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ON THE METHOD OF « IN SITU » MEASUREMENT OF GROUNDWATER CHLORINE CONTENT: AN IMPROVEMENT FOR ITS UTILIZATION IN SMALL DIAMETER WELLS (UNDER 30 CM)

SUMMARY

The observations concerning the dynamic behaviour of the transition fresh-water intruding sea-water are run in observation wells, the diameter of which is lesser than 30 centimetres. This work refers to the results obtained during the various theoretical, planning and experimental laboratory phases of a new probe; though based on traditional principles of nuclear physics, the work uses an americium-fluorine radioactive source, a counter helium 3 and an original geometry source-counter.

This way the influence sphere characterized by neutrons emitted by the source allows measurements for wells having a small diameter.

1. FOREWORD

A previous note [4] proposed a new method for « in situ » measuring chlorine content of groundwater or seawater. This method is based on a chlorine nucleus characteristic which clearly differenziates this element from any other element generally contained into groundwater or seawater: its remarkable capacity of capturing thermal neutrons.

In practice, the measurement is perfomed by putting into water a « neutron probe », formed by a fast neutron source and a thermal neutron counter. Fast neutrons, by colliding with hydrogen atoms of water, lose their own ener-

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gy thus changing into thermal neutrons (initial energy: 5 Mev - final energy: 0,025 eV). The thermal neutron counter records their quantity, which is practically constant in water lacking or poor in chlorine content and depends on the neutron source intensity as well as on the position of this source respect to the counter.

When water contains chlorine, a remarkable lessening of thermal neutron counting takes place, owing to the fact that a portion of such thermal neutrons is captured by the chlorine nucleus thus being unable to react within the counter. A calibration curve correlates the chlorine content to the neutron count carried out everytime.

The probe described consists of a source of Americium-241 - Berillium (300 mCi of Am-241) and of a proportional boron fluoride counter. It can detect a minimum chlorine content equal to 0.13 gram per liter into low chlorine content water and to 0.22 grams per liter into high chlorine content water. These limits, which can however be lowered through a more sensible neutron counter, are widely sufficient to obtain the determination required by the hydrogeological studies aiming at clarifying freshwater/saltwater connections which are especially remarkable in coastal regions having geological characteristics similar to the ones of Apulia, where groundwater is supported by seawater.

On the other hand, the nuclear method of chlorine measurement is of special interest in the determination of quick chlorinity changes in groundwater, like the ones which take place in the diffusion zone separating a full fresh-water from a full salt-water.

In case of high chlorine content, the normal conductometric salinometers actually undergo polarization and electrode fouling phenomena, being more sensitive to chemical composition differences.

Anyway, the above mentioned neutron probe allows measurements only in wells at least 30 cm in diameter. Actually, the neutrons emitted by the Am-Be source have an average energy equal to 5 MeV approximatively and, for being reduced to a 0.025 eV value they must undergo 20 collisions approximatively against hydrogen nuclei.

Even if the probability of collision with these nuclei is rather high, a miminum water volume is required around the source in order to make the emitted neutrons thermal.

In volumes under 30 cm in diameter a fast neutron runaway takes place; part of them comes back, reflected by walls. However the quantity of returning neutrons is not constant because neutron reflection depends on the reflecting material characteristics: chemical composition, soil porosity and water content, etc.

Such inconvenience does not distort measurements at sea or at marine swallows or wells 30 cm or over in diameter, but it distors any measurement in other wells having a smaller diameter; therefore, in order to overcome this disadvantage, a study has been carried out aiming at finding the possibility of using neutrons under 5 MeV in initial energy, thus reducing the water volume needed for moderating them.

2. CHOICE OF NEUTRON SOURCE

An ideal neutron source for this measurement should provide low energy neutrons (few KeV) whose number must be sufficient to minimize the statistical errors due to the radioactive emission casualness.

Furthermore, such source should not emit any gamma radiations and should have a long half-life, a small size and a low cost.

One of the available sources which can be utilized among the ones based on the photodissociation phenomenon (gamma-neutron reaction) is represented by a source consisting of an antimony-124 / berillium mixture.

Antimony-124 provides the gamma rays with the 1,666 MeV energy necessary for extracting neutrons from Be-9 with an approximatively production of 1.3×10^5 n/s for 1 curie of Sb^{124}. Neutrons have a 26 KeV energy value approximatively which is excellent for this end; anyway, the Sb^{124} half-life is only equal to 60.9 days thus requiring continuous neutron counting adjustments owing to the too quick decay.

The source ought to be replaced or regenerated too often; furthermore, the operator protection against such an energetic gamma radiation requires very heavy lead shields, which make in field work difficult, especially in areas which are impracticable by car.

Greater advantages are offered by sources of the (α, n) type, where neutrons are obtained by bombarding with alpha particles low atomic number elements such as: Li, Be, B, C, O, F, Na, Mg and Al.

Alpha particles can be provided by different natural or artificial radioisotopes, such as actinium-227, americium-241, polonium-210, radium-226, thorium-228, etc. Excepting polonium-210 and thorium-228, whose half-life is too short (138 and 1.91 days respectively), the other isotopes offer good characteristics.

Americium-241 seems to be preferable because it emits fewer gamma radiation together with alpha radiations, in comparison with other elements; furthemore, these gamma radiations are less energetical.

At a distance of 1 meter, the gamma dose ratio is 1:8 with actinium-227 and 1:60 with radium-226.

As previously specified, the target choice should meet the requirement of obtaining low energy neutrons in sufficient number.

The energy of neutrons originating by (α, n) reactions depends on the energy of the incident alpha particle, on spent energy or on energy deriving from reaction as well as on the way the resulting nucleus de-energizes. If reference is made to americium-241, the energy of incident alpha particles is 5MeV approximatively. Neverthless, this is the maximum alpha particles energy; in pratice, before producing a nuclear reaction, a large part of alpha particles can undergo collision of the elastic type thus losing a portion of the energy held.

Such being the case, the above nuclear reaction take place with polyenergetic alpha particles and for this reason neutrons emitted are not monoenergetic.

Furthermore, the neutron energy depends on the emission angle with reference to the incident particles direction: maximum energy value corresponds to 180°, the minimum energy value corresponds to 0°.

The reaction energy depends also on nuclear bonds of nuclei affected by the reaction, i.e., the target nuclide and the resulting nuclide.

The energy can be calculated on the ground of the following formula:

$$Q = 931\ (m_1 + m_2 - m_3 - m_4)$$

where: m_1 = target nucleus mass

m_2 = incident particle mass

m_3 = resulting nucleus mass

m_4 = neutron mass

931 = conversion factor of masses into neutron energy.

If the mass difference is positive, the reaction produces energy and this could also take place with alpha particles of 0 kinetic energy (disregarding the Coulomb barrier).

If the mass difference is negative, it is necessary to give energy. So, in this case, a part of kinetic energy of alpha particles is spent for letting reaction take place.

The quantity of energy which can be got by emergent neutrons is lower than the one available when reaction takes place with energy release.

Nevertheless, it is to kept in mind that the energy resulting anyhow from reaction is divided between the neutron and the resulting nucleus.

When the nucleus has low energy excitation levels, a part of energy may be spent for bringing the nucleus to one of such levels, from which the nucleus comes back to its fundamental state emitting gamma radiations.

When, taken in its complex, the resulting energy does not allow this operation, the whole energy can spent as a neutron critical energy. In conclusion, as regard the source choice, the following factors are to be taken into account under the same conditions of energy of incident alpha particles: the reaction Q, the existence of nucleus levels compatible with the energy originated by reaction, the density of the radioisotopes-target mixtures which conditions the energy of incident particles on the basis of the above mentioned collision slowing-down of the elastic type (these factors are to be considered merely for the sake of neutron energy).

As a general rule, a prevision about the energy of neutrons resulting from different possible reactions can be obtained through the following formula:

$$E_n = \frac{Q}{1 + \frac{m_n}{m_r}} + E\,\frac{m_G\ m_n}{(m_r + m_n)^2}\left\{2 + \frac{m_r\ (m_n + m_r)}{m_G + m_n}\left[1 - \frac{m_G}{m_r}\right] \pm \right.$$

$$\left. \pm 2\sqrt{1 + \frac{m_r\ (m_n + m_r)}{m_G\ m_n}\left[\frac{Q}{E} + \left(1 - \frac{m_G}{m_r}\right)\right]}\right\}$$

where:

Q = reaction energy
E = max. energy of incident particle
m_r = mass of the nucleus resulting from reaction
m_n = mass of the neutron
m_G = mass of the incident alpha particle.

The $\pm$ sign preceding the root is referred to the max. emission angle ($- = 180^\circ$ / $+ = 0^\circ$) or to the min. angle between the direction of the incident particle and the one of the originated neutron. The resulting nucleus is considered to be in its fundamental state, wherefore the neutron is given the whole resulting energy.

A further factor that is to be considered when chosing a target regards the number of neutrons emitted, which, in order to attain the above object must be as high as possible. This number depends on the availability of alpha particles having energy enough to make reaction take place as well as on the target nucleus cross section for stated reactions. This energy can be raised by increasing the density of the radioisotope-target nucleus mixture.

The limit to this increase lies in the fact that, as previously described, an excessive thickening causes a remarkable slowing-down of alpha particles, so that a lot of them have no longer the energy needed for getting a reaction, especially when Q is considerably negative.

On the other hand, a considerable increase of alpha particles involves also a considerable increase of gamma radiation, which accompanies alpha particles. In that case, it is necessary to get a compromise between neutron ideal energy and quantity.

The following table 1 shows the light nuclei available for reaction alpha-neutrons - their reaction Q - the indicative number of neutrons which can be produced through a flux of 10^6 alpha particles when react with the under reported nuclei - the maximum and minimum energy of the obtained neutrons [1].

TABLE 1 - List of the target-nuclei, of Q of reaction and of neutrons maximum and minimum energy, available by nuclear reaction shown on the right of the table.

Target	Reaction Q	Neutrons emitted every 10^6 α particl.	NEUTRON ENERGY		Nuclear Reactions
			max E (MeV)	min E (MeV)	
Li^7	−2.790	2.6	1.518	--	$Li_3^7 + He_2^4 = B_5^{10} + n$
Be^9	5.704	80.0	11.051	5.077	$Be_4^9 + He_2^4 = C_6^{12} + n$
B^{10}	1.061	13.0	7.42	2.027	$B_5^{10} + He_2^4 = C_6^{13} + n + \beta^+$
B^{11}	0.158	26.0	5.126	0.305	$B_5^{11} + He_2^4 = N_7^{14} + n$
C^{13}	2.215	10.0	7.42	1.349	$C_6^{13} + He_2^4 = O_8^{16} + n$
O^{18}	−0.700	29.0	4.42	1.192	$O_8^{18} + He_2^4 = Ne_{10}^{21} + n$
F^{19}	−1.949	12.0	3.37	0.23	$F_9^{19} + He_2^4 = Ne_{11}^{23} + n + \beta^+$
Na^{23}	−2.971	1.50	5.32	0.37	$Na_{11}^{23} + He_2^4 = Mg_{12}^{26} + n + \beta^+$

By examining the features of target elements one can notice that – as far as the neutron energy is concerned – Li^7 has the best characteristic; nevertheless, the neutron production appears extremely low, the lowest after the one of Na^{23}.

The second element - with respect to the emitted neutrons energy –, is F^{19}.

The production of neutrons having energy comprised between 3.37 and 0.23 MeV is only lower that the one of Berillium, O^{18}, B^{10} and B^{11}, which on the other hand product neutrons having a higher energy.

Therefore, an Am-241 / F-19 source has been chosen.

In order to get a sufficient quantity of neutrons, it has been necessary to mix 1 Curie of Am-241 with fluorine thus obtaining a 1.5×10^5 n/s emission. This source is 22.4 mm in diameter and 31 mm in length.

3. NEUTRON COUNTER

A proportional helium-3 counter has been utilized, containing a 10 atm compressed gas. Helium-3 capture cross section is 5400 barn for thermal neutrons, so it is especially sensitive to this kind of neutrons. This neutron counter (T.D. Texlium Model) is 15 cm in working length – 2.5 cm in diameter.

Owing to the gas high pressure, the counter is sensitive also to the gamma rays of americium-241 and of Na^{22}, limit of changing from Fluorine to Neon. Therefore, a discriminator has been included in the amplifier circuit of signals by which gamma rays are rejected.

Thermal neutrons react with Helium-3 on the ground of the following reaction:

$$He_2^3 + n = H_1^3 + p$$

4. SOURCE-COUNTER ARRANGEMENT

Fig. 1 shows a layout of the probe built-up.

The probe to be used should have as small as possible diameter; so it has been necessary to disregard the usual arrangement where the source is placed at the level of center line of the counter. The source of this probe is placed directly under the counter. Obviously, this is not the best arrangement as regards sensibility, because, notoriously, the thermal neutron flux is measured with a better efficiency when the source is in the middle part of the counter. Anyway, the thermal neutron capture has a slight per cent increase going

little by little away from the source, thus counterbalancing partly the reduced counting efficiency. The above mentioned arrangement allows the source to keep within its lead shield thus minimizing the risk due to gamma radiations of Am-241 during the Am-241/Fluorine-19 reaction and other gamma-rays produced.

5. PROBE CALIBRATION

The probe calibration has been preceded by a calculation of the thermal neutron flux considering different source-neutron counter distances, carried out both in fresh-water and in salt-water.

A formula of the following type has been used:

$$\phi = \frac{Q\ e^{-\sqrt{\tau}}}{4\pi\ r\ \Sigma_i\ \tau_i}$$

where:

ϕ = thermal neutron flux ($n/cm^2 s$)
Q = neutron source intensity (n/s)
$\sqrt{\tau}$ = neutron slowing-down length (cm)
r = distance from source (cm)
Σ_i = summation of microscopic absorption cross section (cm^{-1}).

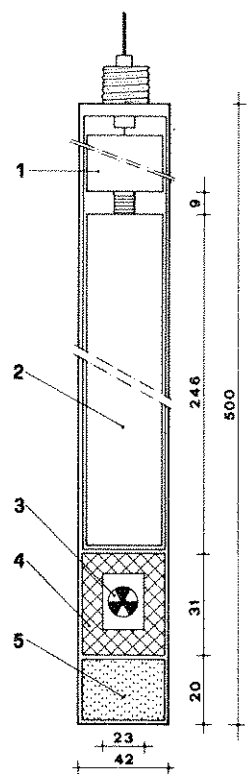

Fig. 1 – Scheme of the neutron probe for measuring the chlorine content of the ground water: 1) preamplifier; 2) counter; 3) neutron source; 4) lead shield; 5) rubber plug.

Table 2 shows results obtained.

TABLE 2 - Thermal neutron fluxs at different source-counter distances in fresh-water and in salt-water respectively. This calculation has taken into account values of utilized parameters relevant to neutrons having a similar energy as the fission ones.

Source-Counter distance cm	A Fresh-water flux $n/cm^2.s$	B Salt-water flux $n/cm^2.s$	Capture ratio % difference A-B/A
5	$6.28.10^{-3}$	$4.47.10^{-3}$	28.82
10	$2.64.10^{-3}$	$1.74.10^{-3}$	33.86
15	$1.12.10^{-3}$	$0.72.10^{-3}$	36.02

Calibration has been carried out in order to attain two objects:

a) verification of the thermal neutron counting constancy in the presence of a reduction of the water mass surrounding the neutron probe. This for stating the min. diameter of wells where accurate chlorinity measurements can be performed by using neutrons having a max. 3.37 MeV energy, i.e., the ones originated through the Am-F reaction

b) plotting of a calibration curve by correlating the thermal neutron counting to increasing chlorine quantities thus having the possibility of deducing immediately in the field the water chlorinity of the ground of the neutron count.

Table 3 shows that the count constancy has been obtained when steel pipes 20.0 and 15.0 cm in diameter have been used, in comparison with the count got by dipping the probe in a container 60 cm in diameter (infinite medium). The count has dropped to a 83.1% of the one obtained in the infinite medium.

The proper probe calibration has been performed by using increasing amounts of sodium chloride - 99% in titre - 58.45 in molecular weight. Precedently, it had been ascertained that the use of this salt does not determine substantial changes in comparison with seawater. The sodium chloride has been dissolved in drinking water (Caposele water - for its chemical composition see [1]) according to different concentrations determined through both the Mohr method and the stoichiometric calculation; a 10 min count has been everytime carried out through the neutron probe.

TABLE 3 – Count of thermal neutrons carried out in water volumes decreasing little by little (60 cm - 20 cm - 15 cm - in diameter).

Diameter cm	count cpm	% difference
60	89,750	–
20	89,750	–
15	89,750	–
10	74,580	–16.9

Table 4 shows the results and Fig. 2 shows the relevant diagram.

The min. measurable chlorine activity, considered as 3 times higher than standard deviation for a 10 min count, is 0.15 g/1 of Cl and 0.26 g/1 at high chlorine concentrations.

On the basis of the before recalled nuclear physics principle (thermal neutron capture), by the same probe it is possible to determine the filtration speed of the groundwater. This measurement is performed putting into a well tracers as boron or cadmium (very active for capturing neutrons) and following from time to time their concentration dilution revealed by increasing of the neutron counting [3,5].

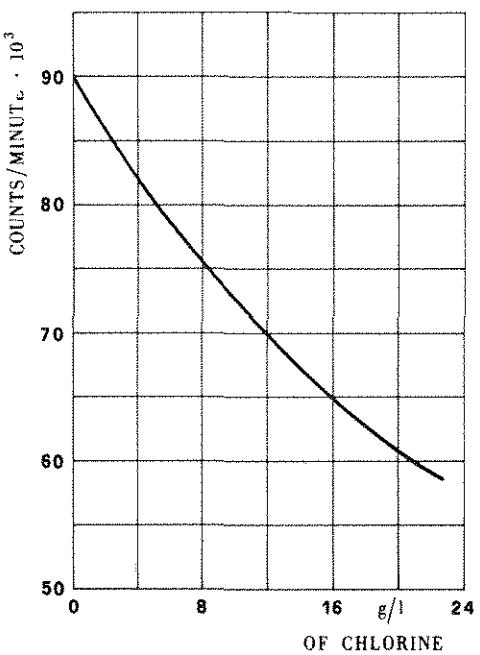

Fig. 2 – Calibration curve of the neutron probe for measuring the clorine content of the ground water.

TABLE 4 – Correlation between water chlorine contents and thermal neutron count.

Concentration NaCl (g/l)	Concentration Cl (g/l)	Count cpm	Difference in comparison with drinking water %
0.026	0.0160	89,750	–
1.026	0.6269	88,521	1.37
2.026	1.2290	87,527	2.70
4.026	2.4430	85,092	5.19
6.026	3.6570	82,403	7.63
8.026	4.8710	81,265	9.86
10.026	6.0850	78,980	12.00
15.026	9.1201	74,260	17.26
20.026	12.1510	69,934	22.08
30.026	18.2160	62,431	30.44
40.025	24.2860	57,423	36.02

The same neutron probe can be employed for water content measurement in not saturated soils, on the basis of the much greater capacity of hydrogen to slow-down neutrons respect to any element constituting the soil [2].

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