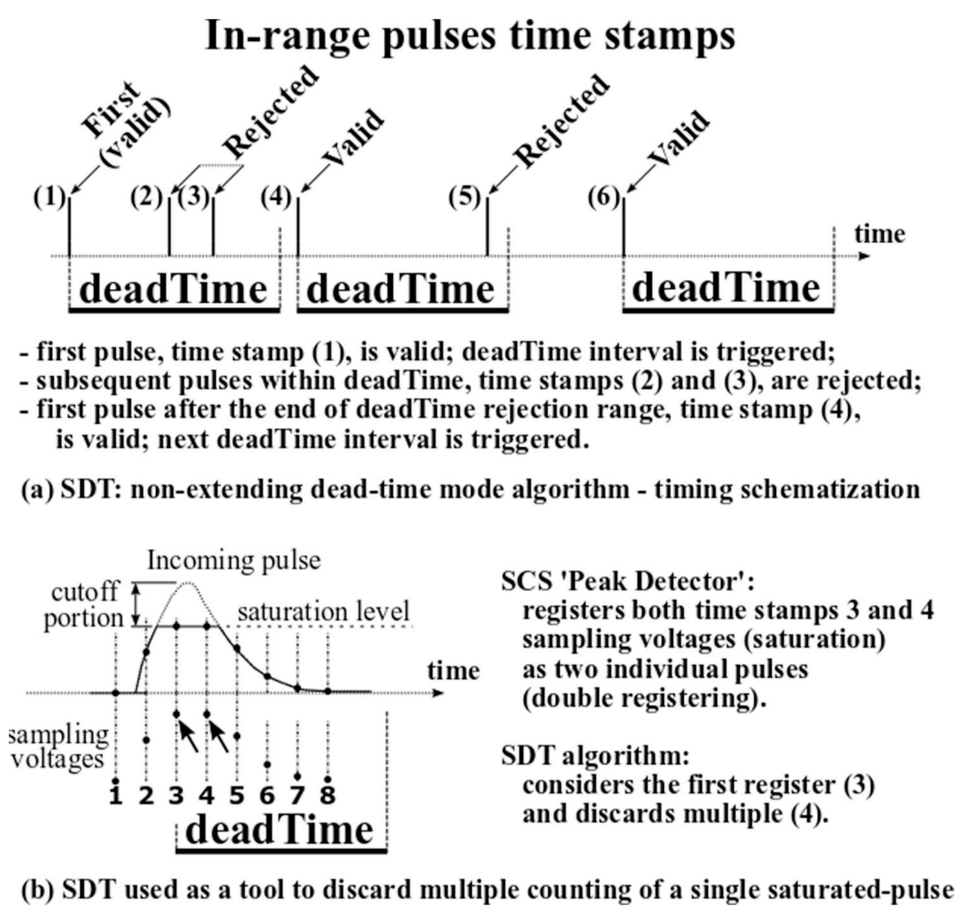

Fig. 3. Schematization of the Software Dead Time algorithm (SDT):
(a) Timing for the implemented non-extending mode dead-time;
(b) Diagram for saturation pulse with doubled registering and multiple counting rejection by SDT.

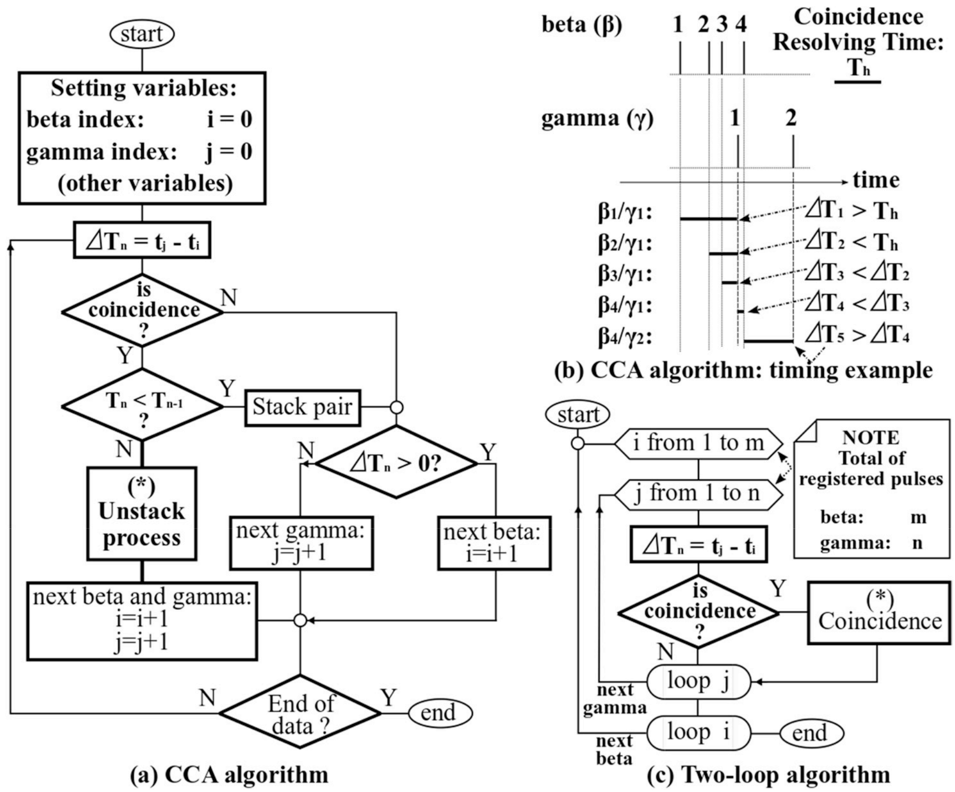

Fig. 4. Simplified flowcharts of the algorithms for coincidence counting:
(a) Coincidence Counting Algorithm – CCA;
(b) CCA timing example, showing β and γ pulses arriving times;
absolute time values (T_h, ΔT_i) are considered; procedure starts with β1 and γ1:
$\Delta T_1 = |t_{\gamma 1} - t_{\beta 1}| > T_h \Rightarrow$ not a coincidence (no stacking); $t_{\beta 1} < t_{\gamma 1} \Rightarrow$ next beta (β2);
$\Delta T_2 = |t_{\gamma 1} - t_{\beta 2}| < T_h \Rightarrow$ coincidence candidate $\Rightarrow$ stack; $t_{\beta 2} < t_{\gamma 1} \Rightarrow$ next beta (β3);
$\Delta T_3 = |t_{\gamma 1} - t_{\beta 3}| < T_h$ and $\Delta T_3 < \Delta T_2 \Rightarrow$ stack; $t_{\beta 3} < t_{\gamma 1} \Rightarrow$ next beta (β4);
$\Delta T_4 = |t_{\gamma 1} - t_{\beta 4}| < T_h$ and $\Delta T_4 < \Delta T_3 \Rightarrow$ stack; $t_{\beta 4} > t_{\gamma 1} \Rightarrow$ next gamma (γ2);
$\Delta T_5 = |t_{\gamma 2} - t_{\beta 4}| < T_h$ and $\Delta T_5 > \Delta T_4 \Rightarrow$ end of stacking $\Rightarrow$
$\Rightarrow \Delta T_4$ chosen as coincidence (minimum value);
$\Rightarrow$ previous stacked values also refer to γ1 and then are discarded;
$\Rightarrow t_{\beta 4} < t_{\gamma 2} \Rightarrow$ increase β index (next pair: β5 and γ2) $\Rightarrow$ restart procedure.
(c) Two-loop beta-gamma iteration algorithm.

3.1.3. *Chemistry and nuclear data base library*

'Chemistry and Nuclear Data Base' classes define chemical elements, isotopes, radioisotopes, decay-constants, radioactive sources and samples, respectively: **ChemicalElement**; **Isotope**; **Radioisotope**; **Constants**; **Source** and **Sample**. Classes **IsotopeX**, **Isotopes** and the singleton **IsotopesTable** are used for instantiation of the table containing relevant nuclear data of isotopes: Atomic Number, Isotopic Mass; name; symbol; constants.

3.1.4. *CAT specific data structure and classes*

Quantities and parameters related to Coincidence Counting Methodology (Baerg, 1966, 1973) are structured into a set of program variables defined in CAT library. Such I/O data hierarchy includes: pulse register; pulse amplitude and time distributions (spectra); pulse and coincidence counts (**PoissonQuantity**) and count-rates; amplitude-ranges or Regions of Interest – ROIs; target sample characterization; etc. Some auxiliary classes and classes' members (intentionally omitted for simplicity) provide setting variables for data acquisition and analysis characterization, including: data-file names; measurement and reference dates; amplitude discrimination ranges; dead-time values; coincidence-time resolution.

3.2. *CAT algorithms*

CAT algorithms, called 'software-modules', provide suitable program functionality, as replacements to 'electronic-modules', commonly used in conventional measurement systems. Such modules are controlled by setting some parameters defined in CAT library, and properly 'interconnected' (program flux or sequence of the algorithms). Then, different measurement system configurations can be emulated and the parameters can be chosen or trimmed within wide setting ranges.

The main CAT 'Software-modules' are:

- Software Amplitude Discrimination (SAD);
- Software Dead-Time (SDT);
- Coincidence Counting Algorithm (CCA).

The current status of the software-modules implementation is presented in the following subsections and the relevant initial simplifications and approaches are discussed.

3.2.1. *Software Amplitude Discrimination – SAD*

SAD module performs the selection of pulses, according to their amplitudes. Multiple ranges of amplitudes or threshold-levels can be freely set by the user, both for beta and gamma channels. 'In-range' pulses are processed, discarding the remaining ones ('out-of-range'). Multiple beta threshold-levels or intervals can be set in order to obtain the extrapolation curve, or the activity as a function of beta counting efficiency. Results can be obtained, iteratively, for different gamma transitions, or pulse amplitude intervals (total absorption peaks).

3.2.2. *Software Dead-Time – SDT*

Dead-time is an intrinsic measurement systems' feature, in principle undesirable, since it causes pulse counting losses. Basically, dead-time is a time interval within which an arriving pulse is processed and the system becomes unable to register other incoming pulses.

Differently, the SCS sampling process is deadtimeless since all the pulses within a time window are registered. However, SDT was originally created to discard multiple counting of single saturated-pulses, which have to be properly computed in the beta channel in order to maximize the beta efficiency counting. Multiple registering of saturation pulses occurs because of their trapezoid shaping (amplifier output saturation), as an intrinsic pulse registering characteristic of the Peak Detector (PD), a LabVIEW built-in Virtual Instrument (VI), used by the SCS acquisition program. PD extracts the time-stamp and the pulse-height information from the signal sampling (signal reconstitution). Usually two or three samplings occur within the flat saturation region, which are interpreted as independent pulses by the PD. Fig. 3 a shows the timing of the non-extending mode SDT algorithm and Fig. 3 b

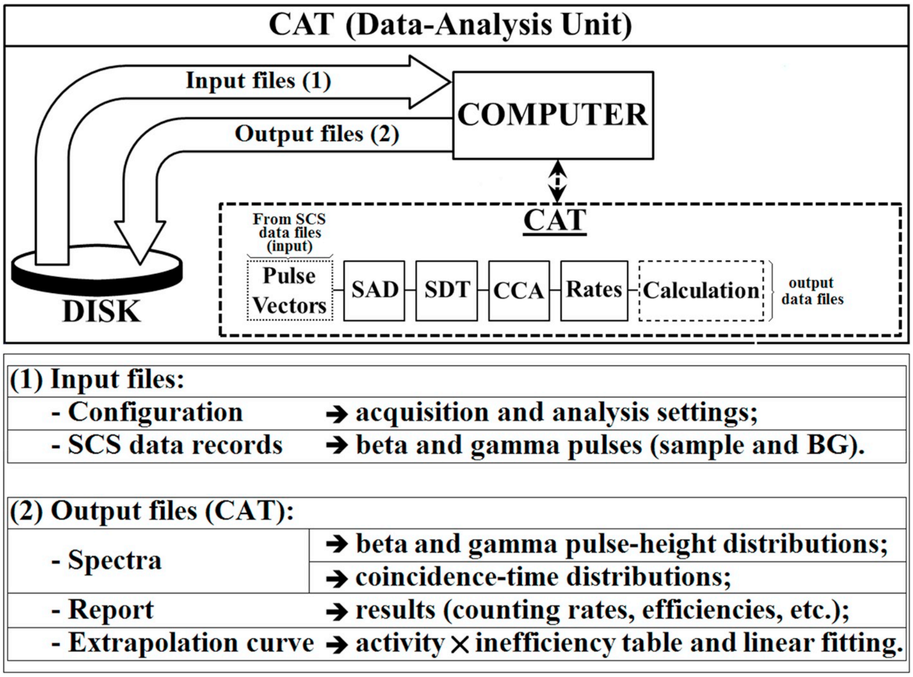

Fig. 5. –Simplified diagram of CAT analysis-program operation. SCS data files are read and processed to obtain the pulse rates, amplitude spectra and the points for the extrapolation curve.
Caption:
SAD Software Amplitude Discriminator;
SDT Software Dead-Time;
CCA Coincidence Counting Algorithm.

illustrates the use of SDT to discard multiple counting of single saturated-pulses. In practice, the minimum stablished dead-time value to discard multiple counting is about 1.5 μs and larger values (2.0 or 3.0 μs) were adopted for safety (eventually, if needed, higher values, in the order of milliseconds, also can be set).

The dead-time values as above were also adopted as a first approach for the compensation of the counting losses caused by the pulse pileup effect. In such case, pulse-height distortions caused by peak overlap were considered insignificant, for the following reasons:

- current DCS version uses a low sampling rate (2.5 MS/s) and was thought to perform low rate measurements, in the order of a few thousands of becquerel;
- pileup affects more significantly the beta channel, because of its high counting efficiency, and only the counting losses are considered, since the whole energy spectrum is computed; usually only low energy threshold discrimination is performed in order to obtain the beta efficiency extrapolation curve;
- much lower gamma channel counting efficiencies results in lower counting rates, reducing significantly the probability of pileup occurrence and its effect on the gamma energy windows were neglected.

As shown in Fig. 3, SDT module imposes an artificial non-extending type dead-time on the in-range pulses. Out-of-range pulses also contribute for the system's dead-time; however, such class of pulses and respective loss are included in the detection counting efficiency calculation.

Time considerations were also done, referring to the sampling time and pulse time: as mentioned in 2.3, the sampling resolution is 4.0×10^{-7} s and time uncertainty is 0.01%, or 4.0×10^{-11} s, which can be considered insignificant for SDT computations; the main requirement for the pulse times is precision, meaning that such time can represent the pulse beginning or its peak instant (it can be considered that all pulses, in average, are delayed by a half-width amount).

3.2.3. Coincidence Counting Algorithm – CCA

Basically, CCA module determines the beta-gamma coincidence rate, searching for the minimum differences between the beta and gamma pulse pair time stamps remaining within the coincidence resolving time (t_C) which value is set in the input configuration file in microseconds. CCA flowchart is shown in Fig. 4 a and Fig. 4 b presents a timing diagram example illustrating the algorithm operation. Fig. 4 c shows a simplified flowchart of the first coincidence algorithm approach, called the two-nested-loop algorithm. Beta and gamma pulses are identified by index numbers, respectively ***i*** and ***j*** in Fig. 4. CCA analyzes all the coincidence combinations (beta-gamma pairs) in a single-iteration mode, using a stack structure and running approximately three times faster than the two-nested-loop algorithm. Each subsequent beta-gamma pair is stacked only if their time-difference is smaller than that of the previous pair, else, stacking is stopped and unstacking takes turn. The stack-top pair (minimum time value) is computed as a coincidence. If stack is not empty (after unstacking), the next stack-top is taken as another coincidence only if pulse-indices (both beta and gamma) differ from that of the last computed coincidence.

The two-nested-loop version, yet simpler, includes some improvements not shown in Fig. 4 c for simplicity. For instance, an index advance is performed, in order to remember the last computed coincidence, avoiding the internal loop to restart from its very beginning (first gamma pulse from the data block), at next iteration of external loop. Also, if a pulse from a given channel (as to say: gamma) has two or more near pulses from the other channel (beta) as possible coincidences, the minimum time-difference is chosen to form the real coincidence pair.

3.3. CAT functionality

CAT analysis-program integrates the above described functionality and objects, in order to perform the correct sequence of tasks needed for the calculation of the radioactive target-sample activity.

The program starts reading the Configuration-file, where measurement information and analysis settings can be retrieved from, namely: I/O data-files names; ROI-values lists (SAD controlling); dead-time values list (SDT) and coincidence-time resolution (CCA). List-format inputs enable the iterative operation, where the entire process is repeated for different variable-values (from the two lists: ROI-values; dead-time values). Then, families of results are obtained, such as the experimental extrapolation points.

Then, beta and gamma pulses information is read from SCS data-files

Table 2

Compilation of an extrapolation output file obtained with CAT analysis program, from a SCS measurement of ^{60}Co. The analysis was performed for eight different beta pulse discrimination levels and for two dead-time values, 3.0 and 4.0 μs. Iterations for each dead-time value, originally formatted vertically, are shown horizontally, side by side, for commodity. Absolute uncertainties are shown and specific-activities are in $Bq.g^{-1}$.

Extrapolation data and linear fitting – Analysis: NaI(Tl) gamma detector; sample ID: #15232 (Cobalt-60)									
Results - dead-time (s): 3e-06					Results - dead-time (s): 4e-06				
Ineff. Par.	Activity (Bq)	Uncertainty	Specific-Act.	Uncertainty	Ineff. Par.	Activity (Bq)	Uncertainty	Specific-Act.	Uncertainty
0.1375666	4946.742	8.974525	151880.3	314.6317	0.1366393	4944.659	8.950274	151816.4	313.9489
0.1610181	4949.098	9.616945	151952.7	332.0753	0.1600483	4946.765	9.593446	151881	331.4011
0.2243926	4948.585	11.1238	151936.9	373.8063	0.2234692	4946.285	11.10598	151866.3	373.2775
0.3071685	4951.135	12.77478	152015.2	420.6534	0.3063391	4949.06	12.76275	151951.5	420.286
0.4045314	4953.67	14.41319	152093	467.9368	0.403773	4951.803	14.40521	152035.7	467.6863
0.5139177	4957.584	15.99756	152213.2	514.2192	0.5133256	4956.165	15.99479	152169.6	514.1252
0.6321785	4959.824	17.48.942	152282	558.1547	0.6316502	4958.578	17.48892	152243.7	558.1296
0.7595148	4964.701	18.9246	152431.7	600.7058	0.7588211	4962.931	18.92097	152377.4	600.5842
Data fitting results -	y = c0 + c1 *x, where: x = Ineff. Par.; y = Activity.				Data fitting results -	y = c0 + c1 *x, where: x = Ineff. Par.; y = Activity.			
Intercept (c0 - Specific-Activity extrapolation value, for x = 0):					Intercept (c0 - Specific-Activity extrapolation value, for x = 0):				
1.5178e+05	±280				1.5170e+05	±278			
Slope (c1):	822.14	±779.89			Slope (c1):	865.35	±778.77		
Chi-squared:	0.028588				Chi-squared:	0.02667			
Covariance Matrix:	78117	−1.8541e+05			Covariance Matrix:	77488	−1.8421e+05		
	−1.8541e+05	6.0823e+05				−1.8421e+05	6.0649e+05		

and loaded into ‘**PulseRegister**’ vectors, aggregated to ‘**PulseData**’ objects. Two groups of data-files are considered, respectively, from the target-sample and background radiation (BKG) measurements.

Data processing, in brief, consists of the selection of the effective-pulses from the total records, starting with the ‘in-range’ pulses, according to the ROI list-setting (SAD), followed by the rejection of pulses within the virtual dead-time interval (dead-time list-setting – SDT). Then, the remaining pulses (effective or valid) are computed to determine the beta and gamma rates (sample, BKG, net) and obtaining the respective pulse-height distributions (histograms or spectra).

CCA software-module, then, seeks the time-coincident beta/gamma pulse-pairs from the effective pulses, according to coincidence-time resolution setting. A ‘**CoincidData**’ object encapsulates both coincidence configuration setting and processing results, including total, accidental and real coincidence rates, and the coincidence-time distribution (beta/gamma time-difference histogram).

Calculation, based on the effective-pulses rates, includes some corrections, according to the formalism of Cox and Isham (1977), considering: virtual measurement dead-times; BKG rates; accidental coincidences; radioactive decay. Current software version saves the following output text-files: report for each iteration (input and output files names; settings; pulse and coincidence rates; counting efficiencies; final activity-value); spectra (beta; gamma; coincidences); table of experimental points of the extrapolation curve (activity as function of beta-counting efficiency) and respective linear fitting data. The functionality and calculation processes described in this section are represented in Fig. 5, and Table 2 presents an extrapolation output file from a ^{60}Co sample measurement, showing the beta threshold iterations performed for two different dead-time iterations, 3 and 4 μs. Both fitting activity values are in very good agreement within the uncertainty ranges, meaning that dead-time corrections are satisfactory. CAT (blocks inside the dashed line in Fig. 5), resembles a typical conventional system, where:

- the detection signals (pulse vectors) are connected to amplitude discriminators (SAD);
- amplitude selected pulses are subjected to measurement dead-time (SDT);
- remaining processed pulses are counted and submitted to the coincidence module (CCA) for counting;
- beta, gamma and coincidence counts are then processed for rates calculation and radioactive sample activity determination (usually after measurement).

3.4. External applications

The present version of CAT provides the essential functionality and techniques for the Coincidence Methodology and, therefore, CAT output files have to be processed by external programs. For instance, any suitable spreadsheet or mathematical program can be used to carry out statistical analysis and data regression, and for elaboration of graphs and tables. Presently, first order linear fitting is achieved by CAT itself (as shown in Table 2) and higher order fittings or regressions including multiple parameters require external programs.

Monte Carlo simulation (MCs) is another powerful tool in LMN experimentation. Radionuclide standardization (including DCS measurements), for instance, makes use of MC to obtain theoretical predictions of the extrapolation curves, as explained below.

As mentioned in previous section, there are some factors that affect the measurements, demanding the proper corrections to be performed. Nevertheless, there are some factors related to decay phenomena and detection geometry limitations, hard to correct algebraically. For instance, because of the small dimensions of proportional counters (effective detection volume), high energy electrons usually lose small fractions of their energies. As a consequence, non-spectroscopic deposited-energy histograms are obtained: energy of primary and secondary electrons are mixed together in the spectrum. Therefore, it becomes a hard task to reduce the beta counting efficiency (extrapolation method, section 1.1) by cutting off beta pulses, according to amplitude selection criterion, because both categories of electrons are rejected. Ideally, only primary electrons (directly originated from decay) should be neglected, and the activity extrapolated value results inaccurate. In order to overcome the above explained disadvantage, LMN developed the ESQUEMA MCs program (Takeda, 2006; Dias et al., 2013). Simulation process includes the modeling of detection geometries and of radioactive samples, in order to obtain the deposited electron-energies spectra; this step is accomplished with MCNP6 (ORNL, 2006) program. Then,

Table 3
^{152}Eu decay branches employed for CAT analysis and branch pairing for bi-parametric fitting (Brancaccio, 2013) presented in the column at right. Asterisks at left indicate the four most intense branches, used for the Monte Carlo simulation, performed to obtain theoretical activity-values.

Decay branch		Energy		Probability	α_T	Combined data pair for bi-parametric analysis
used for analysis		(keV)		(%)		
*	β	($\gamma_{1,0}$)	344.28	26.59	0.0399(12)	[344 /964] [344 /1112] [344 /1408]
	β-	($\gamma_{3,1}$)	411.12	2.238	0.0239(7)	[411 /964] [411 /1112] [411 /1408]
*	β-	($\gamma_{7,1}$)	778.91	12.97	0.00190(6)	[779 /964] [779 /1112] [779 /1408]
*	EC	($\gamma_{9,1}$)	964.08	14.5	0.00270(8)	
	EC	($\gamma_{10,1}$)	1112.08	13.41	0.00200(6)	
*	EC	($\gamma_{13,1}$)	1408.01	20.85	0.000600(18)	

experimental parameters, analysis settings and the spectra obtained in first step are entered into ESQUEMA program, which generates the theoretical previsions for the extrapolation curves. Considering proportionality, the experimental curves (CAT) can be normalized by simulated data (MC), obtaining the best proportionality factor by means of least square method. Therefore, the desired activity-value can be obtained by multiplying the theoretical extrapolated-value (the number of histories of MCs) by the fitted proportionality factor (Takeda, 2006; Dias et al., 2013).

4. Methodology application and system validation

Basic functionality of DCS was implemented and initially tested by the standardization of a^{60}Co sample (Brancaccio et al., 2009), with satisfactory results. However, in order to start the DCS software expansion (section 3), the standardization of a^{152}Eu sample was chosen because of its challenging decay scheme and high potential for software implementation, testing and validation. Procedures and results details refer to (Brancaccio, 2013). Since the ^{152}Eu target sample was the same of a previous conventional standardization (Fonseca, 2002) the results from both works could be directly compared.

4.1. ^{152}Eu standardization

^{152}Eu presents a complex decay scheme, with three main branches (β^-, β^+, EC) and many gamma transition energies. ^{152}Eu is often employed in LMN calibrations, such as for attaining the 'gamma counting efficiency versus energy' calibration curve, for gamma detectors.

A previous LMN standardization of ^{152}Eu (Fonseca, 2002) was performed, employing a conventional system, as part of an international comparison organized by the BIPM (Bureau International des Poids et Mesures). Participant laboratories were supplied with an amount of the matrix solution, in the form of 26 µg de $EuCl_3$/ml HCl 1 M. LMN received 0,891321 g of the material and made fifteen samples for measurement. One of these samples, with a mass of $0.011534\,g \pm 0.2\%$, was set available for development of the DCS bi-parametric fitting methodology. Therefore, DCS results could be directly compared to that obtained previously, by internationally recognized laboratories.

For better understanding, the bi-parametric formulation (Fonseca, 2002; Brancaccio, 2013), for ^{152}Eu standardization, is briefly presented. Only the more intense decay branches, EC (≈73%) and β^- (≈27%), are considered, neglecting the β^+ (≈0.03%). Then, the beta count (N_β) can be derived from the activity (N_0) by the following expression:

$$N_\beta = N_0\,(a\,\varepsilon_{EC} + b\,\varepsilon_\beta) \quad (1)$$

where:

a, b respectively, the electron capture (EC) and β^- branches abundances;
ε_{EC}, ε_β respectively, the EC and β^- counting efficiencies.

Dividing expression (1) by the product of the efficiencies, $\varepsilon_{EC}\,\varepsilon_\beta$, follows:

$$\frac{N_\beta}{\varepsilon_\beta \varepsilon_{EC}} = N_0\left[1 + a\left(\frac{1-\varepsilon_{EC}}{\varepsilon_{EC}}\right) + b\left(\frac{1-\varepsilon_\beta}{\varepsilon_\beta}\right)\right] \quad (2)$$

Expression (2) shows the double dependency on the two efficiency inverses (or inefficiencies), corresponding to the decay branches (EC and β^-). In such case the beta efficiency extrapolation method (bi-parametric) may take both ε_{EC} and ε_β efficiencies into account in order to make both of them to tend to unity ($\varepsilon_{EC} \approx 1$and $\varepsilon_\beta \approx 1$) and, then, making the beta count, N_β, to tend to the desired activity, N_0. With some algebraic manipulation, expression (2) can result more understandable and useful:

$$y = p_1 x_1 + p_2 x_2 \quad (3)$$

where:

$\boldsymbol{y}$ equal to: $\frac{N_\beta}{\varepsilon_\beta \varepsilon_{EC}}$;
$\boldsymbol{p_1}$ equal to: N_0 (radioactive sample activity);
$\boldsymbol{p_2}$ equal to: $\boldsymbol{a}\,N_0 = \boldsymbol{a}\,\boldsymbol{p_1}$;
$\boldsymbol{a}$ intensity (or abundance) of the electronic capture decay branch;
$\boldsymbol{x_1}$ equal to: $1 + \left(\frac{1-\varepsilon_{EC}}{\varepsilon_{EC}}\right)$;
$\boldsymbol{x_2}$ equal to: $\left(\frac{1-\varepsilon_\beta}{\varepsilon_\beta}\right) - \left(\frac{1-\varepsilon_{EC}}{\varepsilon_{EC}}\right)$.

The sample measurement was accomplished in a 20000 s acquisition (SCS) for good counting statistics. Background (BKG) counting was evaluated from a 5000 s acquisition (SCS).

The signals from two gamma detectors – NaI(Tl) and HPGe – and from a 4πβ proportional counter (Detection Assembly #1; see Fig. 1) were simultaneously acquired. As gamma data, only the pulses from HPGe were employed for the analyses processes, because of its better energy resolution, needed for the proper gamma peak discrimination.

CAT main parameters setting was:

- Coincidence resolving time (CCA): ±1 µs;
- Virtual measurement dead-time (SDT): 3 µs;
- Activity reference date (local ISO 8601 format): 1999-07-01T09:00:00.

Six 4πβ-γ coincidence analyses were performed by CAT program, centered on gamma transitions originated from β^- branches (344.28, 411.12 and 778.91 keV) and electronic capture branches – EC – (964.08,

Table 4
Activities obtained by Monte Carlo for ^{152}Eu and fitting parameters, related to one out of nine experimental data combinations (Nine-pair). Parameters p_1 corresponds to the obtained activity-values, from each fitting process. Four data-pairs were allowed for Monte Carlo simulations (Four-pair).

Analysis gamma energy pair [β^- /capture] (keV)	Theoretical Activity (Monte Carlo) $\times 10^2$ kBq g^{-1}	Experimental activity (Fitting Parameter p_1) $\times 10^2$ kBq g^{-1}	Fitting Parameter p_2 $\times 10^5$ kBq g^{-1}
[344 /964]	5.81(7)	5.908(4)	4.57(3)
[344 /1112]	–	5.971(8)	5.08(6)
[344 /1408]	5.79(4)	5.801(4)	4.32(3)
[411 /964]	–	5.898(5)	3.99(4)
[411 /1112]	–	5.957(8)	4.58(8)
[411 /1408]	–	5.805(5)	3.73(4)
[779 /964]	5.70(9)	5.832(6)	4.37(4)
[779 /1112]	–	5.917(9)	4.92(7)
[779 /1408]	5.62(9)	5.722(5)	4.12(3)

Table 5
Uncertainty budget and calculation, applied to the final activity-value of ^{152}Eu.

Uncertainty budget	Value (for u = 1) (%)
Decay Correction	0.15
Mass of ^{152}Eu Sample	0.20
Efficiency parameter	0.11
Beta counting statistics	0.01
Experimental fitting procedure	0.69
Monte Carlo fitting procedure	0.73
Total – Experimental	0.74
Total – Monte Carlo	0.78

1112.08 e 1408.01 keV).

Reference date was set to be the same as previous BIPM measurements in order to allow direct comparison of results. Such six analyses were combined into nine pairs (gamma from β^-/gamma from EC), as shown in Table 3.

The bi-parametric fitting process was applied for each analysis-pair. Table 4 presents the results for each pair of energies (β^-/EC), resulting in nine sample activity values (called the nine-pair results). The four energy pairs used in MCs (called the four-pair) were also used for the four-pair experimental calculations for direct comparison of results (MCs/experimental). The final value was obtained from average calculation, weighted by variance inverses. Experimental and MCs results are presented in section 5.

5. Results and discussion

The present section presents the DCS' main results of the ^{152}Eu standardization, as explained in section 4.

5.1. ^{152}Eu standardization

DCS ^{152}Eu measurements (Brancaccio, 2013) were carried out with an available sample from a previous standardization (Fonseca, 2002), performed with conventional techniques. Results obtained from bi-parametric fitting are given in Table 4. The nine pair combinations (β^-/capture branches) shown in Table 3 are repeated at first column in order to present individual fitting parameters. Activity-values from MCs, presented in the second column, were obtained for the most intense gamma transitions (344 and 779 keV) and β^- branches (964 e 1408 keV, from capture). The last two columns at right in Table 4 show the parameters, p_1 and p_2, obtained from fitting. The target sample-activity is given directly by the fitting parameter p_1 (column 3) from expression (3), section 4.1.

Uncertainties in parameters p_1 and p_2 (Table 4) were obtained directly from bi-parametric fitting, taking into account the contributions shown in Table 5 for uncertainty propagation. Table 6 shows the final activity values compared to previous results (Fonseca, 2002) for the same ^{152}Eu matrix solution, with very good agreement, within uncertainties.

6. Conclusion

DCS, made up of SCS (data-acquisition unit) and CAT (analysis software package which is the main focus of this work), was presented in this work. The main software elements are described and the experimental results discussed.

System validation was suitably accomplished by the standardization of ^{152}Eu radionuclide, which measurements provided the material for software implementation and testing. OOP was employed to create an internal data representation (objects). Algorithms and software-modules (SAD; SDT; CCA) were checked out for a wide range of parameter settings, with good results.

Amplitude discrimination, SAD, proved to be a useful tool for gamma-pulse windowing and for the attaining of beta efficiency extrapolation curves, from a single data set, by varying beta-pulse thresholds.

The virtual dead-time generator, SDT, set, typically, to 2.0 or 3.0 μs showed to be appropriate to discard multiple registering of single saturation pulses. Higher values settings, up to 1 ms, were also tested and the proper compensation was performed by the correction algorithm.

The coincidence time resolution, usually set to a ±1 μs range for the most of cases, was successfully tested with CCA settings ranging from ±1 to ±200 μs.

The results of ^{152}Eu target sample measurements could be compared to those from its previous standardization, showing good agreement within uncertainty ranges.

DCS' standardization process showed to be easier and faster than that performed by conventional measurement systems; just a few previous settings are required, since most adjusts are carried out by software. Obtained precision and accuracy levels are compatible or even

Table 6
Obtained activity-values (kBq g^{-1}) of ^{152}Eu. The Nine-pair value, obtained from all nine energy pairs (Table 4), is compared to the results from previous work (Fonseca, 2002). The Four-pair calculation uses only the four energy pairs also used for the Monte Carlo simulation, allowing direct comparison between experimental and theoretical values. Percent differences in the bottom two lines are the rounded up values of 100 × |PWv – Lv| /PWv, where PWv represents the present work values and Lv correspond to the values found in the literature.

	LMN (DCS) (Four-pair) (Nine-pair)	Monte Carlo Simulation (Four-pair) –	LMN (Conventional) Fonseca (2002)	BIPM average of four main laboratories (Fonseca, 2002)	BIPM main average Fonseca (2002)
(Four-pair)	578.1 ± 4.2	577.1 ± 3.6	575.7 ± 7.8	578.1 ± 9.8	582.0 ± 8.1
(Nine-pair)	583.6 ± 2.7	–	–	–	–
Difference (%)					
(Four-pair)	–	0.2	0.4	0.0	−0.7
(Nine-pair)	–	1.1	1.4	1.0	0.3

better.

CAT is a versatile software package and can be used with any coincidence system or easily adapted for such purpose. Non-nuclear-event counting can also be considered, involving or not coincidence criteria.

Even though DCS is considered a pilot project, the presented considerations show that the software methodology, developed for coincidence counting analysis, is trustworthy, representing an important step to improve the LMN measurement capabilities.

New improved DCS versions are planned, with the following features:

- use of more powerful hardware, with sampling rates up to 1 GS/s;
- increasing of software autonomy, flexibility and efficiency, including:
  - extensions of the Coincidence Methodology concepts and procedures;
  - friendly graphic user interface (GUI);
  - new software-modules, (extending dead-time mode, for instance);
  - new math capabilities for data fitting, graphs, etc.;
  - web connection and cloud computing, to provide remote access to databases (DB) and software functionalities, as follows:
  - DB: nuclear data; radioactive samples catalogs; measurement and analysis log files for configurations and results;
  - software: modules; Application Programming Interfaces (API) and programs.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apradiso.2019.108900.

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