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Autonomous sample switcher for Mössbauer spectroscopy

J. H. López[1] · J. Restrepo[1] · C. A. Barrero[2] · J. E. Tobón[3] · L. F. Ramírez[3] · J. Jaramillo[3]

Abstract In this work we show the design and implementation of an autonomous sample switcher device to be used as a part of the experimental set up in transmission Mössbauer spectroscopy, which can be extended to other spectroscopic techniques employing radioactive sources. The changer is intended to minimize radiation exposure times to the users or technical staff and to optimize the use of radioactive sources without compromising the

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Edited by Juan A. Jaén

✉ J. Restrepo
jrestre@gmail.com

J. H. López
jolobotero@gmail.com

C. A. Barrero
cesar.barrero.meneses@gmail.com

J. E. Tobón
nobotj@gmail.com

L. F. Ramírez
luisf.ramirez@udea.edu.co

J. Jaramillo
jdex87@gmail.com

[1] Group of Magnetism and Simulation, Institute of Physics, University of Antioquia, A.A. 1226, Medellín, Colombia

[2] Group of Solid State Physics, Institute of Physics, University of Antioquia, A.A. 1226, Medellín, Colombia

[3] Group of Scientific Instrumentation and Microelectronics, Institute of Physics, University of Antioquia, A.A. 1226, Medellín, Colombia

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resolution of measurements or spectra. This proposal is motivated firstly by the potential hazards arising from the use of radioactive sources and secondly by the expensive costs involved, and in other cases the short life times, where a suitable and optimum use of the sources is crucial. The switcher system includes a PIC microcontroller for simple tasks involving sample displacement and positioning, in addition to a virtual instrument developed by using LabView. The shuffle of the samples proceeds in a sequential way based on the number of counts and the signal to noise ratio as selection criteria whereas the virtual instrument allows performing a remote monitoring from a PC via Internet about the status of the spectra and to take control decisions. As an example, we show a case study involving a series of akaganeite samples. An efficiency and economical analysis is finally presented and discussed.

Keywords Mössbauer spectroscopy · Optimization · Virtual instrument · Data control · Radiation safety

1 Introduction

The International Commission on Radiological Protection (ICRP) has determined some protocols and regulations regarding the use of radioactive sources in order to minimize exposure times to the people involved in the management of that sources in such a way that radiation doses remain below certain standards already established [1, 2].

In practice however, it is easy to find misconducts that put the safety of personnel at risk. An example takes place in laboratories employing spectroscopic techniques based on radioactive sources as is the case of Mössbauer spectroscopy where radioactive sources are used [3–5]. Tasks in these laboratories involve research, teaching, services, training, technical features like source or sample replacement, etc. and all of them imply certain degree of radiation exposure. Moreover, when these spectroscopic techniques are used as the experimental counterparts in teaching experimental courses (e.g. a solid state physics course, etc.) students become part of itinerant groups of individuals non-occupationally exposed. In some other cases, laboratories managers are not aware of informing about the existing regulations or even simply they do not know the normative. Otherwise, suitable knowledge and information can even exist but misconducts may persist. Examples of these misconducts can be observed for instance when spectra are recorded pretty quickly with no good enough resolution or statistics. This usually happens when fast results are needed, many users are involved, there is a long queue of samples to be measured in a short period of time, or as an attempt to optimize airtime. All these aspects represent potential hazards in terms of radiation exposure time that must be cushioned [6–8].

Another relevant fact deals with life times which become critical as they are of the order of days or even less. Here, an optimal up-time is required and the use of radiation must be as effective as possible. Therefore, it is desirable to control the time for every single sample in order to spend only the necessary and avoiding greater times that do not provide additional information. This situation is pretty common in cases where the collection of a spectrum takes place during nights, weekends or holidays resulting in time-wasting. This fact is critical when using new sources having more radiation fluency at the first stages of their life times and where some few hours usually are enough for recording a good spectrum.

For the above reasons, an automatic sample changing system becomes useful. The objectives of such a device are: a) to minimize exposure times of exposed individuals, b) it is not

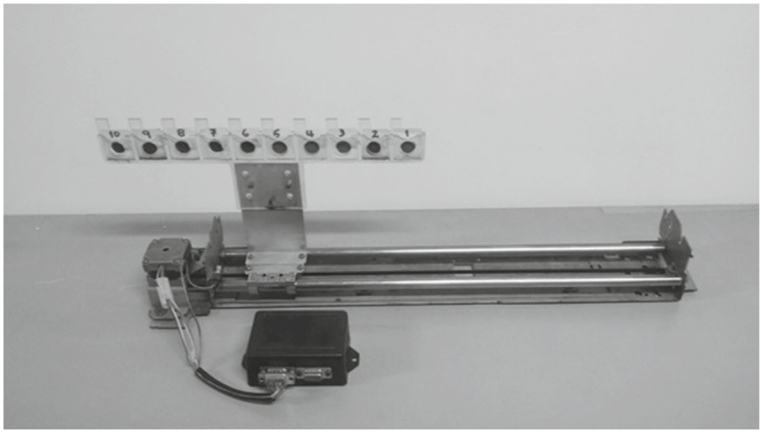

Fig. 1 Sample holder switcher for Mössbauer spectroscopy and circuit

necessary to be physically close to the equipment to change a sample or to follow the evolution of a spectrum. This can be done via remote control through a virtual instrument and the web, c) to optimize the radiation use times according to the signal to noise ratio and d) to allow a sample change (manual or automatic) to begin with another new spectrum with the respective data storage for analysis.

2 Sample switcher

In order to comply with the above goals we have designed an autonomous sequential sample switcher to be used in spectrometer devices (e.g. Mössbauer spectroscopy). It consists of a sample holder assisted by an electromechanical part responsible for the holder movement, sample alignment (hardware) and a virtual instrument (software) operated via PC for data acquisition and analysis of the sample change criteria.

2.1 Hardware

The switcher consists on a kind of rule, transparent to $\gamma-$rays (when using ^{57}Co sources), and formed by compartments or pockets storing each of the samples as are shown in Fig. 1 in addition to a circuit. The rule can be in principle of a linear type with a number of pockets defined in terms of the needs. Other different geometries could also in principle be implemented (e.g. an endless circular geometry with the source at the center). The size of the pockets can be also redesigned as a function of the particular type of spectroscopy or geometry. The electronic circuit is composed of a PIC microcontroller 12F1822 (http://www.microchip.com/wwwproducts/en/PIC12F1822) and a power system. Its function is to control a step motor to move the holder and the respective source-absorbent alignment. Communication between the microcontroller and PC is carried out by means of the serial RS232 protocol. The system has a switch that is pressed each time the first sample (sample number one) is in front of the radioactive source. This switch also serves as a sensor to restart the sample holder and to take control of the sample position according to the physical dimensions of the pockets.

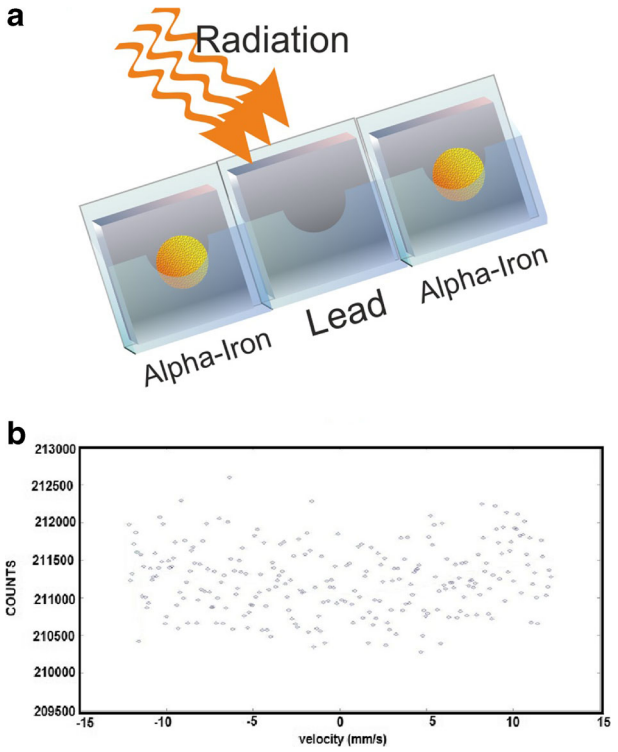

Fig. 2 Experimental set up of a lead sample sandwiched between two enriched $\alpha-$Fe calibration samples. (**b**) Spectrum obtained from (**a**)

The size of the pockets fulfills the standards of Mössbauer absorbents and the usual geometrical criteria described elsewhere [9]. Likewise, the separation between the pockets, of the same order of the pocket length, was designed in such a way to ensure that signal arising from a particular sample was not influenced by absorbent neighbors [9]. This fact is endorsed by Fig. 2 where the recorded signal is in the range of noise, i.e. no absorption lines were evidenced for a sample of lead with two α-iron samples as nearest neighbors. This means that for a given sample being measured, its neighbors do not interfere in the measurement.

On the other hand, removing or replacing a sample in order to introduce a new one can also be carried out at any time without disassembling the exchanger equipment. However, even though the system as a whole is mounted upon an antivibratory set up, undesired external vibrations can be introduced while changing a sample by hand and, at the same time, another sample is being measured. To avoid that, it is recommended to pause data collection during the exchange of a sample by hand.

2.2 Software—virtual instrument

For developing the virtual instrument (V.I), LabVIEW (http://www.ni.com/labview/esa/) was chosen as the graphical programming software. The main function of V.I. is to control the switcher by establishing the communication with the spectrometer, data storing,

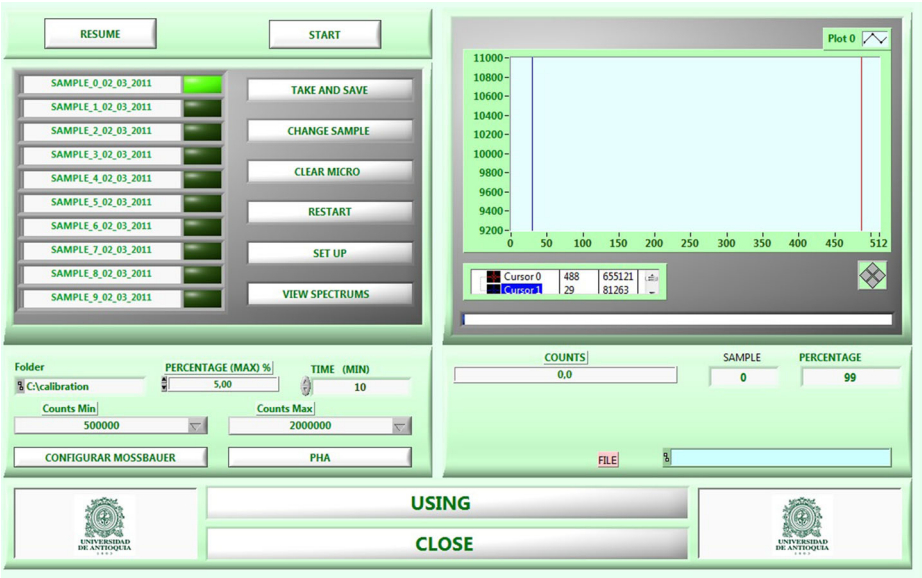

Fig. 3 Front-page of the virtual instrument

data visualization and data analysis. The latter implies a data comparison with the so-called change criteria previously programmed. When criteria are fulfilled, the V.I. triggers the signal for sample switch. Data backup is also performed for further analysis or either for data recovery in case of any electrical failure or continuation of any measurement. After recording the spectrum of the last sample in the rule, the switcher stops and it gets ready for rebooting. Another great advantage is data visualization and switcher control through V.I. from any PC with Internet access in the same way as in the lab. Thus, the researcher can monitor the Mössbauer spectrum being recorded at any time. Manual decisions about stopping a spectrum, rebooting or retaking an old one instead of autonomous decisions are also available at any time. Figure 3 shows a snapshot of the virtual instrument and Fig. 4 is the flowchart showing the logic implemented. The virtual Instrument generates a panel to visualize the spectrum of the current sample, the number of counts in the baseline and spectral information stored in a file for later analysis. The switcher control generates a list with the particular filename for each sample (default sample_X). The number of files to be displayed in the screen is ten by default, but the list goes from sample_0 to sample_99 to avoid overwriting files. Every time a sample is read, a light indicator is turn on in front of the name of the sample. This indicator shows what sample is being currently analyzed.

2.3 Meeting the criteria

As we have already mentioned, at any time the user can take any decision about a spectrum being recorded including a new measurement. However, in the absence of the user during certain periods of time, the system attends the following default criteria before changing a sample and getting a new record. These criteria, which also can be changed by the user, are [10, 11]:

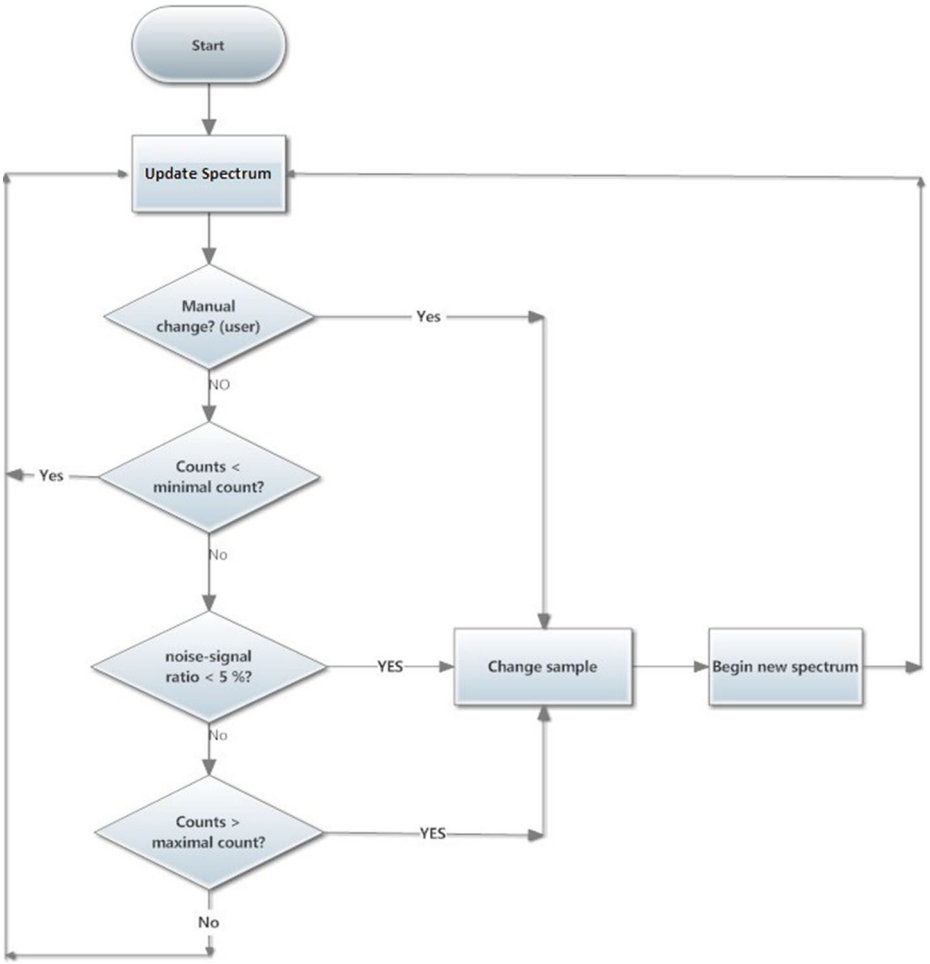

Fig. 4 Flowchart of the logic implemented in the virtual instrument

Number of counts First, it is important to stress that not necessarily a great amount of counts implies a good spectrum. Moreover, a good spectrum can be also achieved with a relatively small or moderate amount of counts in some samples as long as the signal-to-noise ratio is high enough, like it is the case in samples containing a high density of the target element compatible with the source isotope as concerns for instance to the nuclear resonance in Fe-rich samples studied by ^{57}Fe Mössbauer spectroscopy. For this reason, this criterion must be considered concomitantly with the signal-to-noise ratio criterion. In the virtual instrument we have fixed a minimum amount of 2×10^6 counts per channel on the baseline, at least for Mössbauer spectroscopy according to our experience with canonical samples. Nevertheless, such an amount can be modified in order to fulfill features of another particular spectroscopic technique where the exhanger sample device can be mounted or depending on the characteristics of the sample [9].

Signal-to-noise ratio As the Virtual Instrument collects the spectrum, a data analysis is performed by estimating the signal-to-noise ratio. To make it, the average is taken from the

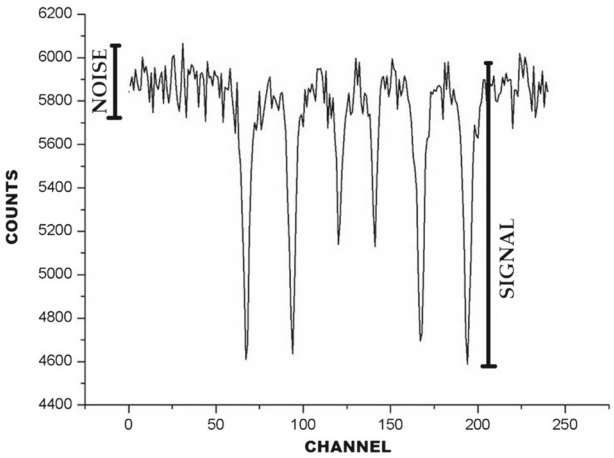

Fig. 5 Analysis of the signal-to-noise ratio of a Mössbauer spectrum

first channels on the baseline in absence of any resonance signal and then it is compared with the deepest resonance absorption line as is shown in Fig. 5 for a typical Mössbauer spectrum. When this ratio reaches the 5%, it is considered that the spectrum has enough information and the switcher makes the change to the next sample [10, 11]. The criterion is considered fulfilled even if the minimum number of counts is not reached. Once more, the percentage of signal-to-noise ratio can be adjusted according to requirements established by the user.

3 Mössbauer spectroscopy results

3.1 Akaganeites as a case study

The V.I., besides controlling the switcher, it also controls the spectrometer from a panel in the PC. Thus, the system stands for a "virtual instrument" as we have called it. The items to be determined from the panel are: a) the number of channels (options are 256, 512 and 1024 by default, however in cases where higher spectral resolution is needed, the virtual instrument can be configured to address more channels, i.e. 2048 or 4096 channels) and b) frequency, speed and type of ramp of the driver containing the radioactive source.

Room temperature akaganeite ^{57}Fe Mössbauer spectra recorded by using the automatic switcher and the above criteria are shown in Fig. 6. Although the spectra appear already fitted (an additional task), they are not normalized in order to make explicit the criteria above mentioned regarding the minimum number of counts on the baseline and the signal-to-noise ratio. A high resolution is observed resulting in a suitable fitting process.

Finally, in order to evaluate the economical impact of our switcher, we address the problem of estimating the cost of a ^{57}Fe Mössbauer spectrum as an indirect measurement of the cost derived from a non-optimum use of the radioactive sources used in this spectroscopy.

3.2 How much does a Mössbauer spectrum cost?

To estimate the cost of a single Mössbauer spectrum the following issues must be addressed:

– Cost of a single 14.41 keV photon.

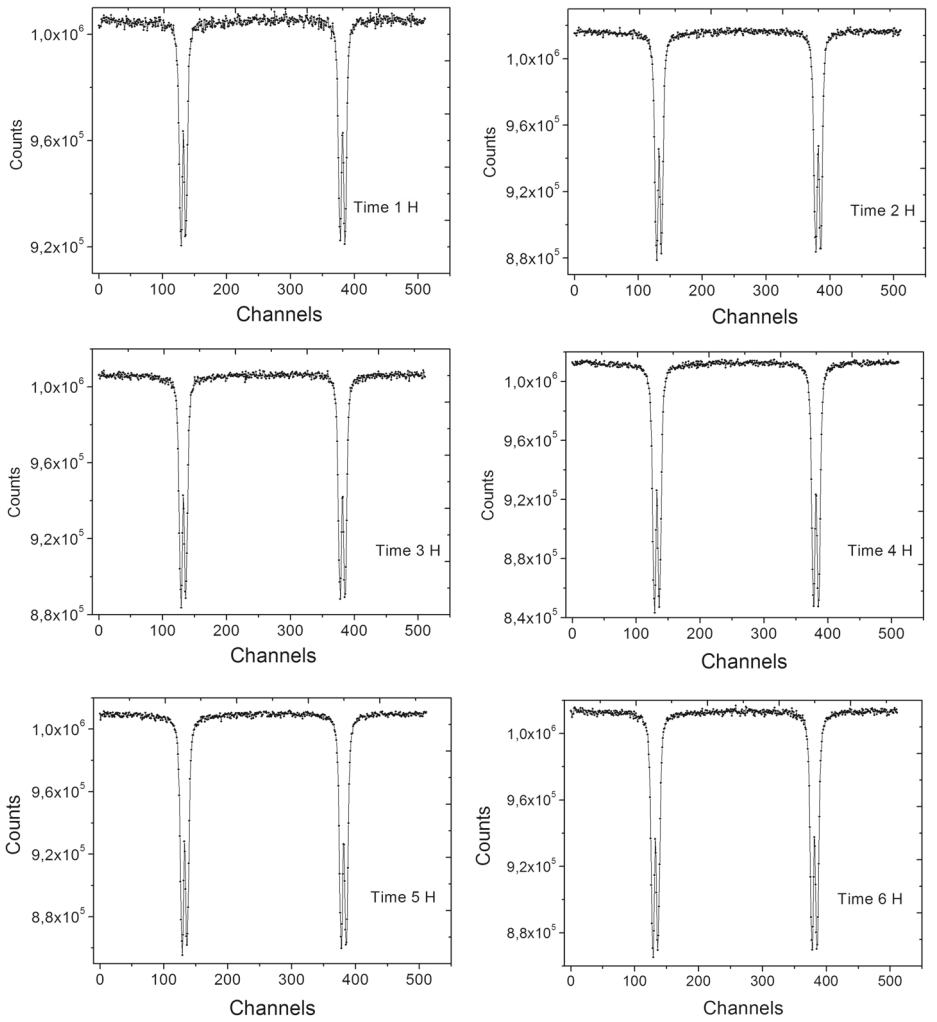

Fig. 6 Set of room temperature ^{57}Fe Mössbauer spectra for akaganeites mechanically milled at different milling times. Spectra were recorded by using the automatic sample switcher and by applying the criteria

– Cost of getting a *count* in a single channel and the corresponding record over certain number of channels (e.g. 1024) of the Multichannel Scaling system.
– Finally, we can estimate a cost on the basis of an average of 2×10^6 counts per channel.

In ^{57}Fe Mössbauer spectroscopy is usual, after getting a new radioactive source of certain initial activity (e.g. 50 mCi, which is typical for ^{57}Fe Mössbauer spectroscopy), to have a useful time of some few lifetimes, i.e. some few years. More concretely, for our calculation we consider a period of 5 years, i.e. 6.7 lifetimes. For ^{57}Co, the lifetime is $\tau = 271.8$ days or 23'483.520 s, and its cost on average, including importation and reexportation, can be as high as 1.5×10^4 US dollars as it occurs in countries of South America. However, it must be stressed on this issue, that costs, taxes and fees can vary depending on the country. On

the basis of the above estimation and by assuming an initial activity $R_0 = 50$ mCi, which means 1.85×10^9 decays per second with 1 Ci $= 3.7 \times 10^{10}$ decays per second, the amount of nuclei to decay in a useful period of around 5 years, i.e. $T \approx 6.7\tau$, is given by [12]:

$$N(T) = \int_0^T R(t)dt = \frac{R_0}{\lambda}(1 - e^{-\lambda T}) = 6.2 \times 10^{16} \quad (1)$$

where the activity is $R(t) = R_0 e^{-\lambda t}$ with $\lambda = \ln 2/\tau = 2.95 \times 10^{-8} s^{-1}$. From this amount of nuclei decaying, around 91% of them correspond to $\gamma-$ rays of 14.41 keV according to the decay scheme of ^{57}Co, i.e. approximately 5.6×10^{16} 14.41 keV photons. Therefore, during the useful time, the cost of a single 14.41 keV photon is around 2.7×10^{-13} US dollars. However, only a fraction of them generated at the source will arrive to the absorbent, and from them, only a fraction will produce Mössbauer effect as long as zero phonon processes are governed by certain probability. Such a probability is determined by the Lamb-Mössbauer factor f, which is a function of temperature and it represents the fraction of the recoil free transitions relative to the total number of transitions.

To finish the calculation we must therefore to estimate the number of 14.41 keV photons that will arrive on average to the detector during the useful time we have considered. By assuming an isotropic distribution of photons emerging from the source in all directions and that the effective circular area of absorption is of the type $\pi(\Delta r)^2$, it is easy to see that such an amount is given by:

$$\Delta N = \frac{N_0}{4}\left(\frac{\Delta r}{r}\right)^2 \quad (2)$$

where r is the source-absorbent distance and Δr is the radius of the effective circular area for absorption which is typically of the order of 1 cm, and N_0 is the total amount of photons emerging from the source. Thus, by assuming a typical distance of $r = 20$ cm we get $\Delta N = 3.1 \times 10^{13}$ 14.41 keV photons reaching the sample. This means that approximately 0.0625% of the total amount of photons reach the absorbent. This factor can be also interpreted as an shortage index in the *photons market*, which means that in principle the cost of a single photon should increase by the inverse of such index ($\approx$1600 times), i.e. around 4.3×10^{-10} US dollars per photon. Additionally, a typical room temperature f value for ^{57}Co sources, after correction for thickness effects, is of the order of 0.7 [13]. Once more, this means that the cost should increase about $\approx$1.4 more times, i.e. 6.1×10^{-10} US dollars per photon. Now, on the basis of 2×10^6 counts per channel and 1024 channels, we arrive to a final naive estimated cost of around 1.3 US dollars per spectrum. Of course, a realistic estimation should include other costs associated to: refrigeration (in case of low-temperature measurements), electric power consumption, staff, administrative affairs, licenses, dosimetry, locatives, etc. It is interesting to remark finally that from around 1300 nucleides, that are known to exist, almost one hundred can be used as Mössbauer isotopes, which belong to some 45 elements. From these, about 2/3 of all Mössbauer studies concern iron, about 1/6 concern zinc and the rest are dealing with other elements.

4 Conclusions

From the above naive economical analysis, if we take into account that usually a Mössbauer spectrum is recorded in some few hours, it is easy to realize the economical impact involved in the number of hours, days or weeks wasted as a consequence of a non-optimum use of radioactive sources employed in spectroscopic techniques like the Mössbauer one analyzed

in this work. From this standpoint, and as a manner of taking advantage of the resources, we have designed and proposed a sample switcher device that we hope can be implemented by the scientific community dealing with spectroscopies based on radioactive sources. The switcher also satisfies our goal of minimizing the radiation exposure times by the users directly involved in the measurements.

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