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A CONTRIBUTION TO THE STUDY OF SEA-WATER
INTRUSION INTO COASTAL AQUIFERS:
A NUCLEAR METHOD FOR MEASURING GROUNDWATER
CHLORINE CONTENT

SUMMARY

The direct measurement of underground water chlorine content gives a contribution to the solution of the problems brought about by groundwater contamination by the sea.

On the basis of such considerations a method of chlorine measurement has been accomplished; it is based on the characteristic of the nucleus of this element having a capture section for thermal neutron far greater (on the average about 200 times) than any other element which normally occurs in underground waters.

The variation of thermal neutrons, followed by a BF_3 proportional counter immersed into water (moderating process) with a source of fast neutrons (neutrons probe), gives the chlorine content immediately, since this element is 98,5% responsible for the above mentioned neutron flux variation.

1. INTRODUCTION

The groundwater supply in a region such as Apulia - that is practically lacking in surface water resources - is of vital importance and becomes an element which conditions the economic-social development of the zone.

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The main groundwater source in this area is the imposing aquifer present in the Mesozoic calcareous and dolomitic substrate; this aquifer is called «deep» or «basical» for being told from those having a much lower capacity contained in the superior Miocene or Quaternary soils. The deep groundwater is supported at its base by the sea-water which invades the continent; the fresh-water / sea-water contact determines molecular diffusion and dispersion phenomena in consequence of which the fresh water is contaminated by the sea water thus originating a more or less extensive transition zone of water whose salinity increased according to the depth.

The harmful sea water influence is made worse during fresh-water drawings, particularly when drawings are not counterbalanced by an adequate recharge: the salt-water is often drawn up thus making well useless in a very short time.

The Laboratory for the Application of Radioisotopes to Hydrogeology of the Nuclear Energy National Committee («ENEA» at present) in Bari and the Institute of Engineering Geology and Geotechnics of the Bari University had purposed to carry out (since 1962) a series of studies aiming at finding techniques (nuclear or not) suitable for verifying the hydrogeological parameters necessary to have a fresh-water balance, taking into account external discharges, defluxions to sea and yields admitted, depending on fresh-water / salt-water interface depth. Within this framework, a special importance has been attached to the study aiming at finding a rapid and easy method for measuring the groundwater chlorinity.

The relevant study which had been carried out through the analysis of the physical properties of the elements forming the fresh-salt-water, has led to the location of a distinctive feature of the chlorine nucleus thus allowing an easy identification by physical means: the much greater capacity of capturing thermal neutrons (slowed down neutrons) in comparison with any other element generally hold by groundwater.

2. PHYSICAL PRINCIPLES. CHOISE OF THE SOURCE – COUNTER AND CALIBRATION

Thermal neutrons originate in the same water when a fast neutron source is immersed into it.

The conversion from fast neutrons into thermal neutrons takes place nearly instantaneously when the fast neutrons emitted by the source collide above all with the water hydrogen/oxygen nuclei.

In practice, 20 collisions of a neutron with an hydrogen nucleus are sufficient for changing energy from a 5 MeV initial value into the 0.025 eV final one, while some additional collisions need when the target is represented by oxigen.

If a thermal neutron counter is immersed at the neutron source side, it is possible to survey the neutron concentration, which keeps practically unchanged, being the same for any kind of fresh-groundwater, depending on fast neutron source intensity as well as on its distance from the source where the thermal neutron counter is placed.

The thermal neutron concentration becomes notably lower in comparison with the one measured in fresh-water when water contains chlorine because this element destroys a large amount of thermal neutrons it collides with (fish effect).

In comparison with the thermal neutron concentration that may be found in fresh-water, a 25% approximately decrement takes place in the Adriatic sea-water (chlorine content: 20.52 g/l). This phenomenon may be utilized for an easy quick determination of chlorine or of its concentration changes according to the depth.

The minimum chlorine content changes which can be detected depend on the fast neutron source intensity and on the efficiency of the thermal neutron detector.

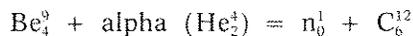
Fig. 1 shows a layout of a neutron probe.

A stainless steel envelope 65.4 cm in length - 4.85 cm in diameter contains a fast neutron source and a thermal neutron counter.

This envelope is watertight; it withstands 20 atmosphere pressure. A cable 200 m in length connects the probe to a battery scaler which is used for counting and recording electric pulses originated in the neutron counter by the collisions of the thermal neutrons with the filling gas.

The fast neutron source consists of a mixture of Americium-241 and Beryllium-9 (Am-241 = 300 mCi).

The fast neutrons emitted on the basis of the nuclear reaction



have the following average energy: 85% = 5.5 MeV (max 11 MeV), 15% = 0.3 MeV. The source intensity is equal to 8.5×10^5 neutron/second.

The Am-241 gamma ray energy is equal to 0.06 MeV, then it is very low. So there is no serious risk due to ionizing radiations or neutrons, considering that the probe is carried within a protective envelope 30 cm in diameter approximately, 40 cm in height, made out of stainless steel covered with lead and filled with paraffin, which is the ideal material for shielding neutrons.

Any damage is avoided when the probe is immersed in water, since this liquid acts as a protective shield against both gamma rays and neutrons.

The thermal neutron counter consists of normal proportional gamma rays detector, but it is filled with boron (boron fluoride).

Boron reacts with thermal neutrons which enter the counter. It captures thermal neutrons and originates electric pulses which are counted by the Nuclear Enterprise 5011 model battery scaler. A thermal neutron flux calculation has been carried out (through the neutron diffusion theory) for verifying actual variations of thermal neutron concentration depending of increasing chlorine contents; then a probe calibration curve had been plotted by correlating chlorine content and thermal neutron counts in three different ways.

A first calibration has been got by mixing Adriatic sea water ($\text{Cl} = 20.50$ g/l) and Apulia aqueduct water ($\text{Cl} = 0.016$ g/l) in different ratios and by recording the thermal neutron counts for each different chlorine concentration.

A second calibration has been obtained by employing sodium chloride, titre 99%.

A third calibration has been obtained directly in the field by correlating neutron counts and chlorine contents of samples taken to this purpose, whose chlorine contents had been ascertained through the Mohr method (silver nitrate - chlorine reaction).

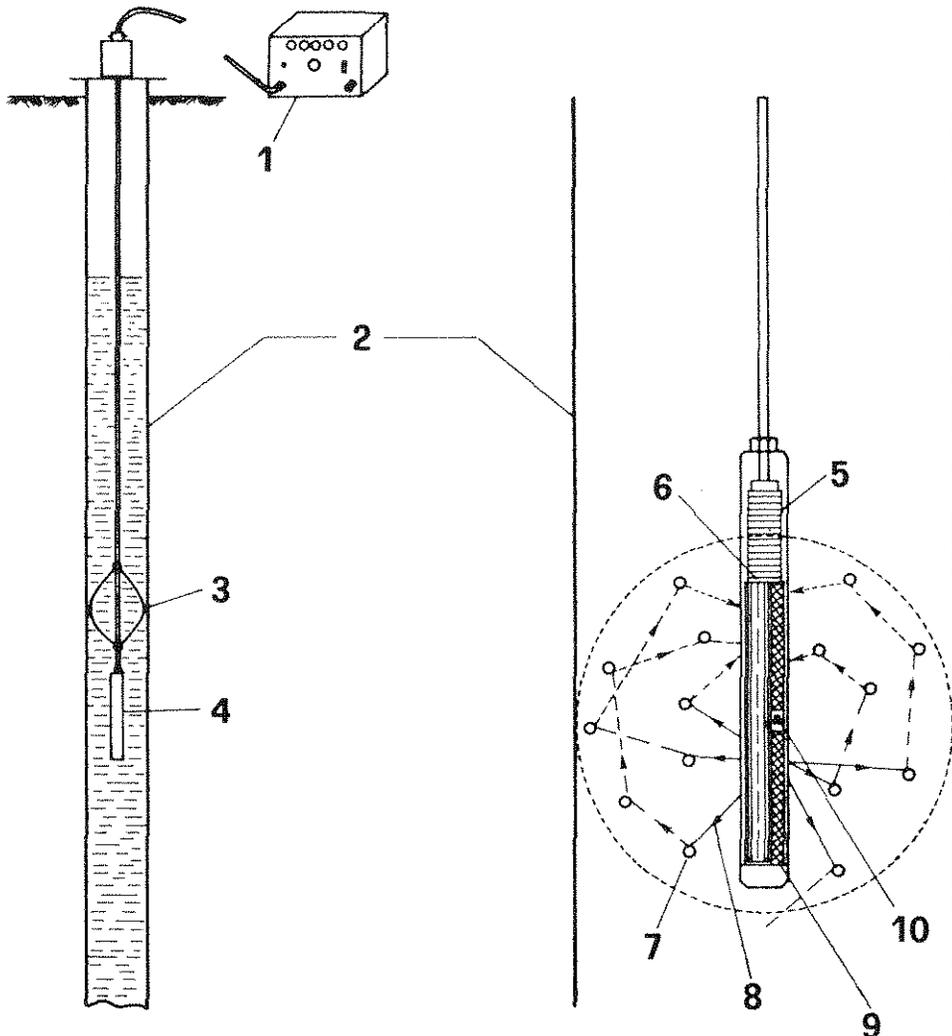


Fig. 1 - In the left side the gauge arrangement in the well: in the right side the drawn of the neutron probe and the scheme of the slowing-down and diffusion of the neutrons (1: Scaler; 2: Covering pipe; 3: Centering device; 4: probe; 5: Pulse preamplifier; 6: Boron trifluoride proportional counter; 7: Atoms; 8: Neutrons; 9: Lead shield; 10: Americium berillium source).

The curve obtained is shown in Fig. 2.

One can immediately notice the remarkable difference existing between the thermal neutron count performed in drinking-water (202,000 counts for minute) and in salt water (155,450 counts for minute) respectively.

By assuming as a minimum detectable chlorine increment the counted quantity three time as big as the standard deviation for a 10 minutes count in fresh-water and in sea-water, a 0.13 g/l and 0.22 g/l is respectively obtained [4].

In order to get a measurement free from any interference, the neutron probe must be surrounded by a minimum water volume such as to allow all fast neutrons emitted by the source to undergo the necessary quantity of elastic collisions and to lose all their energy.

Minimum water volume range forming the so-called probe «influence sphere» is 15 cm (sphere radius) for the energy of fast neutron emitted by the Am-241 - Berillium source.

This condition (called of «infinite medium») is respected in most cases where the neutron probe appears to be helpfull: large diameter digged or drilled wells, marine pools or swallow, sea density currents, etc. Measurements in minor diameter wells are differently influenced; therefore they are distorted by several factors such as the presence of cavities or fissures in the rocks where wells are drilled, water spaces between boreholes and casings, etc. In order to carry out accurate measurements even in such conditions, it is necessary to utilize sources which emit neutrons whose energy is lower than the one of neutrons produced through the Am-241-Be mixture.

A second study, presented in this Meeting [3], had been performed for finding a source of such nature and for getting an appropriate probe.

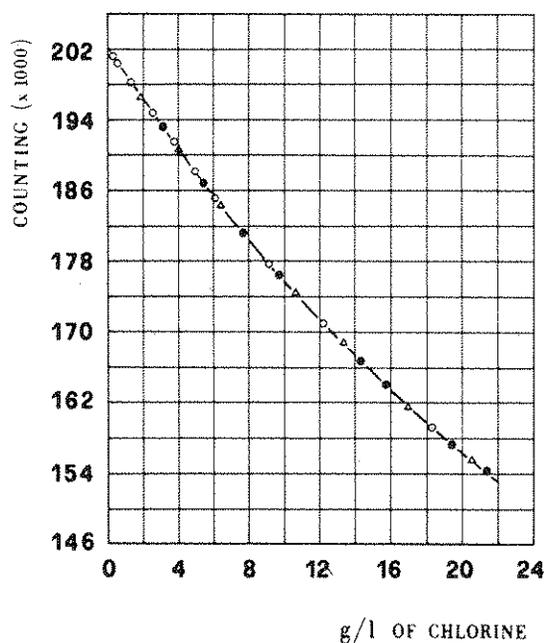


Fig. 2 - Calibration curve of the neutron probe for measuring the chlorine content of the ground-water. The triangles the circles and the squares show the points corresponding to the calibration performed respectively with sea water, with sodium chloride and with fresh water having a known chlorine content.

In that work the reason bringing to choose a source of Americium 241 - Fluorine 19 of 300 mCi (yield of 1.5 million of neutrons / second) are discussed.

Summarily these reasons are: the lower energy (maximum 3.5 MeV) of neutrons produced respectively to neutrons emitted from a Am-241-Be mixture and the good yield of neutrons with reference to an Am-241-Litium-7 source.

The thermal neutron counter is represented by a proportional counter filled with helium-3 instead of boron fluoride. Helium 3 has a higher capacity than boron in reactions with thermal neutrons.

The model adopted (Texlum T 10) has a very pure 10 atmospheres compressed gas.

In studying this neutron probe, the following aspects has been considered: the source-counter unit location with respect to the problem of having a probe as thin as possible to keep around it a water volume sufficient for neutron thermalization even in wells having a very small diameter. In this case it was not possible to adopt the traditional geometry where the source is placed to the counter side. Therefore a preliminary calculation has been performed in order to examine the possibility of placing the source under the counter.

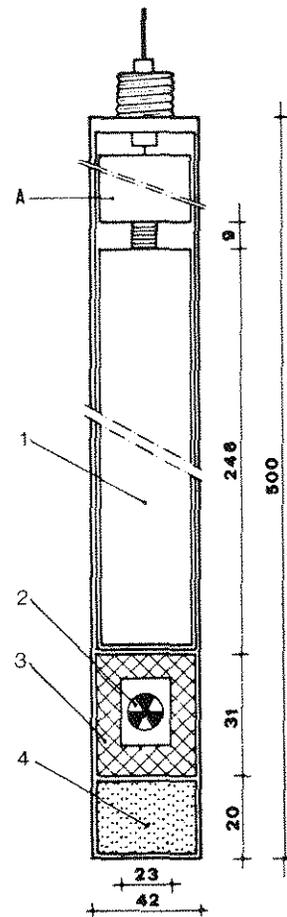


Fig. 3 - Scheme of the neutron probe for measuring the chlorine content of the ground water: 1) preamplifier; 2) counter; 3) neutron source; 4) lead; 5) rubber plug.

This calculation (made through the neutron diffusion formulas) confirmed that the measurement sensibility kept good because, even if the thermal neutron concentration decreases when the source goes away from the counter, the thermal neutron percentage captured by chlorine increases a little.

A layout of such a probe is shown in Fig. 3.

The first purpose of calibration has been to ascertain the count uniformity at water volumes decreasing little by little.

So it has been observed that if containers up to 15 cm of diameter are used, a count uniformity is found with reference to the count obtained in an infinite medium (that is a container 60 cm in diameter), whereas through a container 10 cm in diameter the count rate was 16.9% lower. The probe calibration has been carried out, as previously described, by correlating counts to increasing chlorine quantities obtained both through NaCl and by mixing fresh-water and sea-water.

The new source position as to the counter diminishes neutron concentration around the counter; nevertheless the difference as to sea-water (36.02%) is higher than the one of the former probe (25.74%) and, taken in its complex, the measurement sensibility is such as to allow – by 10 minutes counts – variations up to 0.15 g/l at low chlorine concentration and up to 0.26 g/l at high chlorine content.

3. CONCLUSIONS

On the basis of the same nuclear physics principle (thermal neutron absorption), by the neutron probe it is possible to determine other hydrologic parameters, such as the filtration speed of the groundwater, its direction, the presence of vertical currents in the well.

The filtration speed, for example, can be measured with the «single well method» by putting into water tracers as boron and cadmium (which have a thermal neutron capture cross-section a thousand time greater than chlorine) and following from time to time the concentration dilutions.

This is made by registration of the thermal neutron counting difference in different times after the tracer is put in (The counting rate increases because the cadmium or boron concentration decreases) [2,5].

The same neutron probe can be employed also to determine the infiltration speed of rainwaters in not saturated soil and generally the water content of soil.

This measurement is performed directly correlating the thermal neutron counting and the soil water content (by a correlation curve) on the basis of the much greater slowing-down quality of hydrogen with respect to any other element constituting the soil [1].

The usefulness of the nuclear technics for the solution of hydrogeologic and geotechnic problems has not been till now well understood and all the possibilities have not been explored; then we hope that this work can stimulate a resolute engagement in this research trend.

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