A New Method Based on Adaptive Neuro-Fuzzy Inference System for Determination of Acid Molarity Using Compton Scattered Photons

Okhtay Jahanbakhsh*, Saleh Ashrafi, and Davood Alizadeh
Faculty of Physics, University of Tabriz, Tabriz, 51666-16471 Iran
*e-mail: o.jahanbakhsh@tabrizu.ac.ir
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Abstract—The simplicity and accuracy in determining the exact concentration of a particular substance in a solution, is one of the major issues in industrial chemistry. In this paper, we introduce a new technique based on Compton scattering of gamma photons and artificial intelligence to determine the concentration of a solute in a solution. We used a $^{137}$Cs gamma ray source with few millicurie activity and a NaI(Tl) scintillator detector to determine the acid sulfuric molarity. We also applied Adaptive Neuro-Fuzzy Inference System (ANFIS) and Artificial Neural Network (ANN) as artificial intelligence techniques to estimate the molarity of the samples. Monte Carlo simulations also support the present experimental results. The results of the proposed methods are in excellent agreement with the measured molarities.

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1. INTRODUCTION

The exact concentration of a substance in a solution, is frequently inquired by medical and industrial laboratories. A widely used classical technique to determine the unknown concentration of any given solution is titration. There are several requirements for analytical titrations. The chemical reaction involved in a titration must be known, quick, complete, unambiguous and observable. However, this method is time-consuming, and when done manually, it is somewhat ambiguous. The most inaccurate part of the titration is deciding when the color change of the indicator occurs. This is hard because although the color change is fairly rapid, it is not always instantly obvious. Furthermore, it needs particular skill and practice for effective results and requires appropriate judgment on what indicator and test to use [1, 2]. Hence it is desirable to evaluate alternative and easier methods to estimate the desired parameters.

In recent years, several detection methods based on nuclear techniques have been suggested for the measurement of density, atomic composition ($Z$) in industrial and medical fields. These techniques have a number of advantages such as: relative safety, reasonable measurement accuracy, quick response and non-destructiveness [3–8].

The purpose of this study is to investigate the possibility of using Compton scattering method to measure the molarity (number of moles of solute dissolved in one liter of solution) of sulfuric acid solution. This method provides an effective tool for inspecting materials, because the number of photons scattered from a sample depends on the electron density of the scattering medium. A detailed analysis of the measured spectrum shows that the number of scattered photons and molarity of examining sample is closely related to each other and by increasing the acid molarity, the number of scattered photons increases. Because of the complexity of the relationship between the number of scattered photons and molarity of acids we used Artificial Neural Network (ANN) [9–11] and Adaptive Network-based Fuzzy Inference Systems (ANFIS) [12, 13] as artificial intelligence techniques to estimate molarity with changes in the number of scattered photons. Comparing the results of the ANFIS with ANN and experimental results shows the good performance of this method to determine the molarity of acid and validation of obtained model. The relative error is less than 3%. Therefore, by using this method, users can get reliable results.

1 The article is published in the original.
2. THEORY

2.1. Compton Scattering

Depending upon the photon energy and the atomic number of the absorber, gamma-ray interacts with a matter through four effects, Rayleigh coherent scattering, photoelectric absorption, Compton scattering and pair production [14, 15]. The Compton scattering is the dominant mode of interaction in the case of intermediate photon energies and Low-$Z$ materials. It refers to the scattering of photons by electrons, when the binding energy of the electrons is much lower than the energy of the incident photons. In such collisions, the photon loses an amount of energy that depends on the incident photon energy $E_0$ and scattering angle $\theta$.

The energy of the scattered gamma ray $E$ is given by

$$\frac{1}{E(\theta)} = \frac{1}{E_0} + \frac{1}{m_e c^2} (1 - \cos \theta), \quad (1)$$

where $m_e c^2$ is the rest-mass energy of the electron.

The number of singly scattered photons that reach to the detector could be written as:

$$f = GI_0 \exp(-x_{in} \mu(E)) n_e \frac{d\sigma^{KN}}{d\Omega} \exp(-x_{sc} \mu(E)). \quad (2)$$

Here, $I_0$ is the intensity of the incident photons; $G$ is a geometrical constant based on the setup geometry; $d\sigma^{KN}/d\Omega$ is known as the Klein–Nishina differential scattering cross-section; $n_e$ is the electron density of scattering target and two exponential functions represent the gamma-ray attenuation in the incident path from the source to the scattering center ($x_{in}$) and in the scattered path from the scattering center to the detector ($x_{sc}$), respectively.

For a fixed geometry and given incident photon energy and intensity, the Klein–Nishina differential cross-section is constant [16, 17]. Therefore the term $GI_0 \frac{d\sigma^{KN}}{d\Omega}$ will be constant and Eq. (2) can be written as:

$$f = C n_e \exp(-x_{in} \mu(E)) \exp(-x_{sc} \mu(E)). \quad (3)$$

Any changes in the molarity of the scattering sample would affect its electron density ($n_e$) and gamma-ray attenuation coefficients for the incident and scattered photons ($\mu(E)$ and $\mu(E_0)$). So the number of scattered photons reached to the detector is a function of sample molarity as bellow:

$$f = F(m, Z_w, \rho_w, A_w, Z_a, \rho_a, A_a, E, E_0), \quad (4)$$

where $m$ is the molarity of the acid; $Z_w$ and $Z_a$ are the number of electrons associated with a molecule of water and pure sulfuric acid (H$_2$SO$_4$) respectively; $A_w$ and $A_a$ are the atomic mass of water and of pure sulfuric acid; $\rho_w$ and $\rho_a$ are the mass density of water and of pure sulfuric acid. In the Eq. (4) all parameters except $m$ (molarity of the sample) are known and by counting the number of scattered photons ($f$), the molarity of the acid samples can be obtained.

2.2. Artificial Neural Network (ANN)

Artificial neural network (ANN) is a modeling method, similar to the human brain. The ANN is used extensively in engineering and technology to produce a particular output based on the input [10, 11]. Neuron is processing element in ANN, that receives input data and sums them with applying weights to each data, and finally applies to the transfer function. The transfer function decides the neuron output. There are exist three different types of layers in ANN namely input-layer, hidden layers and an output layer, which each of them consists of one or more neurons. The schematic form of the artificial neural network is shown in the Fig. 1.

2.3. Adaptive Network–based Fuzzy Inference Systems

Adaptive Neuro–Fuzzy Inference System (ANFIS) is a hybrid artificial intelligence technique that has attracted growing interest in various scientific and engineering areas. It is a simple data learning technique and has the ability to transform a given input into a target output using Artificial Neural Network (ANN) and fuzzy logic control. Fuzzy inference systems (FIS) are well known applications of fuzzy logic.
theory [18, 19]. A typical ANFIS architecture is shown in Fig. 2, which contains two inputs \((x, y)\), four rules and five layers. Each layer described by the node function and consists of adaptive and non-adaptive nodes with a single output.

The first layer is the fuzzy layer and fuzzification of input data is performed here. Every node in this layer is an adaptive node with node membership functions (MFs). Some examples of membership functions are shown in Fig. 3 [20, 21].

The second layer involves fuzzy operators and every node in this layer is a non-adaptive node. Each node multiplies incoming signals and the output is the product of all the incoming signals. The node output represents the firing strength of a rule. In third layer every node computes the ratio of the \(i\)th node firing strength of the sum of all rules firing strength and the output from this layer are the normalized firing strengths. The fourth layer consists of adjustable nodes that each one contribution of the \(i\)th rule to the overall output. In this stage, a normalized signal is gained again through a linear equation that is formed from the membership function of the output signal. The single node in fifth layer is a non-adaptive node and calculates the overall output as the summation of all input signals [20–22].

3. EXPERIMENTAL SETUP

In the present study, a \(^{137}\text{Cs}\) point source (of strength 5 mCi emitting 662 keV gamma rays) with stainless-steel cylindrical encapsulation was used; where the thickness of the capsule is sufficient to absorb all the beta particles and 32 keV \(K\) X-rays emitted due to internal conversion processes. To reduce the biological hazards and minimize the intensity of the source photons reaching directly to the detector, the source was enclosed in a lead container with one face aperture of 5 mm. Sulfuric acid solutions with different concentration were prepared and each sample filled in a test tube of diameter of 1 cm and placed in the front of the gamma beam.

A 3in \(\times\) 3in NaI(Tl) scintillation detector used for measuring of the scattered photons. The detector was placed at the scattering angle of 90° to detect the radiation scattered from the target and it was shielded with lead blocks to minimize the background radiation. The energy resolution \((R)\) and photo-peak efficiency \((\epsilon)\) of the scintillator at 288 keV (the energy of the scattered photons) were 10% and 4%, respectively and the distance between the sample and the detector was fixed at 6 cm. A schematic representation of the experimental setup is shown in Fig. 4.
We used the MCNP code for simulating the experiments and evaluating the effect of density on the number of scattered photons. The code is widely used for complex geometry and continuous-energy problems. The structural and geometrical information (dimensions and materials) of the experimental setup was introduced to the code and the net intensity of the scattered photons was obtained from the simulated spectrum by integrating the counts in the energy window from 188 to 388 keV.

4. RESULTS AND DISCUSSION

Energy spectra of the photons scattered from the samples were recorded by a PC-based ATOMTEX Multi Channel Analyzer (MCA). The spectrometer was calibrated by using standard calibration sources. The net intensity of scattered photons was computed from the difference between the gross counts, and the background counts in a 200 keV energy interval around 288 keV \((E(90^\circ))\). Figure 5 shows a typical observed spectrum.

Figure 6 shows the experimental and simulated results of the intensity of the detected photons as a function of sample molarity for various acid concentrations. As mentioned previously, the Compton scattering cross section is sensitive to the electron density of scattering volume. By increasing the molarity of samples, the density of electrons in unit volume from the difference between the gross counts, and the background counts in a 200 keV energy interval around 288 keV \((E(90^\circ))\). Figure 5 shows a typical observed spectrum.

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>Real molarity</th>
<th>Number of detected photons ((f))</th>
<th>Molarity estimated by ANFIS</th>
<th>Molarity estimated by ANN</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0</td>
<td>297267</td>
<td>0.01</td>
<td>0.02</td>
</tr>
<tr>
<td>2</td>
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<td>305831</td>
<td>1.01</td>
<td>1.03</td>
</tr>
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<td>2.02</td>
<td>2.05</td>
</tr>
<tr>
<td>4</td>
<td>4.0</td>
<td>332923</td>
<td>4.10</td>
<td>4.13</td>
</tr>
<tr>
<td>5</td>
<td>5.5</td>
<td>345386</td>
<td>5.61</td>
<td>5.67</td>
</tr>
<tr>
<td>6</td>
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<tr>
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<tr>
<td>11</td>
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<td>17.86</td>
<td>18.19</td>
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</tbody>
</table>

Obtained value for molarity by ANFIS and ANN models

<table>
<thead>
<tr>
<th></th>
<th>Molarity estimated</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>by ANFIS</td>
</tr>
<tr>
<td>Maximum relative error percentage</td>
<td>2.5%</td>
</tr>
</tbody>
</table>
increases, which consequently increases the number of scattered photons.

The number of scattered photons from targets with various molarities, which were recorded by a NaI scintillator, are applied as the input to the ANN and ANFIS network and the output data is the molarity of samples. The results obtained by the artificial models are shown in table. It can be observed that the results of the ANFIS model have less error than the results obtained by the ANN.

To check the performance of the ANFIS model, the root mean square error (RMSE) and the coefficient of determination ($R^2$) were calculated. As is shown in Fig. 7 the low value of RMSE (0.616) and value of coefficient of determination (0.987) close to 1, indicate the good reliability of the proposed model.

5. CONCLUSIONS

In this paper, we introduced a new method based on the Compton scattering of gamma photons and artificial intelligence technique to non-destructively measure the molarity of the acid samples. We used a gamma source and a NaI(Tl) scintillation detector for measuring the sulfuric acid molarity and we showed that this method can be an alternative to classical titration. This approach has an obvious advantage because it does not require a priori knowledge about the titration reaction and it is very simple, so it does not need professional skill. We also used Adaptive Network-based Fuzzy Inference Systems (ANFIS) and artificial neural network (ANN) to estimate morality of samples by analyzing the number of scattered photons from each sample. Obtained results by ANFIS show relative error less than 3%, and it can be used in industrial and laboratory measurements with confidence.

REFERENCES


