DETERMINATION OF NUCLEAR MATERIALS' ELEMENTAL COMPISTION FOR THE NEEDS OF NUCLEAR FORENSICS

D.O. Dolmatov

Scientific Supervisor: Ph.D. Demyanyuk D.G.

Tomsk Polytechnic University, Russia, Tomsk, Lenin str., 30, 634050

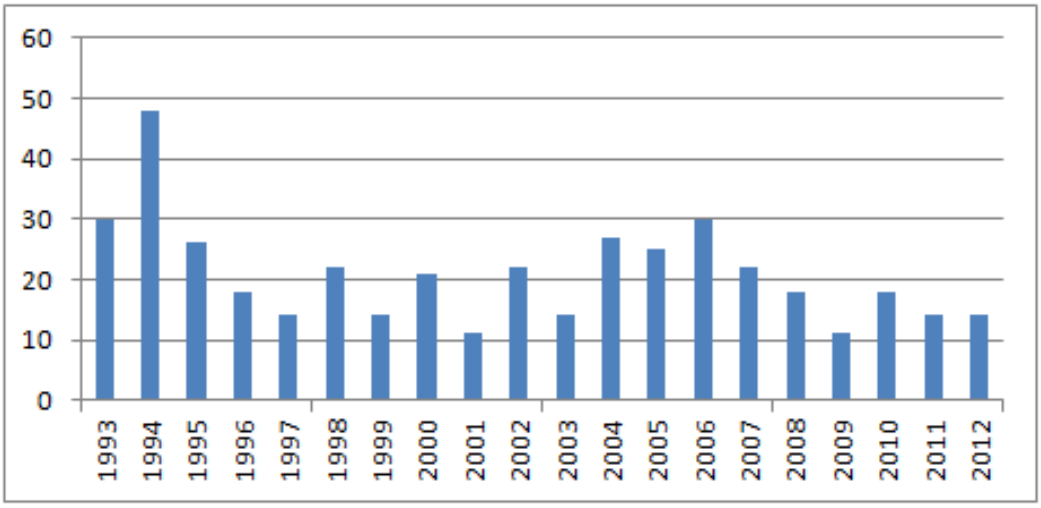
E-mail: dolmatovdo@tpu.ru

Annotation

According to International Atomic Energy Agency there were 419 incidents of illicit trafficking of nuclear and other radioactive materials in 1993-2012. The problem of illicit trafficking of nuclear materials concerns international community due to the increasing of risks which are connected with proliferation of nuclear weapons and nuclear terrorism. IAEA adopted a plan of activities to fight against illicit trafficking of nuclear and other radioactive materials. Nuclear forensics is included to thisplan. Nuclear forensics is analysis of illicit nuclear materials to determine their origin, place of production and routes of transportation. One of the parts of this investigation is elemental analysis. Elemental analysis is important due to the fact that presence or absence of some elements in nuclear material may indicate about origin or some features of nuclear materials. In this article the most useful methods for elemental analysis are reviewed.

Introduction

The problem of illicit trafficking of nuclear and radioactive materials became important in the early 1990's. According to the International Atomic Energy Agency 419 incidents of illicit trafficking of nuclear and other radioactive material were registered. Sixteen of these incidents were connected with high enriched uranium and plutonium.



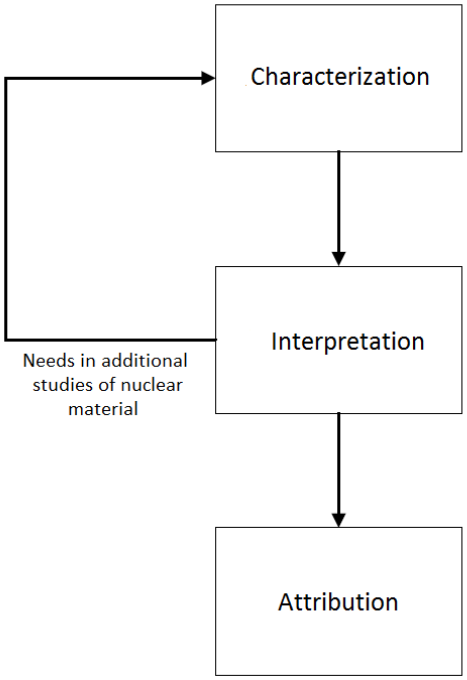
Picture 1. Illicit trafficking of nuclear and other radioactive material in 1993-2012

After the acts of terrorism in USA in 2001 international community started to pay closer attention to the problem of illicit trafficking of nuclear and other radioactive materials. It was connected with the risk of nuclear

and radioactive terrorism. In 2002 IAEA adopted plan of activities to resist the possibility of nuclear materials' possession by terrorist organization. Nuclear forensics is a part of this plan. The main purpose of this activity is to determine origin of nuclear material, feature during its production and routes of its transportation. For purposes of nuclear forensics mainly destructive methods of analyses are used due to the need of precise measurements. In this case right set of methods of analysis is a vital factor for the specialists to achieve the goals of nuclear forensics.

Stages of nuclear forensics

The process of nuclear forensics can be divided into three stages. On the first stage elemental and isotopic compositions of nuclear material are determined. Also particle analyses and phase analysis of nuclear material are important parts on this stage which is called characterization. On the next stage information which was received during characterization is matched with known information about features of nuclear materials' origin and production. In other words, specialists interpret results of characterization. On the last stage experts come to a conclusion about origin, features of production and route of transportation of nuclear material. This stage is called attribution .Nuclear forensics is an iterative process. During interpretation a set of hypotheses are built. These hypotheses are verified by additional study of nuclear material which allows to exclude wrong hypotheses.



Picture 2. Stages of nuclear forensics

Methods of determination nuclear materials' elemental composition

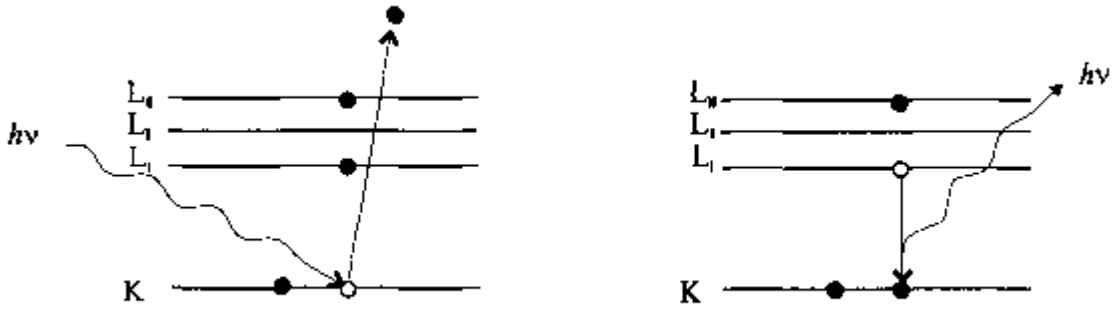
Determination of nuclear materials' elemental composition is important part of characterization due to the fact that presence or absence of some elements in sample may be connected with origin of nuclear material or some features of its production. For example, rare-earth elements can remain in nuclear material during its

reprocessing. Amount of these elements in nuclear material can be measured and this information can help specialists to determine the origin of nuclear material.

For determination of nuclear material's elemental structure spectral methods and mass-spectrometry are used. The main advantage of these methods is possibility to conduct multielement analysis with high precision and sensitivity. Nowadays X-ray fluorescence analysis (XRF), atomic emission spectroscopy with inductively coupled plasma (ICP-AES) and mass spectrometry with inductively coupled plasma (ICP-MS) are the most widely used methods of elemental analysis.

Description of methods

X-ray fluorescence analysis is based on the phenomenon of X-ray fluorescence which is emitted after interaction between Roentgen rays and atom. X-ray knocks out electron from the inner orbital. After that electrons from higher orbital move to the lower orbital and fill the vacancy. As the result characteristic X-ray is emitted. Frequency of secondary X-ray emission depends on atomic number of atom. This relationship was discovered by Henry Mosley in 1912.



Picture 3. Phenomenon of X-ray florescence

Mass-spectrometry is a method of sample analysis which is based on determination of abundance between charge and mass of ions which are produced by ionization. For this purpose laws of ions' movement in electrical or magnetic field areused.

Atomic emission spectroscopy based on analysis of emission spectrums of excited atoms which are unique for every element. This uniqueness is used for qualitative analysis. During quantitative analysis intensive of elements' lines are analyzed.

For the purposes of mass-spectrometry and atom emission spectroscopy analyzed sample should be atomized. In other words it means destruction of material's molecular structure. For the purposes of mass spectrometry atoms must be ionized whereas for purposes of atomic emission spectroscopy atoms must become excited. Inductively coupled plasma can provide all conditions which were described above. Inductively coupled plasma is a type of plasma which is generated by the fluent magnetic field which is created by induction coil. The main advantage of this type of sample atomization, ionization and excitation is high concentration of sample's atoms. Also temperature inside ICP burner reaches the amount of more than 6000 degrees. This fact means the full atomization of analyzed sample. The main disadvantage of using ICP is its dependence on argon. Argon is used to create ICP, to transport sample through the tube to the torch, and to cool the burner.

Comparison of methods

Methods which can be used for purposes of nuclear forensics must meet some requirements. The first requirement is possibility to conduct multielement analysis. It is necessary due to the fact that large set of elements must be determined during analysis for the needs of nuclear forensics. The second requirement is time of analysis. This factor significantly affect on the time of nuclear forensics analysis at all. Third requirement is high sensitivity of methods. In some cases very little amount of elements is important factor which allow building hypothesis about origin and features of production of nuclear material. XRF, ICP-MC, ICP-AES spectrometers meet these demands. So, they can be used for determination of materials' elemental composition for the needs of nuclear forensics.

One of the biggest advantages of the XRF analysis is the possibility to conduct it nondestructively. In addition to this merit there are portable models of XRF spectrometers which allow making on-site nondestructive inspection of material. Generally XRF is used for solid materials analysis. Materials in powder are pressed into the tablets with the cellulose. Metallic materials can be analyzed without any preliminary actions. The main disadvantage of XRF analysis is significant influence of matrix effects to the result of analysis. Matrix effects include x-ray fluoresence's absorption in sample, secondary Roentgen fluorescence and Auger electron's emission spectrums. Matrix effects affects strongly on quantitative analysis's precision. Typical detection limit for elements is 10 particles per million. Method allows analyzing elements from sodium to uranium.

The main advantage of mass spectrometry with inductively coupled plasma is its precision. Typical detection limit for elements in this method vary from nanograms to pictograms per liter of sample. Another advantage is that mass spectrometers can be used for determination isotopic and elemental structure of material. It means that this method is rather versatile .Only liquid samples can be analyzed by using this method. In this case solid samples must be dissolved firstly for the analysis. Mass spectrometers can't be made portable due to the size of spectrometers and their dependence on large amount of argon. Method allows analyzing elements from lithium to plutonium.

Atomic emission spectroscopy with inductively coupled plasma is a rather precision method. This method's typical detection limit varies from nanograms to micrograms. This method is less sensitive than the ICP-MS. ICP-AES can't be used for determination of isotopic composition. Only liquid materials can be analyzed. Also ICP-AES spectrometers cannot be made portable due to the reasons which were described for ICP-MS. Method allows analyzing elements from lithium to uranium.

Conclusion

Methods which were described above have their own merits and demerits and all of them are suitable for the needs of nuclear forensics. However there are some features of these methods' application in elemental analysis

The main advantage of XRF analysis is its possibility of analyzing solid materials. Also XRF spectrometers can be made portable. These facts make XRF portable spectrometers very are available. But low sensitivity with big affection of matrix effects on this method makes it useless for precise analysis of material's

element composition.

For the precocious analysis ICP-MS and ICP-AES can be used. In this case mass spectrometry is more useful method. It has higher sensitivity. Another advantage of ICP-MC is its possibility to determine isotopic and elemental composition of material. Advantage of using ICP-AES spectrometers are connected with their lower salary than ICP-MS spectrometers.

REFERENCES

1. Nuclear Forensic Support. (2006) International Atomic Energy Agency
2. Kenton J. Moody, Ian D. Hutcheon, Patrick M. Grant. Nuclear Forensics Analysis. Taylor & Francis group, 2005
3. M. Otto. (2003) Modern methods in analytical chemistry
4. Application of Nuclear Forensics in Combating Illicit Trafficking of Nuclear and Other Radioactive Material. (2014) International Atomic Energy Agency