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CONSIDERATIONS ABOUT THE NEURAL NETWORK APPROACH FOR ATOMIC AND NUCLEAR SPECTRAL ANALYSIS

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Although Neural Networks have proliferated for spectroscopic data interpretation, this paper shows, with simple ideas and well-known examples that they are not well suited for this kind of analysis. This conclusion can be understood after we demonstrate that spectroscopic results are absolutely equivalent, from a statistical point of view, to a sequence of statistically independent events. Stochastic independent events, like a dice throw or a coin tossing, cannot be better described than by a purely statistical method. This conclusion remains unchanged independently if the results are linear or not with respect to some parameters of the studied system, as is depicted in this work. If a method performs better than statistical methods for spectral analysis, it could be understood that it is violating the causality principle. As a representative example, it is described the particle transport equation (Boltzmann's Equation) solved by Monte Carlo methods, with which we can simulate a general spectroscopic technique. Also are described the Total Reflection X-ray Fluorescence and the Neutron Activation Analysis Techniques. In none of these systems it would be expected that Neural Networks perform better than statistical methods for spectral analysis.

Keywords: Atomic spectral analysis; spectroscopy; neutron activation analysis.

1. Introduction

In the last years Neural Networks (NN) [Fausett (1994)] have been generalized, presenting wide scientific use, particularly in physics and chemistry, in the interpretation of results obtained by spectroscopic techniques. Separately and independently, these results were historically interpreted with "classical methodologies," derived from statistical formulations.

The purpose of this paper is to analyze why NN can not perform better than Statistical Methods when processing a spectrum obtained by a molecular, atomic or nuclear spectroscopic technique. To this end, we divide this article into five numbered sections, each one of them describing a particular independent and complementary issue.

Section 2 describes the generalities of spectroscopic techniques. Section 3 describes a sequence of simple, pure statistically independent experiments in order to confirm that they are absolutely equivalent, from a statistical point of view, to

the spectra produced by many linear spectroscopic techniques. After that demonstration, we present our first conclusion, which is: Stochastic independent events can not be better described than by a purely statistical method.

Section 4 presents the particle transport equation which describes the behavior and distribution of subatomic particles (neutron, photon, electron, etc.). Consequently, its predicted results, under specific conditions, should represent the results obtained by spectroscopic techniques. This comparison is usually faced by nuclear reactor designers, who need to validate their calculation tools against a wide set of representative experiments (benchmarks). We describe the transport equation applied to the case of neutron transport because there are obvious (familiar) examples, from nuclear reactor experience, which corroborate the deduced conclusions.

The transport equation can be solved by two totally different methodologies: (A) Deterministic, and (B) Statistical methods (Monte Carlo methods). Deterministic methods solve the transport equation for the average particle behavior; its numeric resolution obtains particle distribution in the whole phase-space. Alternatively, Monte Carlo methods do not solve any explicit equation but obtain answers simulating many individual particle stories and they collect information about their behavior. The possibilities that rule interactions and reactions of each particle, in the physical system, are statistically sampled (from its birth to its death), so the set of "events" which accumulate particles in the system describe the real physical phenomenon. Monte Carlo methods are well suited to solving complicated problems, obtaining doses, criticalities, etc. in atomic or nuclear systems — independently of whether those systems are linear or not. These results indicate that purely statistical methods are well suited for describing any sequence of stochastic independent events.

In Sec. 5, we apply deductions obtained throughout the paper to two representative spectroscopic techniques (Total Reflection X-Ray Fluorescence and Neutron Activation Analysis).

The paper finishes by making a general balance of the conclusions obtained from each section.

2. Spectroscopy

Originally spectroscopy was the study of the interaction between radiation (the excitation source) and matter, registered as a function of wavelength (the response because the effect of the source). Historically, spectroscopy referred to the use of visible light dispersed according to its wavelength — e.g. by a prism. Later, the concept was expanded to indicate any measurement of a quantity as function of either wavelength or frequency. A further extension of the scope of the definition added energy (E) and time (t) as variables. A plot of the response as a function of wavelength (λ), frequency (ν), energy (E), time (t), etc. is referred to as a spectrum.

Spectrometry is the spectroscopic technique used to assess the concentration or amount of a given species. In these cases, the instrument that performs such

measurements is a spectrometer or spectrograph. Spectroscopy is often used in physical and chemical sciences to identify substances, chemical structures, crystal parameters, etc. through the spectrum emitted from, absorbed or scattered, etc. by them.

The type of spectroscopy depends on the physical quantity measured as an effect of the excitation source. Normally, the quantity measured is an intensity, either of energy produced or absorbed.

- Electromagnetic spectroscopy involves interactions of matter with electromagnetic radiation, such as light.
- Electron spectroscopy involves interactions with electron beams. Auger spectroscopy involves inducing the Auger effect with an electron beam, where the measurement typically involves the kinetic energy of the electron as variable.
- Mass spectrometry involves the interaction of charged species with magnetic and/or electric fields, giving rise to a mass spectrum, which has the mass m as variable, but the measurement is essentially one of the kinetic energy of the particle.

Some other techniques could be described [De Soete (1972); Johansson (1995); Teo (1986); Siegbahn (1965); Carlson (1978)].

Some spectroscopic methods are differentiated as either atomic or molecular based depending whether or not they apply to atoms or molecules. Along with that distinction, these espectroscopies can be classified according to the nature of the interaction. A quite short description follows:

- Absorption spectroscopy uses the range of the electromagnetic spectra in which a substance absorbs. It includes atomic absorption spectroscopy and various molecular techniques, such as infrared spectroscopy in that region and nuclear magnetic resonance (NMR) spectroscopy in the radio region.
- Emission spectroscopy uses the range of electromagnetic spectra in which a substance radiates. The substance first must absorb energy, which can be from a variety of sources, which determines the name of the subsequent emission, like luminescence. Molecular luminescence techniques include spectrofluometry.
- Scattering spectroscopy measures the amount of light that a substance scatters at certain wavelengths, incident angles, and polarization angles. The scattering process is much faster than the absorption/emission process. One of the most useful applications of light scattering spectroscopy is Raman spectroscopy.

Some spectroscopic techniques present linear properties, that is, their response when doubling the source intensity or the amount of sample analyzed, is a double intensity of the acquired signal. A detailed description of this kind of response is given in Secs. 4 and 5.

3. Analogy Between Linear Spectroscopy Results and Statistical Sequences

In the following section, we describe a sequence of simple pure statistically independent experiments in order to attest that they are absolutely equivalent, from a statistical point of view, to the analyzed linear-spectra produced by many techniques [Bennun (2009)].

Let us start by considering a coin tossing. A coin, when thrown into the air and allowed to fall, will rest with either side (head or tail) face up with equal probability. If we train correctly a NN in order to predict the outcome of the next toss, it will give a right answer half of the times, and a wrong answer the other half, on the average. No NN could show a better performance than statistical methods because the system under study is composed of independent stochastic events. The best performance of any NN (applied to any linear-statistical system) would be as good as the statistical method performance.

Let us now assume that we throw a dice many times. The chance of each face is $1/6$. A given sequence of data for each face, being divided by the total number of throws, is our first spectrum a_1, which only takes discrete values. In the limit of an infinite number of events, a_1 tends to the probability ($p_i = 1/6$, $i = 1, \ldots, 6$). If we throw a 5-faces device, the chance obtained for this second spectrum b_1, for each face is $1/5$ ($p_i = 1/5$, $i = 1, \ldots, 5$). If we put both devices into a recipient that is shaken, one is extracted, and the bottom face is studied, we will obtain a spectrum c_1, which is a linear combination of spectra a_1 and b_1. Spectrum c_1 will be more similar to a_1 if the frequency α_1 of extracting the six-face dice is bigger than the frequency β_1 of extracting the five-face dice, and vice versa. Using the χ^2 distribution statistical method it is possible to determine the more probable values of α_1 and β_1, and their corresponding standard deviations ($\Delta\alpha_1$ and $\Delta\beta_1$) for a given sequence of data c_1.

Many linear spectroscopic techniques produce spectra totally analogous to the example introduced above, where the only difference is the spectra a_1 and b_1 are discrete. This can be explained as follows: Let us start by considering an electromagnetic system where we follow many photons incident on a sample. The spectrum produced by the sample a_2 is the result of the stochastic interactions that each one of those incident photons undergoes in the sample. Some of their possible probabilistic interactions were described by Einstein [Einstein (1905)] and Compton [Compton (1923)]. If we analyze another sample, a new spectrum b_2 is obtained. If we analyze both samples simultaneously, the obtained spectrum c_2, will be a linear combination of a_2 and b_2. Thus, as in the previous example, the measured spectrum can be written as: $c_2 = \alpha_2 a_2 + \beta_2 b_2$.

In our example, the exciting source is the shake; in the linear spectroscopy studied, exciting sources are the photons that interact with the studied sample. In the former, for each shake, the results obtained are faces of a device; in the latter, the result is the emitting reflectance light of the sample. Both systems are described

by a Poisson statistics [Poisson (1837)]. That is, the nature of the studied events is the same; therefore, the conclusions extracted from our example should also hold for the results of linear spectroscopy techniques.

Therefore, it is not clear the advantage of training a NN for evaluating a measured spectrum c since the best performance for a NN would be as good as the statistical method performance, and gives no information about the determination uncertainties ($\Delta\alpha_1$ and $\Delta\beta_1$).

Finally, we mention a common representative example that shows that NN cannot perform better than statistical methods, when analyzing a purely statistical system, as is the case of roulette, obtained from gambling activities. We may assume that with adequate training, a NN can perform better than statistical methods, and then win systematically in the roulette. However, even when the NN have been worked on for decades, they have not affected (and will not affect) the economical profits of the casinos, which in each event simply have more probabilities to win than to lose. Identical conclusion can be applied to the lottery. It would be economically profitable to educate a NN to tell us the next winning lottery number. Unfortunately, the result produced can not improve the probability of occurrence of each number. Moreover, if NN systematically win in gambling activities (like the roulette), we may interpret that they would be violating the principle of causality.

Based on this description, when analyzing a purely statistical system, the use of NN cannot be more convenient than statistical methods. Thus, when using linear spectral techniques in chemical sciences, the same conclusion remains.

4. Radiation Transport Equation and Monte Carlo Methods

The following section describes Boltzmann's Equation because it studies the behavior and distribution of subatomic particles (neutron, electron, photon, etc.). The solution of the transport equation can describe the results of spectroscopic techniques, which use those particles as a tool in experiments. We decided to describe the transport equation applied to the case of neutron transport because there are obvious examples, from nuclear reactor experience, which corroborate the deduced conclusions.

Time, space and energetic distribution of a neutron population are described by Boltzmann's equation. The source characteristics and cross sections of the materials must be known. Some considerations should be taken to solve this equation: (1) The system is assumed to be in a stationary equilibrium, so that the neutron distribution does not depend on time. (2) Neutrons are supposed to be non-charged punctual particles, with a constant mass and position and velocity well-defined. In fact, neutrons also behave like waves; being ruled by Heisenberg's uncertainties, although these aspects are not very important in their behavior or they can be incorporated in nucleus cross-sections. (3) Neutron-neutron interaction is not considered since the usual particle abundance (even in environments with high flux) is much lower than atoms in air. Therefore, the transport equation solution for a set of sources

is equal to the sum of solutions obtained separately for each source. (It is a linear system on the source intensity). This is an important result for the analysis that we are carrying out because the history of each particle (the reaction it produces) is independent of the history of the rest of the particles emitted by the source.

Boltzmann's equation can be expressed according to angular flux ($\Phi(\vec{r}, \vec{v})$) like [Stamm'ler (1983); Zweifel (1973)]:

$$\vec{\Omega} \cdot \vec{\nabla}\Phi(\vec{r}, \vec{v}) = S(\vec{r}, \vec{v}) + \int d^3\vec{v}\,'\Phi(\vec{r}, \vec{v}\,')\Sigma_s(\vec{r}, \vec{v}\,' \rightarrow \vec{v}, \vec{\Omega}' \cdot \vec{\Omega}) - \Sigma(\vec{r}, \vec{v})\Phi(\vec{r}, \vec{v})$$

(1)

where velocity vector $\vec{v}$ is expressed as:

$$\vec{v} = v \cdot \vec{\Omega}$$

being $\vec{\Omega}$ the unit vector that has the neutron velocity. Angular flux $\Phi(\vec{r}, \vec{v})$ can be interpreted as the number of neutrons which cross with velocity $\vec{v}$ a surface located in $\vec{r}$, with its normal parallel to $\vec{\Omega}$, per cm^2, steradian, and unit velocity.

The source $S(\vec{r}, \vec{v})$ emits neutrons with some spatial and velocity distribution. The total cross-section $\Sigma(\vec{r}, \vec{v})$ is proportional to the interaction probability per centimeter of neutron travel, undergoing any kind of reaction (i.e. fission, absorption). The differential scattering cross-section $\Sigma_s(\vec{r}, \vec{v}\,' \rightarrow \vec{v}, \vec{\Omega}' \cdot \vec{\Omega})$ is proportional to the neutron scattering probability in the physical system, per centimeter of travel through a given material, in order to carry out the indicated changes in the neutron velocity, direction and module.

Equation (1) means that in a certain point of space, the amount of neutrons with a given velocity and direction will change according to: (1) Source, if there is a production of neutrons at that point, with the given velocity and direction; (2) The "in scattering" process, if neutrons in this place, modify their original velocity and direction to those being considered, (3) The "out scatter" process, if neutrons with the considered velocity and direction in the point of the space, modify their direction or velocity (or both) undergoing any reaction.

Transport equation can be solved by two totally different methodologies: (1) Deterministic or (2) Probabilistic methods called Monte Carlo. Deterministic methods solve the transport equation for the average particle behavior and its numeric resolution obtains neutron distribution in the whole phase-space (e.g. the discrete ordinates method). Alternatively Monte Carlo methods do not solve any explicit equation but obtain answers simulating many individual particle stories, duplicating theoretically a statistical process (such as the interaction of nuclear particles with materials) and they collect the information about their average behavior. The possibilities that rule the interaction and possible reactions of each particle in the physical system are statistically sampled (from its birth to its death), so the set of "events" accumulated by the particles in the system describe the real physical phenomenon. In particle transport, the Monte Carlo technique is pre-eminently realistic — a numerical experiment. The events sampling process that can occur is

based on the selection of random numbers, determining if any interaction occurs and the type of interaction produced. Each event is ruled by Poisson statistics and the composition of those events is also ruled by a Poisson statistics.

Monte Carlo is well suited to solving complicated three-dimensional, time-dependent problems. This method does not need any approximation in space, energy or time; it allows a detailed representation of all aspects of physical data. The Monte Carlo MCNP code [Briesmeister (2006)] (probably the most used MC code) is a general-purpose Monte Carlo N-Particle code that can be used for neutron, photon, electron, or coupled neutron/photon/electron transport, including the capability to calculate eigenvalues for critical systems.

As a common practice in nuclear reactor design, the results provided by codes are matched against a wide set of specific experiments (benchmarks) in order to validate the "calculation line." Then, it is possible to infer a system behavior in hypothetic conditions, much more complicated to put into practice than to be modeled by transport codes.

Monte Carlo codes are standard tools in critical systems design. The example of a nuclear reactor is chosen because it shows a strong non-linear response in relation to many constituents — e.g. volume and shape of components, fuel enrichment, moderator quantities, position of the control bars, etc. For specific values of those elements, the system is critical and the nuclear reaction remains the same. Slight changes of any of those values in one direction will produce a subcritical system, and the nuclear reaction decreases exponentially. If there are tiny changes in the opposite direction, the system becomes supercritical and the rate of reaction increases.

Section 3 shows that NN methods can not be better performed than statistical methods when analyzing a sequence of statistically independent linear events. Here we deduce that if the system is non-linear, but composed of well-known statistically independent events, statistical methods of analysis (like Monte Carlo methods) give correct results because they accurately represent each step of the total sequence that composes the global phenomena; and thus, NN can not be better performed in those systems either.

5. Total Reflection X-Ray Fluorescence

Total reflection X-ray fluorescence (TXRF) is a surface multi-elemental analysis technique used for the ultra-trace analysis of particles, residues, and impurities deposited on smooth surfaces. TXRF is basically an energy dispersive X-Ray Fluorescence (XRF) technique in a special geometry [Klockenkamper (1997)]. An incident beam impinges upon a sample at angles below the critical angle of external total reflection for X-rays resulting in reflection of almost 100% of the excitation beam photons. Due to its unique configuration, the main advantage of TXRF over conventional XRF is reduced background measurement by eliminating of sample scattering, resulting in increased elemental measurement sensitivity — detection limits are on the order of 10^{-8} g/g.

The basic equation for TXRF for a thin film is given by [Markowicz (1993)]:

$$I_i = K_i I_0 N_0 \sigma_i \omega_i m_i / A_i \tag{2}$$

where the intensity of the line originated in the element i, I_i, is written in terms of the following parameters: K_i depends on experimental geometry and energy detection efficiency; I_o stands for the intensity of the excitation source; the Avogadro's Number N_o; m_i represents the mass surface density of the element i with atomic number Z_i and atomic mass A_i, the element i collision cross section σ_i and the photon emission probability per ionization ω_i.

As is deduced from Eq. (2), the measured intensity of line I_i doubles if the source intensity or the mass of the sample is doubled. We may consider under what conditions the technique become non-linear. If the amount of mass analyzed increases systematically, three kinds of non-linear effects will appear: (A) The exciting source will suffer an attenuation inside the sample and the inner material will become less studied (it will become protected or "hidden" by the outer atoms). (B) The emerging radiation produced inside the sample will also be attenuated in its travel outside the sample and will arrive at the detector with decreased intensity — this attenuation is in a different proportion for the photons produced by each element of the sample. (C) The fluorescent radiation emitted by the sample could produce an enhancement effect, exciting other atoms in its travel away from the sample. That is, the signal produced by an element can depend on the presence of other elements in deeper places.

Thus, if the mass of the sample is excessive, doubling the mass of the sample will not double the intensity of the measured spectrum, and the linear relationship proposed in Eq. (2) is no longer valid.

This technique analyzes very small amounts of sample, where is not expected any non-linear effect, being then an example where NN methods would not be perform better than statistical methods.

6. Neutron Activation Analysis

Neutron activation analysis (NAA) is a technique for quantitative multi-element analysis of major, minor, trace and rare elements. NAA begins with neutron bombardment of a sample to convert stable isotopes into radioactive isotopes. The usual procedure involves placing the samples plus a number of suitable standards into a neutron field. Radioisotopes created during the irradiation will decay with time. A portion of the energy released during decay is often in the form of gamma radiation, which can be detected. The gamma rays possess unique energies and characteristic evolutions in time that are distinctive of the radioisotope undergoing decay. Gamma rays detected are indicative of a specific radionuclide's presence.

The basic equation for NAA [Alfassi (1990)] is quite similar to Eq. (2), which also is linear on the source and on the amount of mass analyzed, but it is not described here. In this technique it is difficult to expect any non-linear effect

because: (A) the excitation source (neutrons) shows very little attenuation inside the sample because it doesn't have electric charge, which allows deep penetration into matter. (B) the sample size could be small ($\sim$0.1 g). (C) the gamma radiation produced by the sample usually has appreciable energy ($E \geq 100$ keV), which usually suffers unappreciable attenuation in its travel to the detector where it is observed.

The NAA would be a typical example where NN methods would not perform better than statistical methods.

7. Conclusions

In Sec. 3 we have shown that linear spectroscopy results are equivalent, from a statistical point of view, to a simple linear independent sequence — e.g. a succession of coin tosses. Analyzing that kind of data, according to the maximum likelihood principle [Barnard (1962); Berger (1988)], there is only one method that produces the best results. If another method is chosen, it would be a conceptually bad application of a tool of data analysis, and the procedure would be redundant or inefficient — in the sense that it loses information. If a NN is used for that analysis, some information will be lost, affecting concepts such as accuracy, detection limit and determination limit, associated with any determination. Moreover, one could require an explanation about the limit of validity of the results.

As a common example that shows that NN cannot perform better than statistical methods, when analyzing a purely statistical system, we mention the case of the roulette, obtained from gambling activities. We may assume that with adequate training, a NN can perform better than statistical methods, and then win systematically in the roulette. However, even when the NN have been worked on for decades, they have not affected (and will not affect) the economical profits of the casinos, which in each event simply have more probabilities to win than to lose. Identical conclusion can be applied to the lottery. It would be economically profitable to educate a NN to tell us the next winning lottery number. Unfortunately, the result produced cannot improve the probability of occurrence of each number.

In Sec. 4, we deduced that, if the system is non-linear but composed of well-known statistically independent events, statistical methods of analysis (like Monte Carlo methods) give correct results because they accurately represent each step of the total sequence that composes the global phenomena; thus the NN cannot perform better in those systems either.

In Sec. 4, describing the properties of the transport equation we mentioned that if there is no interaction between the particles emitted by the source, then the measured signal always shows a linear response to the excitation source because each particle's history will be independent of the other particle's history. If there is no interaction between the particles emitted by the source, the transport equation's results are always proportional (linear) with the intensity of the excitation source.

This property is not true if we increase indefinitely the sample size. The characteristics of a spectroscopic technique should be closely studied before the use of a NN.

Analyzing spectroscopic techniques such as TXRF or NAA, whose results consist of a sequence of statistically independent linear events, the NN methods would not perform better than statistical methods.

The results of a NN can be understood as an approximation since it has a simple implementation sequence. If a mathematical description of the physical phenomena studied is possible, as is expected in well-known spectroscopic techniques, then the statistical methods should perform better than any NN. But, if there is not a mathematical description of the system (like Pattern recognition case) or the system shows a not well-predictable behavior (economical or sociological reactions, etc.), in these case NN would perform better. Those are clearly not the case of spectroscopic techniques.

Acknowledgement

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