APPLICATION OF THE NUCLEAR REACTION ANALYSIS FOR AGING INVESTIGATIONS

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We used the nuclear reaction analysis to investigate quantitatively the content and the depth concentration profiles of the light elements (carbon, nitrogen, oxygen, fluorine, etc) in the surface region of the gold-plated tungsten wires with the thickness from 25 to 50 microns. The wires are used in the straw drift-tubes. These investigations permit to study the aging phenomenon of the gas filed detectors under extremely high-accumulated dose of the radiation (up to 10 C/cm).

Exact and comprehensive knowledge of the composition of solids and films is necessary for obtaining reliable and significant results in many branches of science and engineering. The study of physical and chemical phenomena taking place in the thin films and near the surface of solids requires a quantitative determination and localization in depth of small amounts of the elements. The knowledge of the quantitative depth concentration provides information on the diffusion phenomena, the radiation damage of an irradiated material, the reemission of foreign atoms (carbon, nitrogen, oxygen, etc) from environment [1, 2]. Such studies are fundamental in solid state physics, in technology of new materials, in metallurgy, in electrochemistry, in medicine, in production of new devices, and etc.

To investigate the element composition and the depth concentration profiles of atoms in thin films and the near surface region of solids without its destruction the Nuclear Reaction Analysis (NRA) and the Rutherford Backscattering Spectrometry (RBS) are used.

The RBS is a good technique for obtaining quantitative elemental analysis in the near-surface region of material [5]. However, analysis of low Z elements (carbon, oxygen, etc), which have implanted into a matrix of medium and high Z elements, is limited by this technique because the scattering cross section is proportional of Z^2. That is why the oxygen will be detected only if that concentration is at list five times more of gold concentration**.** ***Therefore the RBS technique can be used for analysis of the element starting from calcium (Z ≥ 20).***

For the detection and quantitative evaluation of the carbon, nitrogen and oxygen content as a function of depth in the gold, the following nuclear reactions with deuterons have been applied

CARBON $^{12}C + d \rightarrow ^{13}C + p + Q_R$

OXYGEN $^{16}O + d \rightarrow ^{17}O + p + Q_R$

NITROGEN $^{14}N + d \rightarrow ^{15}N + p + Q_R$

$^{14}N + d \rightarrow ^{12}C + \alpha + Q_R,$

where $\mathbf{Q_R}$ – an energy emitted in the reaction.

These reactions have not a resonance structure in the energy range of 0.7–1.1 MeV (Fig. 1).

If we need to measure the fluorine concentration the resonance nuclear reaction with protons has to be used (Fig. 2):

FLUORINE $^{19}F + p \rightarrow ^{16}O + \gamma + \alpha + Q_R.$

The characteristics of these reactions at a usually used energy of deuterons and protons are presented in Table 1.

Technique of non-resonance nuclear reactions.

The number of protons counts in the experimental spectrum is proportional the concentration of element in the sample and the cross section of reaction

$$Y = C(t) \cdot I \cdot \sigma(E_d) \cdot \Delta\Omega \cdot t \,,$$

where: $C(t)$ - the concentration of investigated element, i.e. the number of element atoms per cm^3
t – the thickness of sample ;
I - the number of incident particles;
$\sigma(E_d(t))$ - the differential cross section for the angle θ;
$\Delta\Omega$ - the solid angle of a particle detection.

The concentration profile can be computed if the excitation curve $\sigma(E_d)$ has already being measured, or by comparing with a sample containing a well-known element composition

The depth concentration profile

The energy of the a detected proton resulting from a reactions *(d,p)* at a depth *t* depends on the relationships between the beam particle energy and the path traveled by the emitted particles.

The reactions which takes place near the surface of the specimen will yield protons of higher energy, while a reaction which occurs at a greater depth will give protons of smaller energy.

THE ***ENERGY-DEPTH*** RELATION.

1. The energy of deuterons $E_d(t)$ with incident energy $E_d(0)$ at the depth t_1 is given by the equation

$$E_d(t) = E_d(0) - \int_0^{t_1} \left| S_d(t) \right| dt$$

where: $S_d(t)$ is a stopping power of substance for deuterons;
t_1 is a path length of deuterons in the target.

2. The energy $E_p(E_d)$ of proton resulting from a reaction (d,p) is given by the well-known kinematics expression

$$E_p(E_d(t) = M_d \cdot M_p \cdot E_d \cdot \{2\cos^2\theta + B + 2\cos\theta \cdot [\cos^2\theta + B]^{1/2}\} / (M + M_p)^2$$

where: $B = M \cdot (M + M_p) \cdot (Q_R / E_d - M_d / M + 1) / (M_d \cdot M_p)$,
θ - the angle between the beam direction and the detector direction;
M_d, M_p, M – the masses of incident particle, outgoing particle, and final nucleus, respectively.

3. The energy of proton in the detector $E_F(t)$ is given by the equation:

$$E_F(t) = E_p(E_d(t)) - \int_0^{t_2} \left| S_p(t) \right| dt - \int_0^{t_{Al}} \left| S_p(t_{Al}) \right| dt_{Al}$$

where: $S_p(t)$ - stopping power of substance for protons,
$t_2 = t_1 / \cos\theta$ is path length of protons in the target,
t_{Al} - thickness of the aluminum foil-absorber.

*Using tabulated stopping power data [15, 16], the **"DEPTH-ENERRGY"** dependence can be calculated from these equations.*

Mentioned above calculations can be greatly simplified because:

1. The energy loss of particles is small **[**16**]** compared with their initial energy (Table 2)

Table 2. The stopping power of the gold and the aluminum for deuterons and protons.

Substance	E_d [MeV]	$\|S_d\|$ [keV/mg/ cm^2]	E_p [MeV]	$\|S_p\|$ [keV/mg/ cm^2]
Gold	0.9	95	1.6	51
Gold	1.0	90	3.0	38
Al (absorber)	-	-	1.6	130
Al (absorber)	-	-	3.0	80

2. The stopping powers have a linear dependence from the energy

3. The stopping powers change less then 15 % for the energy regions: from 0.7 to 1.0 MeV for deuterons, from 1.0 to 1.6 MeV and from 2.7 to 3.1 MeV for protons [15, 16].

In this case the energy scale can be converted into a linear depth scale:

$$E_F(t) = E_F(E_0) - G_{NR} \cdot t,$$

where: $$G_{NR} = (\partial E_p/\partial E_d) \cdot S_d + S_p/\cos\theta$$

is an effective energy loss related to single length.

The thickness of layer Δt_i (Fig. 4a) on which is deleted the investigated sample connects with energy interval ΔE_i (Fig. 4b) by equation

$$\Delta t_i = \Delta E_i / G_{NR}$$

1.The accuracy of the element concentration measurements is determined by the following parameters:

1. The accuracy of measurements of beam intensity – up to 3 %
2. The of the cross section up to 3 %
3. The geometry parameters ***(ΔΩand θ), - up to 1%.***

So, in totally, an accuracy of the element concentration measurements is better than 5%.

2. The thickness of investigated layer depends on the range of particles in sample (for protons and deuterons) and in the absorber (for protons and α-particles).

The thickness of investigated layer is equal to 5-6 μm

3. An effective energy losses of particles for the gold in the 0.5-3.0 MeV energy region are known with precision up to 3 % [16].

4. The NRA technique allows to evaluate the light element content (oxygen) in the gold coating of the tungsten wire with the depth resolution better than 0.15 μm up to the depth of (1÷1.2) μm.

4. The depth resolution

The depth resolution δt of the method is determined by the energy spread effects appropriated to the real experimental set-up $\delta E(t)$ and could be calculated from (12) as:

$$\delta t = \delta E(t) / G.$$

where:

$$(\delta E(t))^2 = (\delta E_{spec})^2 + (\delta E_{beam})^2 + (\delta E_{geom})^2 + (\delta E(t)_{str})^2$$

δE_{beam} – energy resolution of the beam; $\delta E_{beam} = 0.1$ **keV**

δE_{geom} – energy spread distribution due to the geometry of experimental set-up (the dimensions of detector, the distance between detector and sample (target) and the kinematics of nuclear reaction). δE_{geom} **=16 keV.**

$\delta E_{str}(t)$ - straggling distribution of particle energy in the sample. For a near surface region of the gold sample with thickness 2 mg/cm^2 (about 1 μm depth) the $\delta E_{str}(t)$ doesn't exceed 30 keV **[**17**]**.

δE_{spec} - the resolution function of the spectrometer/ The resolution function of spectrometer δE_{spec} includes both the detector resolution that is 25 keV for E_α = 5.5 MeV and the straggling distribution of energy*)* in the aluminum absorber $\delta E_{str}(Al)$. For a near surface region of the gold sample with thickness 2 mg/cm^2 (about 1 μm depth) the $\delta E_{str}(t)$ doesn't exceed 30 keV.

The contribution of $\delta E_{str}(Al)$ was so strong that the energy resolution of the spectrometer achieved $\delta E_{spec} \cong 50$ keV for E_α= 5.5 MeV.

Taking into account the G values that are presented in Table 1, it become clear that the NRA technique allows to evaluate the light element content (oxygen) in the gold coating of the tungsten wire with the depth resolution better than 0.15 μm up to the depth of (1÷1.2) μm.

Starting from the depth more than 2.5 mg/cm^2 the straggling contribution begins visible **[**17**]** and should be taken into account in the analysis of obtained data.

Technique of the resonance nuclear reactions.

To investigate the fluorine content and depth profile in sample the resonance nuclear reaction $^{19}F(p,\alpha\gamma)^{16}O$ is used. This reaction has very high resonant cross section at E_{res} = 874 keV and E_{rec} = 935 keV with the widths δE_{res}= 5.0 keV and 8.6 keV, accordingly (Fig. 2) The energies of γ-rays are 6.13, 6.92 and 7.12 MeV.

The depth profiles are obtained by total yield measurements of γ-rays or α-particles step-by-step, with incident energies equal to and slightly higher then the resonance energy, i.e. by "shifting" the resonance through the sample. The energy resonance E_{res} and protons in the beam E_b, a depth t_{res} where this resonance occurs are given by the equation:

$$E_{res} = E_b - \int_0^{t_{res}} \left(\frac{dE}{dt} \right)_p dt \qquad (17)$$

where $(dE/dt)_p$ – energy losses of protons in substances.

Then the depth t_{res} on where the resonance occurs can be calculated from a equation:

$$t_{res} = (E_b - E_{res})/(dE/dt)_p$$

Since the energy dispersion of the beam particles in the electrostatic accelerator is much more smaller then the resonance width (ΔE_{beam}=100 *eV* at E_b=1 *MeV*), the depth resolution can be calculated as

$$\delta t = \Delta E_{res} / (dE/dt)_p$$

The δt is equal about 0.1 mg/cm^2 (Table 1).

Combination of different techniques for aging investigations.

The using of different methods for the study of aging of irradiated wires permit to investigate the hear-surface and depth regions of sample.

The characteristics of mentioned methods (the XEM/SEM, the RBS, and the NRA) are presented in Table 4.

1. The SEM analysis yielded information on the morphology of the wire surface with lateral resolution up to one μm. Using these investigation we can say about the destruction and the porosity of the gold coating after the wire irradiation.

2. The XEM analysis yielded qualitative information on the elements containing in the near surface layers of wires with the thickness of 0.1-0.2 μm only. The carbon atoms are not found by this technique because of the strong X-ray absorption by the wire material.

3. The quantitative information about content and depth concentration profiles of middle and heavy elements can be obtained by the RBS analysis.

4. The quantitative information about content of light elements (carbon, oxygen, nitrogen) can be obtained by the NRA analysis with the accuracy better 5 % only.

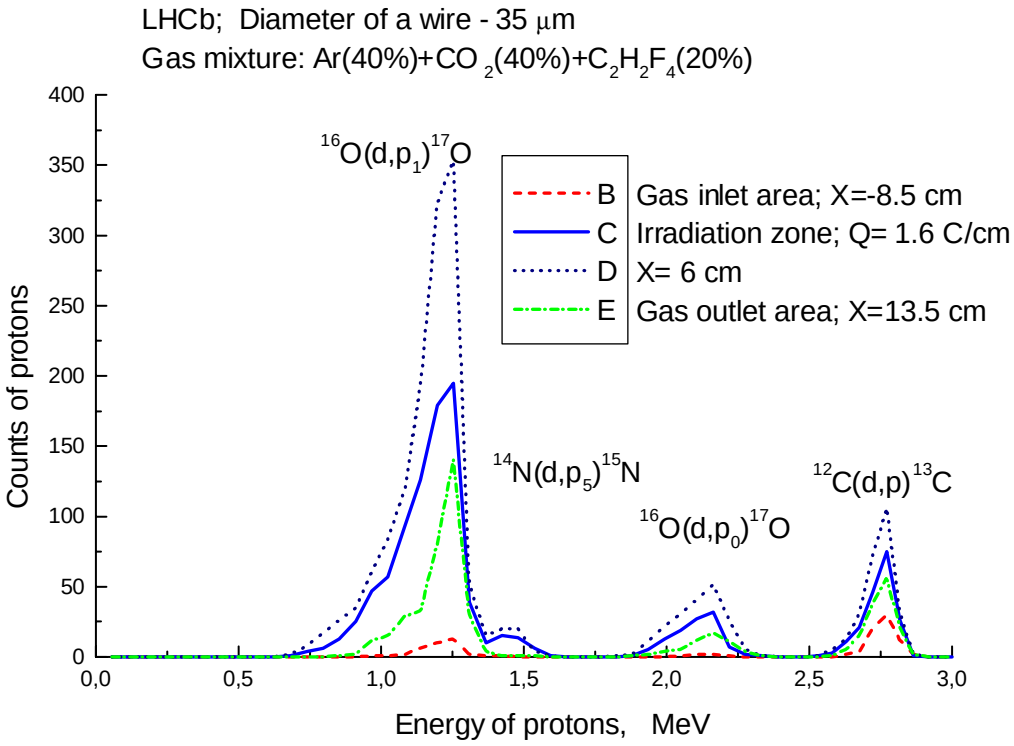
5. Using this method we can measure the concentration depth profile of the light elements with the depth resolution up to 0.1 mg/cm^2.

PROTON SPECTRUM FROM (d,p) REACTION ON THE WIRE TARGET

LHCb -

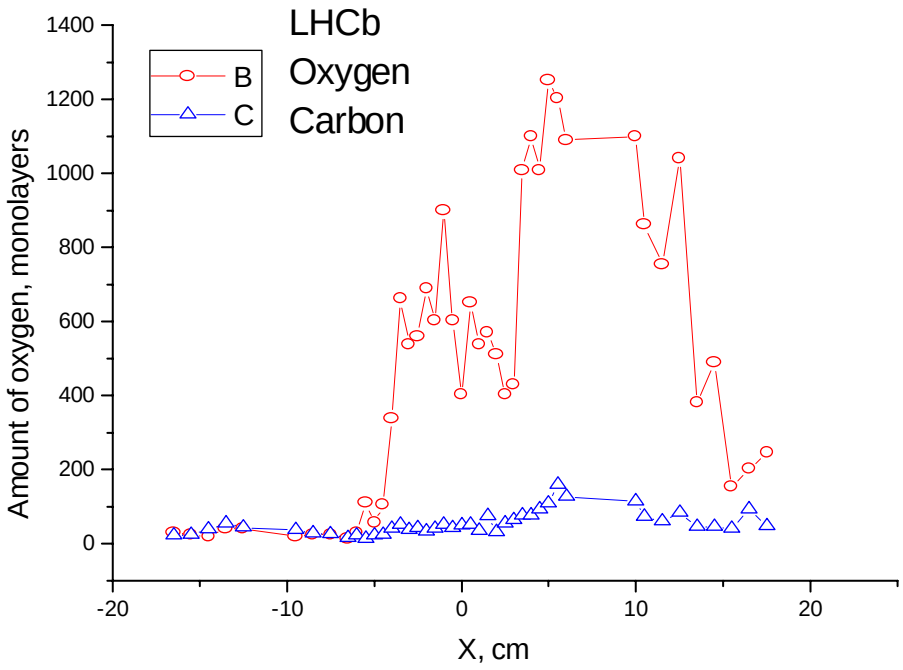
ACCAMULATED DOZE - 1.6 C /cm

DIAMETER OF WIRE - 35 μm



The distribution of oxygen and carbon along the LHCb wire

1 monolayer = 10^{15} at/cm^2



The distribution of oxygen concentration as a function of depth into the gold wire surface

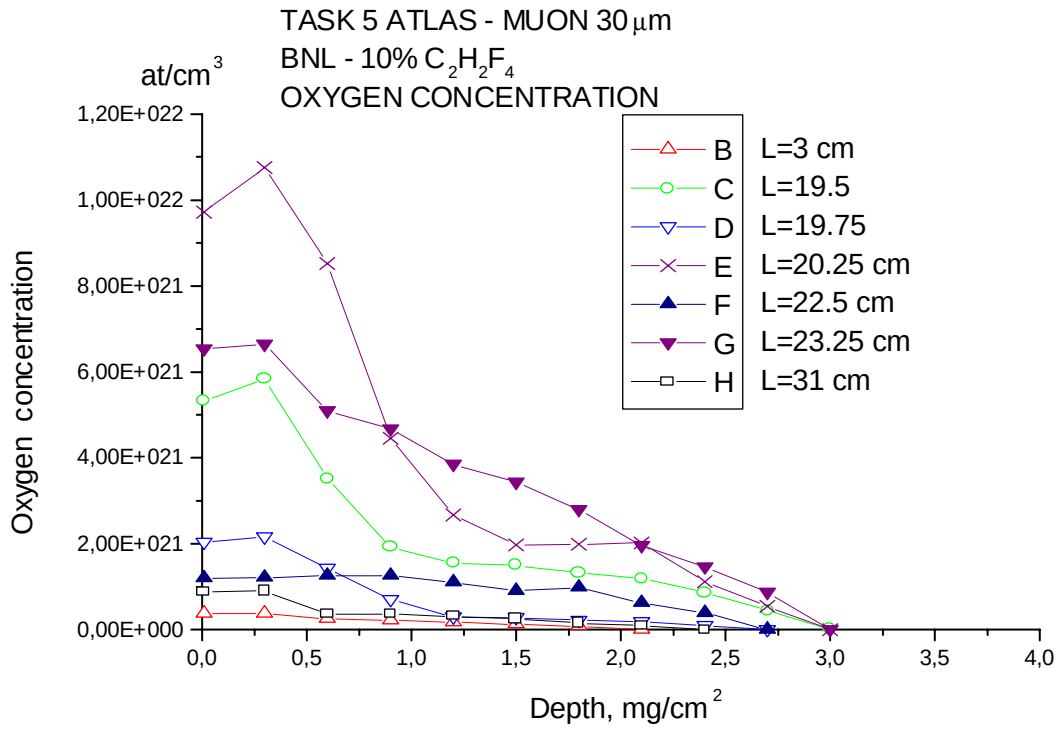


Table 1. Nuclear reactions, which are used to measure carbon, nitrogen, oxygen and fluorine content in the gold.

Measured element	Types of Nuclear Reactions	Emitted Energy Q_R, [MeV]	Beam Energy, E_{beam}, [MeV]	The thickness of the aluminum absorber in front of a detector, t_{Al} [mg/ cm^2]	Energy of emitted particles, $E_p(E_d(t=0))$, [MeV]	Effective energy loss, G_{NR}, [keV/mg/cm^2]	Depth resolution, δt [mg/ cm^2]
C	$^{12}C(d,p)^{13}C$	2.72	1.0	3	3.0	114	0.44
N	$^{14}N(d,p_5)^{15}N$ $^{14}N(d,\alpha_0)^{12}C$ $^{14}N(d,\alpha_1)^{12}C$	1.30 13.6 8.14	1.0 1.0 1.0	3 3 3	1.8 10.0 6.8	170 270 360	0.31 0.19 0.14
O	$^{16}O(d,p_0)^{17}O$ $^{16}O(d,p_1)^{17}O$	1.92 1.05	0.9 0.9	3 3	2.4 1.6	165 180	0.3 0.28
F	$^{19}F(p,\alpha\gamma)^{16}O$	8.114	1.25	6	6.8 (α) and ~7 (γ)	$(dE/dt)_p$=65 keV/mg/cm^2	0.1

a) Yield/ 1μC from a $1\cdot 10^{16}$ at/cm^2 surface layer of element for solid angle $\Delta\Omega$ = 0.1 sr at θ=135^o. Charge 1 μC corresponds to $6.25\cdot 10^{12}$ particles for protons and deuterons.

Table 3. The attenuation of X-ray in the gold.

Element	Energy of X-ray, E_x, [KeV]	Coefficient of attenuation, μ [μm^{-1}]	Attenuation $I(x) / I(0)=e^{-\mu \cdot x}$		
			x = 0.1 μm	x = 0.2 μm	x = 1 μm
Gold	2.2	2	0.82	0.67	0.091
Tungsten	1.8	3	0.74	0.55	0.050
Fluorine	0.68	17	0.18	0.033	$3.7 \cdot 10^{-8}$
Oxygen	0.52	23	0.10	0.01	$1.0 \cdot 10^{-10}$
Nitrogen	0.39	29.5	0.052	0.0027	$1.5 \cdot 10^{-13}$
Carbon	0.28	29	0.055	0.0030	$2.5 \cdot 10^{-13}$

Table 4. Comparison of the SEM/XEM, RBS and NRA methods for aging investigations of the gold coating of tungsten wire.

Technical Characteristics		**Different techniques for thin films investigation**		
		Scanning Electron Microscopy with X-ray Emission Spectroscopy (SEM/XEM)	**Rutherford Backscattering Spectrometry (RBS)**	**Nuclear Reaction Analysis (NRA)**
Elements identified		C – U	Li – U	Li,Be,B,C,N,O,F
Morphology of surface		Yes	None	None
Lateral (space) resolution		< 1 μm	~3 mm	~3 mm
Sensitivity	For the middle and heavy elements (Z> 20)	$\geq 10^{15}$ at/cm^2 (gold)	$\geq 1 \cdot 10^{12}$ at/cm^2 (gold)	The nuclear reactions for these nuclei are forbidden at this energy of ions
	For the light elements (Li, B, C, N, O, F) in the gold matrix	Only element Identification	$\geq 1 \cdot 10^{20}$ at/cm^2 (Oxygen in gold)	$\geq 1 \cdot 10^{16}$ at/cm^2 (Oxygen in gold)
Accuracy of the element concentration measurements		Only element Identification	~ 5 %	~ 5 %
Thickness of investigated layers in the gold matrix		Not more 0.05 μm (at the electron energy of 10 keV)	up to 6 μm	up to 6 μm
Depth resolution in the gold matrix		Equal to thickness of investigated layer	≤ 0.1 μm	up to 0.1 μm
Depth analysis		Destructive	Non-Destructive	Non-Destructive

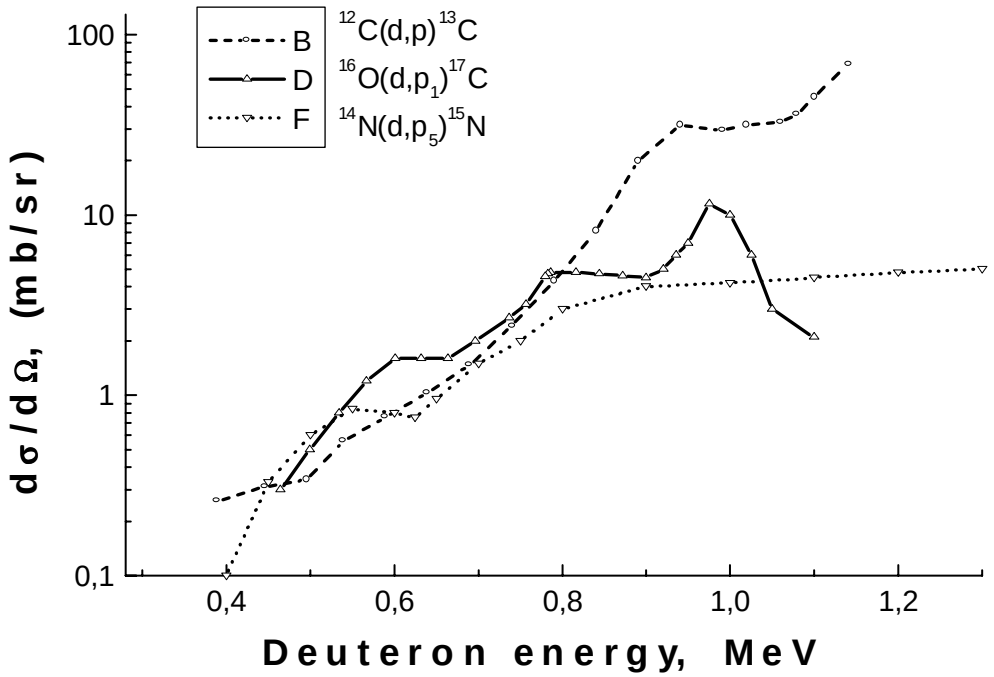


Fig. 1. Cross section of the ^{12}C(d,p)^{13}C (dashed line), ^{14}N(d,p$_5$)^{15}N (dot line) and ^{16}O(d,p$_1$)^{17}O (solid line) nuclear reactions at $\theta_{lab} = 135^{o}$ [7, 8, 11].

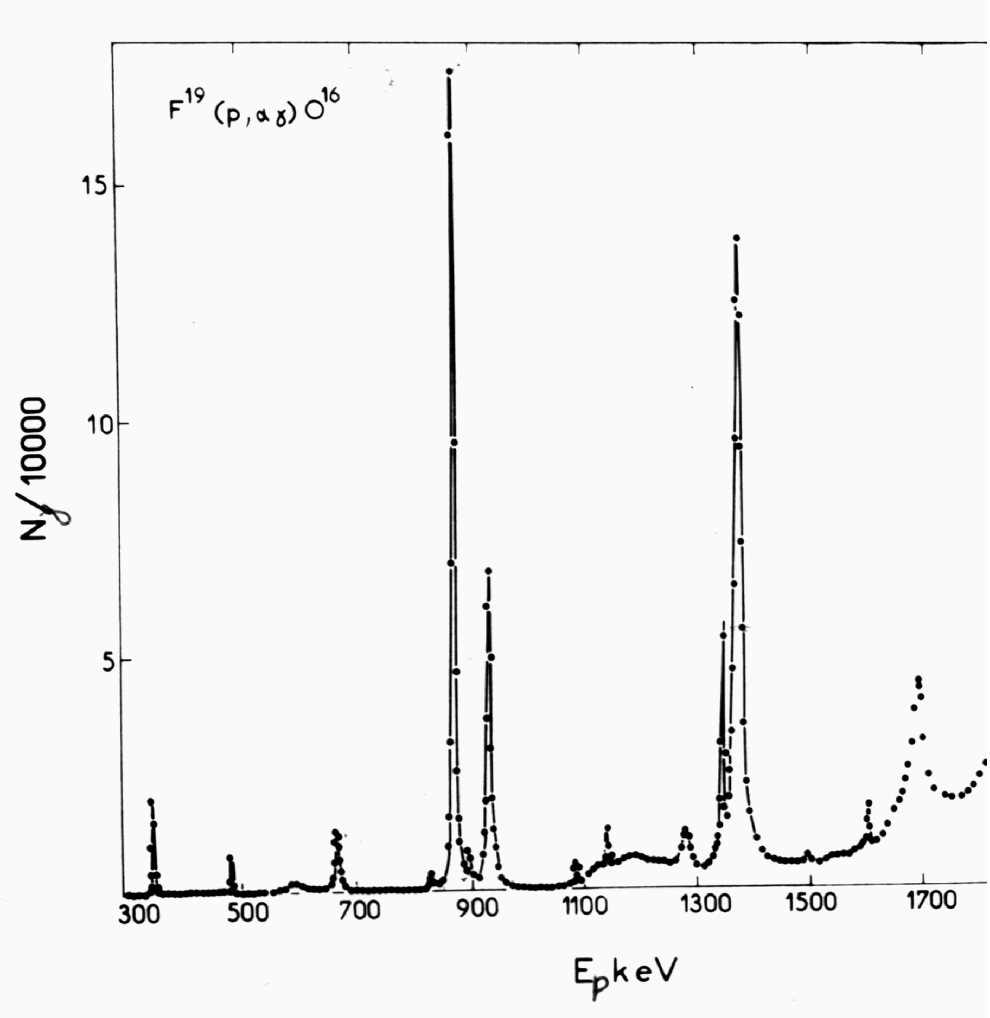


Fig. 2. Overall view of the $^{19}F(p,\alpha\gamma)^{16}O$ excitation curve at $\theta_{lab} = 90^{o}$, normalized to a 3.8 μg/cm^2 fluorine target, 60 μC per point; 3" x 3" NaI(Tl) detector at 7 cm; detected $E_\gamma > 4.7$ MeV [4,12]

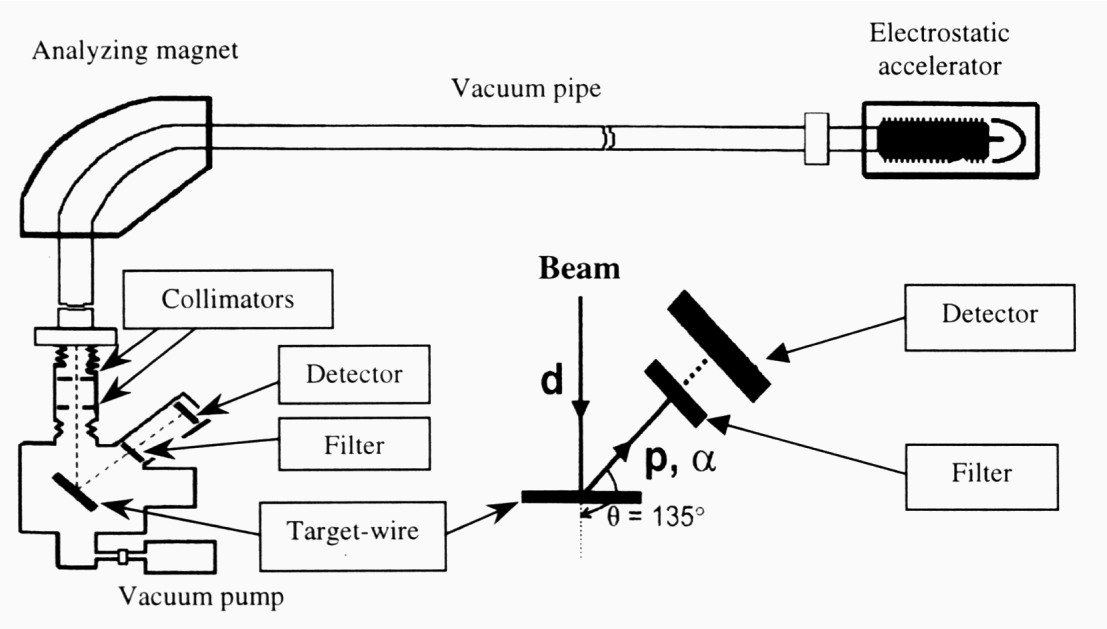


Fig.3. Schematic representation of the experimental set-up in the Nuclear Reaction Analysis.

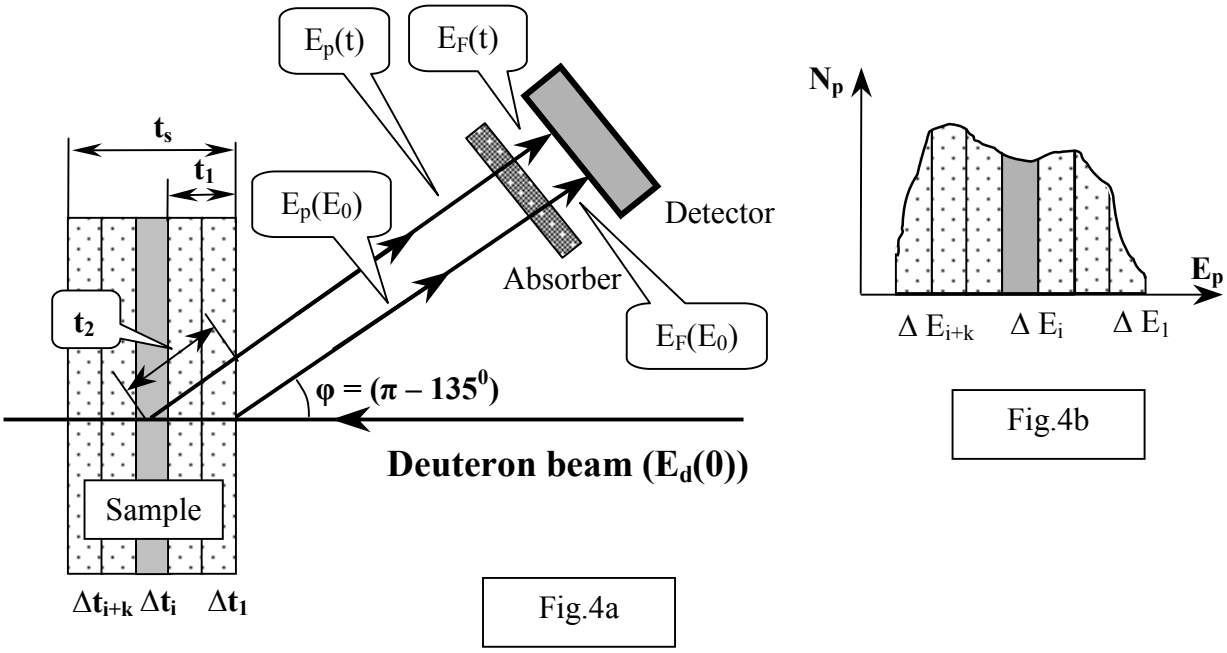


Fig.4. Schematics and nomenclature for the (d,p) nuclear reaction process in sample (Fig.4a) and reconstruction of the resulting energy spectrum (Fig.4b).

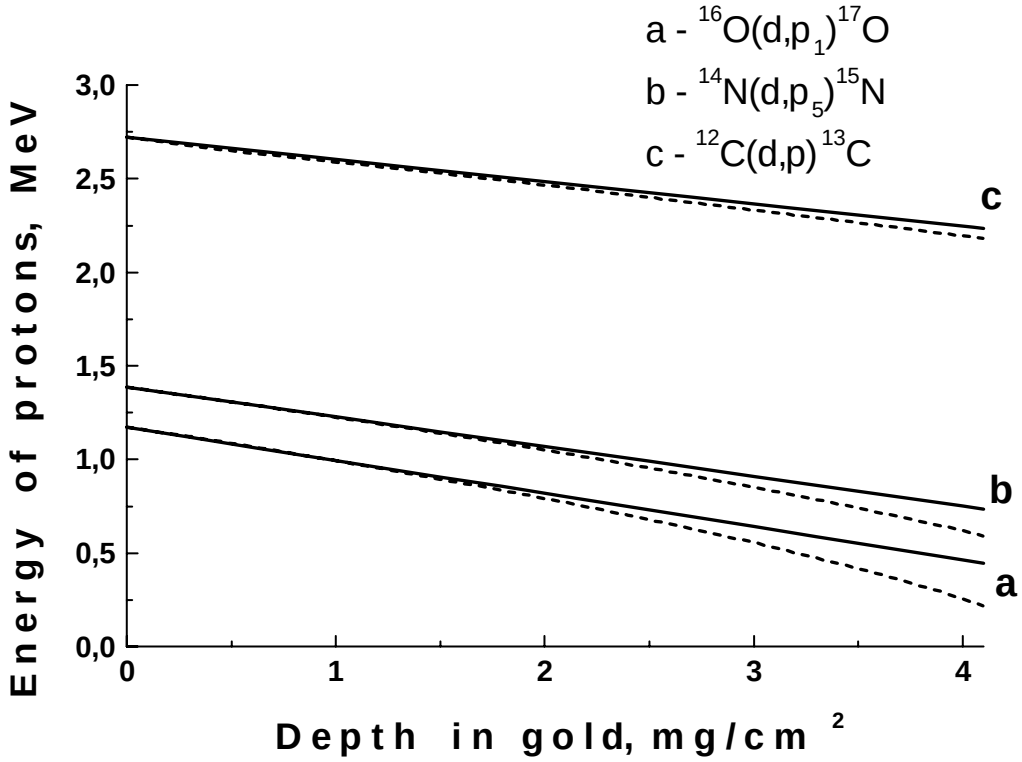


Fig. 5. Variation of the energy of the detected protons $\boldsymbol{E_F}$ with depth $\boldsymbol{t}$, at which the $^{12}C(d,p)^{13}C$, $^{14}N(d,p_5)^{15}N$ and $^{16}O(d,p_1)^{17}O$ reactions took place (E_0=0.9 MeV, θ_{lab} = 135^o), computed for the gold sample with the 5% admixture of the light nucleus(**a** –oxygen, **b** – nitrogen, **c** –carbon). The calculation with the G_{NR} is the continuous line. Exact calculations are dashed lines

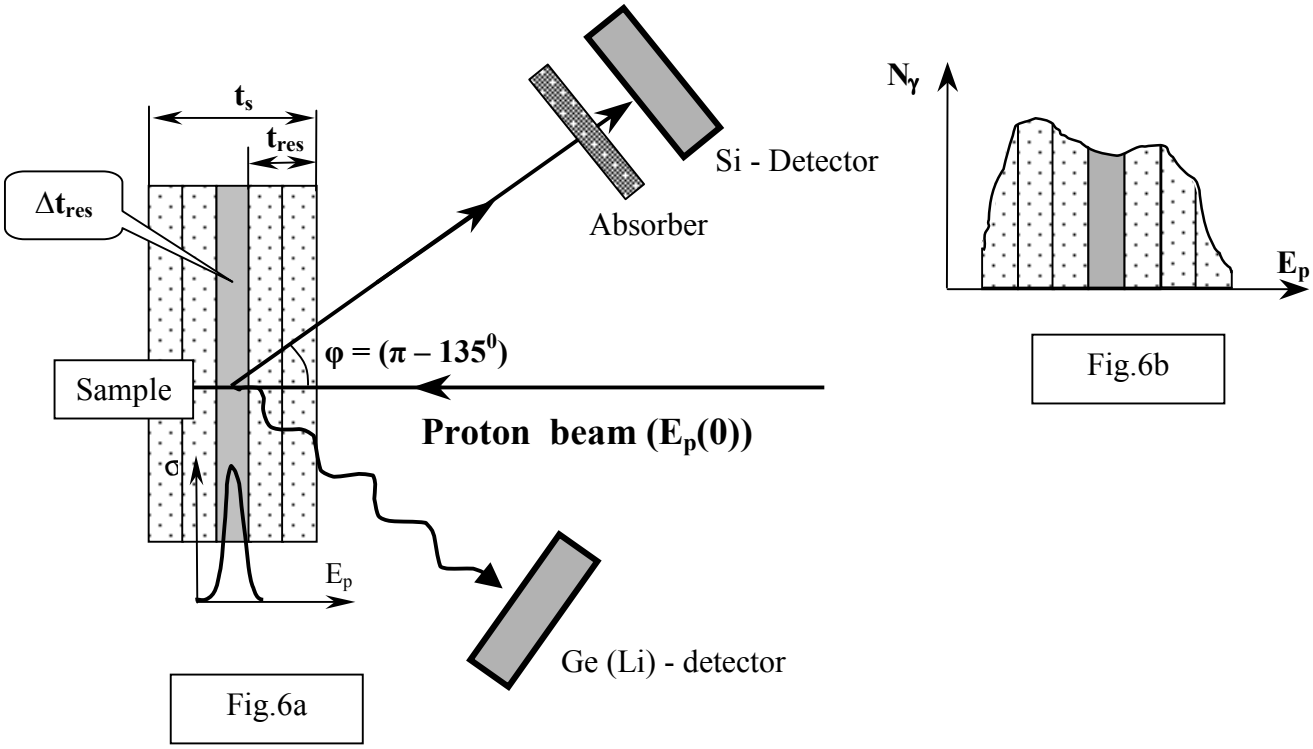


Fig.6. Schematics and nomenclature for resonance (p,αγ) nuclear reactions process in sample (Fig.6a) and reconstruction of the resulting energy spectrum (Fig.6b).

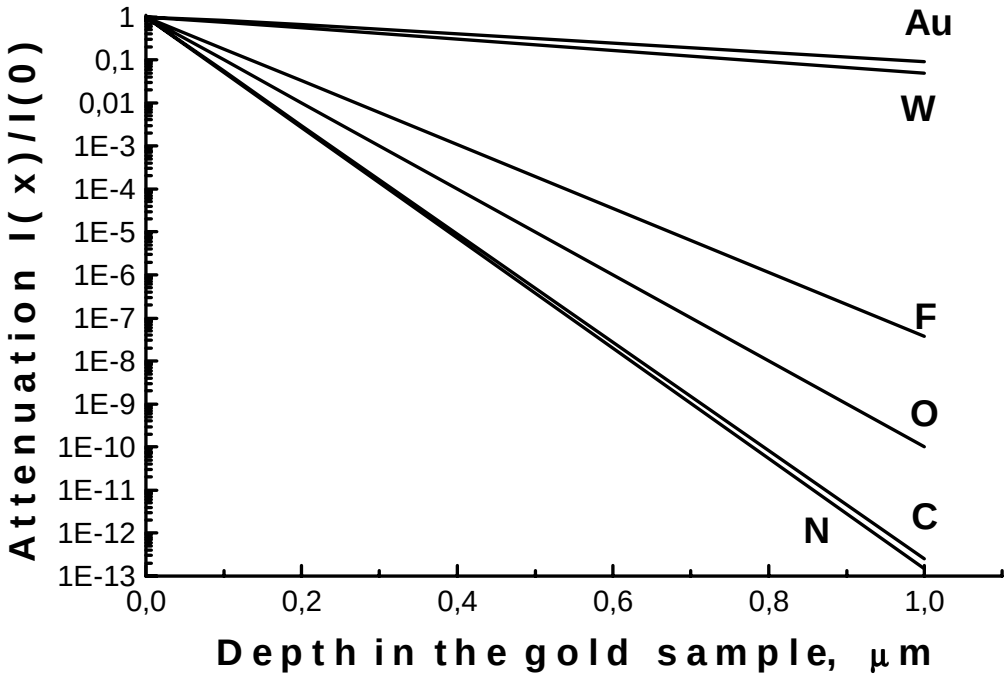


Fig. 7. The attenuation of X-ray of the gold, tungsten, fluorine, oxygen, nitrogen, and carbon atoms in the gold sample.

CONCLUSION

- ✓ **NRA method looks very promising and unique due to high sensitivity, the lack off etching, quantitative evaluation of the light elements and the large depth of analysis. It is very probably that this Nuclear Reaction Method will become a customary technique for aging investigations of the new generation of the gas-filled detectors. Using NRA method in combination with Scanning Electron Microscope and X-ray spectroscopy (SEM/XEM), we can obtain much more precise and complete information about the aging processes in detectors.**

- ✓ **Only NRA method gave us unique opportunity to detect present above unpredictable peak distribution of the oxygen concentration along the wire.**

- ✓ **Application of NRA to the aging investigations allowed us to show both the important role of oxygen in the wire aging and a kinetic of oxygen transportation into the depth of gold coating of the wire.**