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Utilizing Gamma Ray Spectrometry for Enhancing Food and Agriculture Quality Assurance

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Abstract - This research paper explores the application of gamma ray Spectrometry as a powerful tool to enhance quality assurance in the fields of food and agriculture. Gamma ray Spectrometry has gained prominence for its non-invasive and highly sensitive capabilities, allowing for the rapid and precise assessment of various quality parameters. Through a comprehensive review of the literature, this paper discusses the theoretical and practical aspects of gamma ray Spectrometry, as well as its potential implications in food safety, crop quality, and agriculture productivity. The methodology section details the instrumentation and techniques used for data collection and analysis. Furthermore, this paper discusses real-world applications in food quality control, including contaminant detection and nutrient analysis, as well as in agriculture quality control for soil and crop monitoring. It also addresses the challenges and limitations associated with gamma ray Spectrometry, along with safety considerations and regulatory aspects. The paper concludes by highlighting the future.

Theory of Gamma Spectrometry:

i. Gamma rays are electromagnetic radiation produced during transitions between excited nuclear levels of a nucleus.
ii. Delayed gamma rays are emitted during the decay of parent nuclei, often following beta decay.
iii. Gamma rays can interact with matter through three main processes:
iv. Photoelectric Absorption: Gamma rays interact with inner-shell electrons, emitting photoelectrons. This is important for detection with semiconductor detectors.
v. Compton Scattering: Some of the gamma ray's energy is transferred to a recoil electron, resulting in a continuous background in the gamma spectrum.
vi. Pair Production: Occurs when gamma rays have more than 1.02 MeV energy, producing an electron/positron pair.

i. Semiconductor Detectors:

ii. Germanium detectors (Ge-detectors) are commonly used for gamma spectrometry.
iii. Early Ge-detectors had to be doped with n-type impurities like lithium (Ge (Li)-detectors) due to purity issues.
iv. Later, pure germanium crystals became available in n-type or p-type forms, with various geometries (closed-end coaxial, planar, borehole).
v. n-type detectors cover an energy range from about 10 keV to 3 MeV, while p-type detectors range from 40 keV to 3 MeV. P-type detectors with certain window materials are best for energies below 100 keV.

Requirements for Proper Gamma Spectrometry:

i. The Minimum Detectable Activity (MDA) of a Ge-detector depends on various factors, including energy resolution, crystal efficiency, background, measuring time, sample geometry, selfabsorption, and the emission probabilities of the gamma emission lines of the radionuclide.
ii. Commercial calibration sources containing a mix of gamma nuclides are used for detector calibration. These sources cover a range of energies.
iii. Calibration sources typically include radionuclides like 210Pb, 241Am, 109Cd, 139Ce, 57Co, 60Co, 134Cs, 137Cs, 88Y, and 85Sr.
iv. Calibration should include energy response, peak resolution, and counting efficiency.

It's important to ensure that gamma spectrometry equipment is properly calibrated and maintained to obtain accurate results. The choice of calibration sources and the correction of summing effects in complex spectra are important aspects of the calibration process. The field of gamma spectrometry plays a vital role in monitoring and managing radiation levels in various applications, from food and environmental analysis to industrial and medical settings. [1][2]

Background:

i. The background of a Ge-detector system is influenced by detector shielding and the laboratory environment.

ii. Shielding materials, such as lead and copper, can be used to reduce background radiation.

iii. Reducing background radiation is essential for accurate measurements, and efforts should be made to minimize cosmic muon interference.

Attenuation Effects:

i. Photon attenuation depends on the elemental composition and density of the sample matrix.

ii. Attenuation effects are more significant for lower energy photons and high-volume sample geometries.

iii. Software, including Monte Carlo simulations, is used to account for attenuation in gamma spectrometry.

Coincidence Summing Effect:

i. Coincidence summing occurs when radionuclides emit multiple photons in sequence.

ii. The effect depends on the source-detector distance and detector efficiency.

iii. To avoid this effect, samples may be counted at a certain distance from the detector.

Dead Time:

i. High-activity samples can result in a loss of peak counts due to dead time.

ii. Dead time occurs when pulse processing electronics cannot keep up with incoming photons.

Proper sample geometry and distance from the detector can help avoid dead time.

Best Sample Geometries: The Minimum Detectable Activity (MDA) depends on the detector efficiency and sample weight.

i. Marinelli beakers (1 or 2 L) are suitable for large sample amounts and provide good efficiency.

ii. Smaller sample devices (e.g., 32- and 77-mL dishes) are better for small sample amounts, reducing gamma ray attenuation.

Interpretation of Gamma Spectrometry Data:

i. Dose-relevant radionuclides in the natural decay series of uranium, thorium, and actinium can be detected via the gamma emissions of their daughter nuclides.

ii. Equilibrium between mother and daughter nuclides is essential for accurate measurements.

iii. Some mother-daughter systems, such as 226Ra and 224Ra, are used for radium detection.

iv. Equilibrium is reached within minutes for certain systems due to short half-lives.

v. Relevant gamma emission lines for various natural radionuclides are listed in **Table 1.**

(see in last page)

Gamma spectrometry is a powerful technique for quantifying and identifying radioactive materials in various samples, and understanding these factors and techniques is crucial for accurate and reliable analysis. [4][3]

Instrumental Neutron Activation Analysis (INAA) is a nuclear technique used for the analysis of various materials, particularly to determine the elemental composition of samples. Here are some key points regarding the principle and operation of INAA, as described in the provided text:

Principle:

i. INAA relies on the production of short-lived radionuclides through nuclear reactions.

ii. Typically, reactor neutrons, particularly thermal neutrons, are used to activate various nuclides in the sample, turning them into radioactive nuclides.

iii. The efficiency of the irradiation process depends on two factors: the neutron flux density and the cross-section of the nuclear reaction that occurs in the irradiated nucleus.

iv. Nuclear reactors, such as the one at the University of Basel (AGN-211-P), are commonly used as sources of thermal neutrons for INAA.

v. The thermal neutrons are generated in the nuclear reactor, and their availability is crucial for the success of the analysis.

INAA is a powerful analytical technique that is particularly useful for determining trace elements in various materials. The irradiation process leads to the creation of radioactive isotopes, and the subsequent measurement of their gamma-ray emissions can provide valuable information about the elemental composition of the sample. The unique neutron flux and reactor setup described here are crucial for the success of INAA experiments.

Operational Procedure:

i. The detection limit of gamma spectrometry is influenced by the half-life of activated nuclides and the background radiation from highly activated nuclides, like sodium or chloride.

ii. Gold foils serve as internal standards and are placed on top of each sample.

iii. A sample series, consisting of 12 samples, each containing 1-2 grams of material, is irradiated for 30 minutes at a reactor power of 2 kW.

iv. Calibration of each irradiation position in the reactor's neutron field is achieved using coagulated salt solutions containing a known amount of the analyte and a corresponding gold foil. This helps determine response factors for each analyte.

v. The response factors are calculated using the comparator method.

Common Applications of INAA: Determination of Total Bromine Content in Food Samples:

INAA is used to determine the total bromine content in food samples, such as tea, coffee, dried mushrooms, vegetables, and spices.

i. This analysis provides insights into the use of methyl bromide, a fumigant, which results in bromide residues in food.

ii. Methyl bromide's bromide residues can be activated by neutrons to form the gamma-active compound 82Br (with a half-life of 35 hours) and analysed using gamma spectrometry.

iii. After irradiation for 30 minutes, samples are cooled down for several hours to allow for the disintegration of activated sodium and chloride nuclides.
iv. The gamma analysis of the samples takes approximately 15 minutes.

It's important to note that the use of methyl bromide as a fumigant has become less common in recent years, with other fumigants like sulfuryl fluoride, hydrogen cyanide, and phosphines gaining more prominence for various food treatment applications. INAA is a valuable technique for identifying and quantifying the presence of specific elements, such as bromine in this case, in food samples, aiding in quality control and regulatory compliance.

Determination of Total Iodine Content in Food:

i. INAA is used to determine the total iodine content in various food samples, particularly in iodine-rich foods like algae.
ii. Iodine is essential for the production of thyroid hormones and the prevention of goitre.
iii. In many European countries, there is a concern about iodine deficiency, so it's important to assess and declare the correct iodine content in food.
iv. About 1 gram of the sample is activated using reactor neutrons for 30 minutes at a power of 2 kW.
v. The radioactive product 128I is then analysed directly using a gamma spectrometer.
vi. The short half-life of 128I (25 minutes) necessitates immediate counting after activation, which can lead to higher detection limits.
vii. This method is applicable to the analysis of iodine content in fish, seafood, algae, and dietary supplements.

Determination of Flame-Retarding Agents in Plastics:

i. INAA can be employed as a screening analysis to detect flame-retarding agents in plastic materials, such as decabromo-bis-phenylether or tetrabromobisphenol A.
ii. The activation and decay processes for bromine analysis in plastic materials are similar to those used for food samples.
iii. INAA provides information about the total content of brominated flame-retarding agents.
iv. Samples with high bromine content are detected using this screening analysis, and further analysis with gas chromatography is conducted to determine the types and amounts of different flame-retarding compounds.

Determination of U and Th in Suspended Matter, Sediment, and Soil Samples:

i. INAA is applied to determine the content of uranium (U) and thorium (Th) in suspended matter, sediment, and soil samples.
ii. Approximately 1 gram of dried and ground material is irradiated for 30 minutes at a power of 2 kW.
iii. After a 2-hour cooling period, the samples are analysed using a gamma spectrometer.
iv. This method is useful for environmental studies and assessing the distribution of U and Th in various geological and environmental samples.

Instrumental Neutron Activation Analysis is a versatile analytical technique used in various applications, allowing for the determination of the elemental composition of samples and enabling the detection of specific elements, like iodine and bromine, in different materials, including food and plastics.

Radioactivity In Food

First Use of Natural Radioactivity:

i. After the discovery of radioactivity, the commercial use of technologically enriched naturally occurring radioactive material (TENORM) began.
ii. Radium and thorium were used for medicinal purposes to treat various diseases.
iii. Products like underwear, soap, lipstick, hair shampoo, toothpaste, and more were spiked with natural or enriched radioactive materials.
iv. Some individuals, like William Bailey, sold radioactive sources as medicinal drugs.
v. Tragedies like the case of Eben McBurney Byers, who died due to radiator therapy, and the "radium girls" who became ill from painting watch dials with radium, led to public awareness and the decline of the popularity of radioactivity.
vi. Today, radon therapy, which involves radon water, inhalation of radon air in tunnels, and drinking of radon water, is still used for health cures against certain chronic diseases.

Natural Radioactivity in Food:

i. Some natural radionuclides from the decay series of uranium and thorium enter the food chain.
ii. Polonium-210 (210Po), a product of the decay of uranium-238 (238U), is enriched in the intestinal tract of mussels and fish.
iii. Lead-210 (210Pb), radium, and thorium nuclides are present in cereals.
iv. Spices and salt may contain elevated levels of radium and potassium-40 (40K).
v. Foods rich in potassium are typically rich in 40K.

Radioactive Sources in Consumer Products:

i. Remnants of natural radionuclide applications from the past may still be found in households.
ii. Radioactive objects may include materials like thorium used in flame detectors and various consumer products.
iii. Examples of such products include dials with radium in watches, colored glass pearls, drinking glasses containing uranium oxides, wall tiles with uranium oxides, and more.
iv. Finders of such items are encouraged to bring them to specialized laboratories or collecting points for radioactive materials, as the included radioactive material may be harmful. [5][6]

Consumer product	**Radionuclide(s)**	**Radionuclide content range**
Radio luminous timepieces	^{3}H 147Pm 226Ra	4–930 MBq 0.4–4 MBq 0.07–170 kBq
Marine compass	^{3}H 226Ra	28MBq 15kBq
Aircraft luminous safety devices	^{3}H 147Pm	10 kBq 300 kBq
Static eliminators	210Po	1–19 MBq
Dental products	natU	up to 4 Bq
Gas mantles	232Th	1–2 kBq
Welding rods	232Th	0.2–1.2 kBq
Optical glasses Ophthalmic lenses [35]	232Th	5–75 Bq
Glassware: vaseline glass, canary flint glass	natU	100 kBq
Lamp starters	^{85}Kr	0.6 kBq
Smoke detectors	241Am	37 kBq
Electron capture detectors	^{63}Ni	370 kBq
Drinking devices “Radium Drinkkur”	226Ra, (222Rn)	100 MBq
Wall tiles, ceramics	natUO3	50–500 kBq
Granitic surfaces	natU	5–10 kBq/kg
Cardiac pacemaker [36]	239Pu	113GBq

Table2: Consumer products containing radioactive materials

Radio Contamination of Food:

i. Food can be contaminated shortly after its release with fallout of short-lived radionuclides.
ii. The contamination of food with long-lived radionuclides from global fallout and the Chernobyl catastrophe continues to be a concern.
iii. Contamination by short-lived radionuclides, such as 131I, 132I, and 134Cs, reduced within two years after the Chernobyl disaster.
iv. Long-lived radionuclides persist in the soil and can be transferred to crops and grass, particularly in feed for cows, with milk being a typical tracer food.
v. A review of radioactivity monitoring in
vi. Switzerland over 35 years showed that between 1990 and 2015, some moderate contamination was observed in specific food categories.
vii. Special cases of contamination were found in hazelnuts and tea from Turkey, with tea containing higher levels of radio caesium and radio strontium.

Wild-grown mushrooms, berries, and game, especially wild boars, remain the most affected food categories due to fallout from the Chernobyl disaster.

The monitoring and assessment of radionuclide contamination in food is essential for ensuring food safety and public health, particularly in regions affected by nuclear accidents and global fallout.

Data Analysis : The gamma-ray data were obtained using a portable passive gamma-ray spectrometer called the Mole, which uses a CsI(Tl) scintillation crystal detector. This device can be mounted on various vehicles or used manually. In this study, it was mounted on a wheelbarrow and moved at 1.2 m/s, with a field of view of about 3 meters in radius. Data was collected from eight rows in each field, spaced 10 meters apart, totalling around 4,000 data points, along with GPS coordinates accurate to 1 meter.[6][7]

To process and interpret the gamma-ray spectroscopy data, several steps were taken:

1. **Transformation of Multichannel Gamma-Ray Data**: The data from the 256 energy channels (ranging from 0 to 3.0 MeV) were converted into corresponding energies using a simple equation.
2. **Conversion to Count Rates**: Gamma-ray counts were converted to count rates by dividing them by the measurement time. This standardizes the data for analysis.
3. **Spatial Filtering**: A moving average filter was applied to smooth the data, reducing noise and fluctuations.
4. **Energy Channel Averaging**: Within each spectrum, a moving average of five energy channels was calculated to further reduce noise.
5. Energy-Windows (EWs) Method: Energy windows for specific radionuclides (like 40K, 238U, and 232Th) were determined by summing the gamma-ray counts in the energy spectrum around the peaks of these radionuclides. Additionally, a broad energy window called “Total Counts (TC)” was used. This process helps in identifying and analysing these radionuclides in the data.

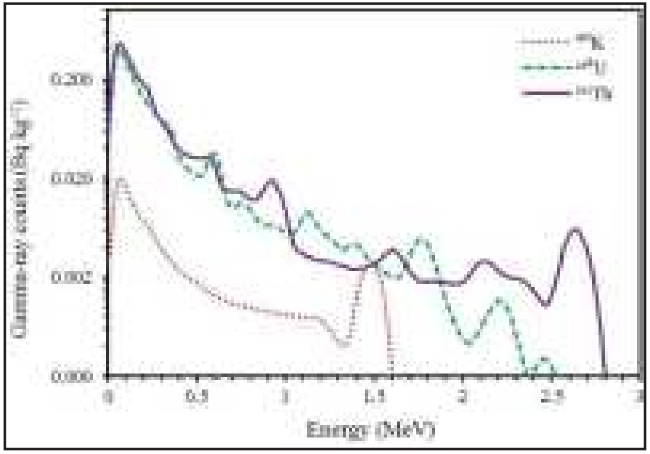

Figure 1: standard spectra of ^{40}K (dotted line), ^{238}U (dashed line) and ^{232}Th (solid line) collected by the Mole at 1Bq·kg^{-1} activity concentration in calibration setup.

The Full-Spectrum Analysis (FSA) method utilizes the entire gamma-ray spectrum to estimate concentrations of radionuclides (e.g., 40K, 238U, and 232Th). In this study, it employs Monte Carlo simulations for this purpose, making it comparable to the Energy-Windows (EWs) method. FSA fits standard spectra for these radionuclides to measured data using a Chi-square algorithm to determine their concentrations in Bq·kg^(-1).

The data analysis involves:

1. **Exploratory Bivariate Analysis**: Using linear regression and correlation to understand relationships between radiometric data and soil and food properties.
2. **Calibration Datasets**: Linear regression of radionuclide concentrations obtained from both EWs and FSA against soil and food properties.
3. **Regression Models**: Developing models from calibration datasets for predicting soil properties in validation datasets.
4. **Model Evaluation**: Assessing model strength using metrics like R^2, RMSEP, and statistical significance tests.
5. **Ratio of Percentage Deviation (RPD):** Calculating RPD to evaluate model prediction ability. RPD > 1.4 indicates potential usability for predicting soil properties.

In addition, spectrum deconvolution techniques are used to identify and separate energy peaks in the raw gamma-ray spectrum, making it easier to distinguish different elements or isotopes.

Statistical Methods Used for Quality Assessment

Descriptive Statistics: Basic statistical measures like mean, standard deviation, and range are employed to summarize the data and understand the distribution of elemental concentrations within the sample.

Inferential Statistics: Inferential statistics, including hypothesis testing and confidence intervals, are used to make inferences about the population of interest based on sample data.

Multivariate Analysis: Techniques such as principal component analysis (PCA) and cluster analysis can be applied to identify patterns and relationships in complex spectral data, helping to differentiate between various samples. [8][9] **9.Challenges and Limitations**

1. Instrumentation Costs: One of the primary limitations is the cost of acquiring and maintaining gamma ray Spectrometry equipment. High-quality detectors and associated instruments can be expensive, making it challenging for smaller farms or laboratories with limited budgets to adopt this technology.

2. Limited Elemental Range: Gamma ray Spectrometry is most effective for the detection and quantification of elements with gamma-emitting isotopes. Elements that lack suitable isotopes or have weak gamma emissions may not be easily analyzed using this method.

3. Limited Depth of Penetration: Gamma rays have limited penetration depth in dense materials. This means that in applications where the material of interest is thick or heavily shielded, gamma ray Spectrometry may not provide accurate results.

4. Sample Size and Homogeneity: The accuracy of gamma ray Spectrometry depends on sample homogeneity. Nonhomogeneous samples can yield inaccurate results. Additionally, small sample sizes can pose challenges, as they may not provide a representative analysis.

5. Safety Concerns: The use of gamma ray sources for activation analysis can introduce radiation safety concerns. Proper training and safety measures are essential to protect operators and ensure regulatory compliance.

Safety Considerations and Regulatory Aspects

1. Radiation Safety: Gamma ray Spectrometry involves the use of ionizing radiation. It is critical to implement safety measures to protect individuals working with the equipment. This includes shielding, proper handling, and adequate training to minimize radiation exposure.

2. Regulatory Compliance: The use of gamma ray Spectrometry may be subject to national and international regulations, particularly in cases involving the use of radioactive materials. Compliance with these regulations is essential to ensure safe and legal operation.

3. Licensing and Certification: Operators and facilities using gamma ray Spectrometry often need to obtain licenses and certifications to handle radioactive materials and operate the equipment. Regular inspections and audits may be required to maintain compliance. [10][11][12]

Future Directions

Emerging Trends and Advancements in Gamma Ray Spectrometry

1. **Miniaturization and Portability**:Ongoing developments are focused on miniaturizing gamma ray Spectrometry equipment, making it more portable and accessible for field applications. This trend opens doors for real-time, on-site analysis in agriculture and food quality control.
2. **Automation and Integration**: Integrating gamma ray Spectrometry with automation and data analytics tools is becoming more prevalent. This allows for faster data processing, real-time decision-making, and seamless integration with other quality control technologies.
3. **Multimodal Sensing**: Combining gamma ray Spectrometry with other sensing techniques like hyperspectral imaging and acoustic sensors enables a more comprehensive assessment of food and crop quality. This multi-sensor approach provides a more holistic view of the sample.
4. **Machine Learning and AI**: Machine learning and artificial intelligence are increasingly applied to gamma ray Spectrometry data analysis. These technologies can improve the accuracy of quality assessments and allow for the detection of subtle patterns and anomalies.
5. **Spectroscopic Imaging:** Advancements in imaging systems are allowing for spatial mapping of elemental composition. This capability can be invaluable for pinpointing areas of concern in larger agricultural fields or during food processing.
6. **Non-Radioactive Sources**: Research into alternative sources of gamma radiation, such as Xray fluorescence, is gaining traction.

Nonradioactive sources reduce safety and regulatory concerns associated with traditional gamma sources.[13][14]

Conclusion: In conclusion, gamma ray spectroscopy represents a valuable asset in the continuous pursuit of safer and more sustainable food production and agriculture practices. As technology continues to evolve and research progresses, its potential to revolutionize quality assurance in these sectors is substantial. Interdisciplinary collaboration and ongoing innovation will be essential in fully realizing the benefits of gamma ray spectroscopy in the

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Radionuclide		**Energy (keV)**	**Emission probability ε (%)**	**Interferences**
7Be	Direct	477.61	10.3	
40K	Direct	1460.8	10.67	
226Ra	Direct	186.2	3.5	235U (185.72keV; 57.2%)
	214Pb	295.21	18.2	211Bi (351.06keV; 12.91%)
	214Pb	351.92	35.8	
	214Bi	609.32	44.6	
	214Bi	1120.3	14.8	
	214Bi	1764.5	15.4	
228Ra	228Ac	338.32	11.3	
	228Ac	911.21	26.6	
	228Ac	968.97	15.8	
228Th	224Ra	240.99	4.0	214Pb (241.98; 7.12%)
	212Pb	238.63	43.3	228Ac (583.41 keV; 0.114%)
	212Pb	300.09	3.3	
	208Tl	277.36	2.3	
	208Tl	583.17	30.5	
	208Tl	860.56	4.5	
227Ac	227Th	235.97	12.1, 11.2	
	227Th	256.5	7.0	
	223Ra	269.5	13.7	
235U	Direct	143.76	10.96	226Ra (186.1; 3.51%)
		163.33	5.08	228Ac (204.10; 0.171%)
		185.72	57.2	
		205.31	5.01	
238U	234Th	63.28	4.3	232Th (63.81; 0.267%)
	234Th	92.37	2.5	Weak line
	234Th	92.79	2.4	
	234MPa	766.37	0.21	Weak line
	234MPa	1001.03	0.84	
210Pb	Direct	46.54	4.2	Weak line

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