Neutrino mass, radioactivity and the dating of wine

Philippe Hubert, CENBG/IN2P3 – Université Bordeaux 1, France

A few weeks ago, when listening to the radio, I heard a journalist interviewing a physicist on the now very familiar fundamental particle called the neutrino. Following the customary explanations about the origin of the particle, its properties (charge, mass, etc...) and its role in nature, or more precisely its role in the Universe, the journalist made a final remark. 'Working on neutrino properties looks very exciting, but it seems to be of absolutely no use in our practical life'. 'You are absolutely wrong', answered the physicist, 'don't you believe it. As an example, I can tell you that some of the techniques developed in this research are now being used to date wine!'

Since neutrinos are chargeless particle and only sensitive to the weak interaction, they are very difficult to detect, and therefore the physics of the neutrino generally involves very large and difficult experiments. Some of them study naturally occurring sources of neutrinos, such as the sun or cosmic neutrinos, while others use neutrinos from artificial sources such as nuclear reactors or particle accelerators. One of the main challenges of this physics is the mass of the neutrino. At the present time, from the recently published results of several neutrino oscillation experiments, the mass appears to be non-zero, but very small. Another, but indirect way to study neutrino mass is to look for a process called double-beta decay. (In ordinary beta decay a nucleus emits an electron, always accompanied by an anti-neutrino). If a nucleus can emit simultaneously two electrons (with no anti-neutrinos), it implies necessarily that the mass of the neutrino is non-zero, and the frequency (or period) of the decay will give the absolute scale. Until now neutrinoless double-beta decay has not been observed. The limits on the periods lead to a mass lower than 0.5 eV, i.e., one million times less than the electron mass, the next heaviest particle. A double-beta decay experiment call NEMO (Neutrino Experiment with MOlybdenum), in which our group is involved, is now running in the Modane underground laboratory in France. A second one called CUORE, with a totally different technique, is in progress in the Gran Sasso laboratory in Italy and several others are being developed around the world.

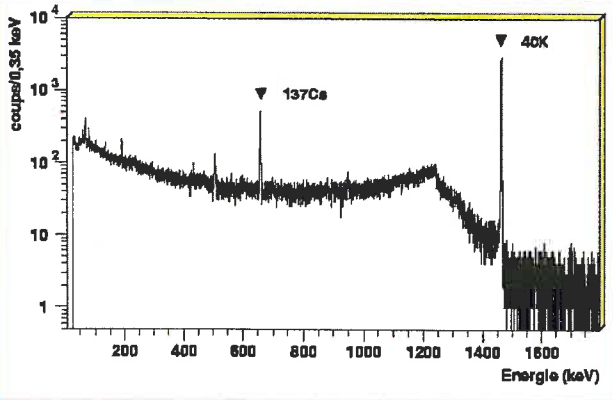
All these experiments have the common feature that they are looking for a very weak signal among a large number of parasitic signals. The physicists involved become rapidly obsessed with the background noise. First of all in order to suppress any effect of the cosmic rays (and the induced neutrons) almost all the experiments have to be installed in deep underground laboratories. Then the detectors have to be protected against the radiation (gamma rays and the remaining neutrons) coming from the surrounding materials, which is achieved by surrounding them with shields made generally of pure lead or iron, or sometimes with water tanks or light Z materials. A third background component is given by the radioactive gas radon that is naturally contained in the air at the level of few Bq/m3. Generally the level of radon has to be decreased by 3 or 4 orders of magnitude, using cold charcoal devices. Finally, the detector itself should be built out of materials containing "no" or only infinitesimal quantities of naturally occurring radioactive elements, such as U, Th, Ra, and their daughters.

Low-background gamma-ray spectrometry

Most of these aforesaid natural radioactive nuclei are gamma emitters. So, in order to be able to select and control the level of radioactive contaminants in materials our group at the University of Bordeaux has been involved over several years in the development of low background gamma-ray spectrometers (in collaboration with an industrial company). Then, hundreds of materials such as metals, glasses, electronics components, glues, screws, etc...have been measured. The expression "low background" in terms of sensitivity means being able to measure activities down to 10 microBq/kg, i.e., about 6 orders of magnitude less than the average level in nature. One may note that the human body contains about 100 Bq/kg, half due to ^{40}K and half to ^{14}C; and in some mushrooms up to 1000 Bq/kg of ^{137}Cs have been recently measured, even almost 20 years after the Chernobyl accident!

The most common technique for gamma-ray spectrometry is based on very pure, large volume germanium crystals (up to 1000 cm^3) cooled down to liquid nitrogen temperature in a special cryostat. The crystal operates like an electronic diode in the reverse bias mode. Any gamma-ray will interact with the crystal through the three most important absorption processes, the photoelectric effect, the Compton effect and the pair-production effect. After interaction the electrons recoil in the crystal, resulting in the creation of electron-hole pairs, which give an electronic pulse whose amplitude is proportional to the absorbed energy. These analogue pulses are then digitised and give an energy spectrum as shown in fig. 1. Any gamma-ray emitted from a radioactive nucleus will give a line in the spectrum with a well defined energy and an intensity proportional to the activity.

In order to be able to measure very low activities, the cryostat is made with highly selected "non-radioactive" materials, is surrounded with a heavy shielding and finally is installed in an underground location. Depending on the required sensitivity, underground means under a mountain such as at Modane (1700 m of rock) or in the basement of a research building at the university of Bordeaux (in this case only a few meters of concrete). In the latter case, a cosmic ray veto made of large plastic scintillators is added to the experimental device. Moreover, special care



▲ **Fig. 1:** Typical gamma-ray energy spectrum of a wine sample recorded with a Ge spectrometer. The 1461 keV ^{40}K and 661 keV ^{137}Cs lines are clearly visible.

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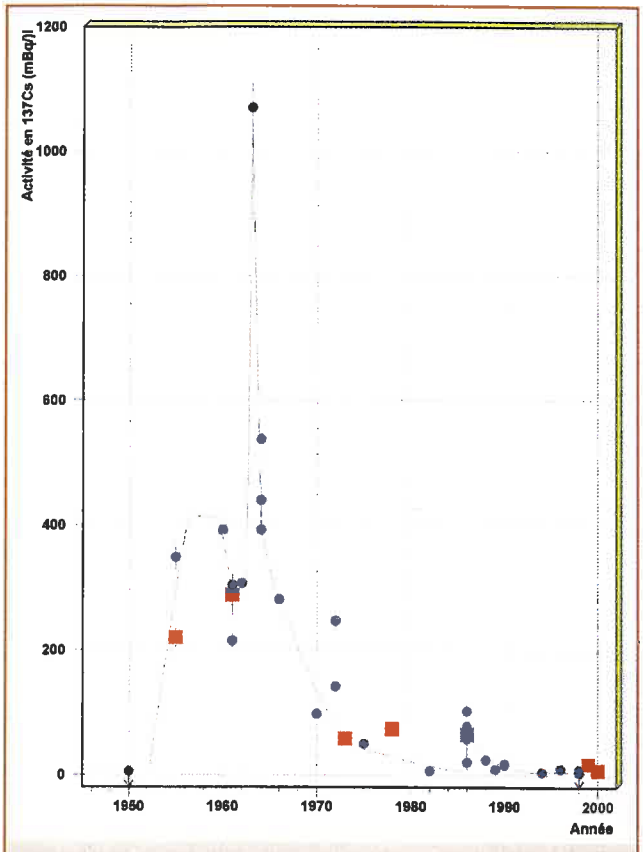

▲ Fig. 2: Activity (mBq/l) of the ^{137}Cs radioactive isotope as a function of the wine millésime. All activities are normalised to the same date, the 1/1/2000. The blue points correspond to measurements after reduction of the wine into ashes, the red squares correspond to non destructive measurements, without opening the bottles. Statistical errors are generally smaller than the dimensions of the points. Points connected with downward arrows are only limits.

has to be taken against any radon penetration inside the shielding. Times of measurements may vary from one day to one month depending on the amount of activity in the sample, and the required sensitivity.

Dating of wine

Such sensitivity in the detection of radioactive isotopes was of obvious interest for other disciplines than physics, and several laboratories in oceanography, geology, archeology, environmental sciences and ...oenology are now currently using these low background gamma ray spectrometers. Why also oenology? It is well known that measuring radioactivity opens the possibility of dating, and therefore why not measuring the radioactivity of a bottle of wine and verify the year or "millésime" written on the label?

Indeed the problem of dating a wine using radioactive techniques is not new: already in 1954 Kaufman and Libby (Nobel prize in 1960 for the ^{14}C dating technique) used tritium ($T_{1/2}$ = 12.3 y) to date several French and Italian wines [1]. About 25 years later, P. Martinière et al. [2] used the ^{14}C isotope to date several Bordeaux wines. The procedure is based on the fact that the amount of ^{14}C in the atmosphere increased by about a factor of 2 after the atmospheric nuclear tests (1950-1963), and then decayed regularly. However these techniques need the bottles to be opened and a sample to be prepared from distillation. Moreover the tritium technique is nowadays more difficult due to the fallout from nuclear reactors, and the ^{14}C technique can give a wrong dating result where there has been the addition of any product such as sugar. As a consequence, the DGCCRF[1] asked if it was not possible to find through our low background gamma-ray spectrometers another possibility for dating. This explains why, when seeking to measure the mass of the neutrino, one opens a bottle of fine old Bordeaux wine to measure its radioactivity!

In order to obtain the best available sensitivity, the first measurements were made after evaporation of the wine, and burning the residue into ashes. The first results showed that wine essentially contains the isotope ^{40}K (see fig. 1) at the level of 30 Bq/l, which corresponds to about 0.9 g of natural potassium in each liter of wine. There is nothing surprising about that, since wine contains, even if connoisseurs may not like it, a fair amount of potassium bi-tartrate. But far more surprising was the fact that certain bottles of wine contain also the isotope ^{137}Cs ($T_{1/2}$ = 30 y), a man-made radioactive isotope, whose origin is from nuclear fission. Even more interesting was the fact that the amount (or activity) varies as a function of the year. All that remained to do was to find a set of bottles with certified "millésimes" and to carry out measurements of the ^{137}Cs radioactivity as a function of the year. The curve shown in fig. 2 was readily obtained [3]. First, wine connoisseurs can reassure themselves, the observed activities are small, always less than 1 Bq/l. Then the results show that the wine keeps a memory of atmospheric nuclear testing (the years 1950-1963) and the accident of Chernobyl (1986). It is obvious that such a curve can be exploited to estimate the age of a given wine, at least to control the year written on the label, and possibly to detect any anomalies. For example, any wine before 1950 should not contain any detectable ^{137}Cs activity!

Even if established only with Bordeaux wines, we think that the 1950-1985 part of the curve can be used for any wine from the northern countries since the ^{137}Cs reservoir is the high atmosphere. For 1986 and beyond, it is well known that the ^{137}Cs fallout due to Chernobyl is strongly dependent of the geographical localization. For example, within the 1986 Bordeaux wines, the ^{137}Cs activity varies from a few mBq/l up to a 100 mBq/l. For French eastern 1986 wines the activity may even reach up to 1000 mBq/l!

At this point it is important to remark that dating using the ^{137}Cs activity from nuclear fallout is not new, but is already used in other disciplines, such as in geology to date sediments. What is new with our technique is the possibility to measure activities down to a few mBq/l

Non-destructive dating of wine

Even if the process of reduction of a wine into ashes is rather easy, it cannot be systematically used, specially with old "grands crus" for which prices can be rather expensive. And beside the price, everybody would prefer to taste and drink such a wine than to reduce it into ashes. Therefore it became rapidly important to work on a non-destructive way of dating.

In principle there is no argument against such a non-destructive measurement since the energy of the gamma-ray emitted by the ^{137}Cs nucleus, 661 keV, is high enough to escape the wine and to cross the glass of the bottle before to making an interaction with the Ge crystal. It is only necessary to set a bottle close to the endcap of the detector, and to record the energy spectrum. Fig. 3 shows the set-up currently used in Bordeaux, where up to four

[1] Direction Générale de la Concurrence, de la Consommation et de la Répression des Fraudes, a French government agency charged with protecting consumers, assuring fair competition and preventing fraud.

identical bottles can be measured at the same time. However, such a procedure implies a serious loss in the sensitivity of the measurement: firstly because the geometry of the source becomes quite large, and secondly, because the rather high level of radioactivity in the glasses (^{40}K, U, Th, Ra and their daughters) increases the background of the spectra. Nevertheless the first measurements have shown that within these experimental conditions, the ^{137}Cs gamma line is still observable down to an activity of 20 mBq/l. The first activities measured this way are shown in fig. 2 as red squares and are in very good agreement with the values obtained from the ashes. Of course we have carefully checked that neither the glass nor the cork themselves contain strong ^{137}Cs contaminations.

Summary and outlook

Since the pioneering measurements given in fig. 2, much more data have been accumulated, mainly for Bordeaux wine. For a given year some dispersion in the activity values is observed, due to statistics and to non-uniformity in the ^{137}Cs fallout which depends on the local amount of rain or aerosols. However the average values are always very close to the curve of fig. 2, and therefore now we consider that the method is validated.

Of course we are well aware that the curve of fig. 2 is not without any ambiguities. A measured ^{137}Cs activity of 100 mBq/l may correspond to the years 1953 (rare!), 1970 or 1986. Another weak point is that the ^{137}Cs activity in a "young" wine, beyond 1985, is rather low and/or strongly dependent on the Chernobyl fallout. But this curve is the only data available that is provided by human activity! For a particular analysis, we would recommend that first a non-destructive measurement should be carried out, and if the answer is not satisfactory, then one can always reduce the sample into ashes and proceed to a more sensitive gamma analysis.

Nowadays we are working on the possibility to use another radioactive isotope that is also present in wine, namely ^{210}Pb ($T_{1/2}$ = 22 y). The origin of this isotope is the radon inside the air. After decay, the radon daughters, such as ^{210}Pb are fixed on dusts or aerosols and then fall down on the soil, plants, grapes, and finally enter the wine. If we know the amount of ^{210}Pb at the time the wine is bottled (t =0), then dating is possible using simply the exponential law of decay. However, there are several drawbacks with this method:1) the activity at the time t = 0 is rather weak, around 100 mBq/l which implies the need for very precise activity measurements; 2) until now we don't know if the t = 0 activity is constant with the year and with the "terroir"; 3) the gamma-ray emitted in the decay of ^{210}Pb is rather weak in intensity (4%) and low in energy (46 keV) and therefore only measurements with a wine reduced into ashes can be envisaged; and 4) the 30 Bq/l of ^{40}K, always present in the wine, gives a severe high background level in the gamma spectra, due to the Compton of the 1461 keV gamma (10% of the decays), and to the bremsstrahlung of the beta-rays (90% of the decays). Nevertheless, recent measurements with a new low-background well-type Ge spectrometer appear to be encouraging.

Finally, it is important to stress that with the present sensitivities, i.e., around 1 mBq/l, obtained with this new low-background well-type Ge spectrometer, a few other natural radioactive isotopes are simultaneously observed in the same spectra, such as Ra, Th and their progenies. The activities are even lower, around 10-20 mBq/kg, but still easily measurable. From the equilibrium situation within the chains we obtain again new information on the age of the wine. As a result, with our present sensitivities, we obtain within the same gamma ray spectrum three or four independent possibilities to date a wine (^{137}Cs, ^{210}Pb, radium and thorium isotopes), and we hope to obtain the necessary accuracy to one year, even for young wines. However many more measurements are still necessary before the method can be validated. And now, we are wondering if these levels of radioactivity could not also be a signature of the "terroir"?

A suivre… as we say in France!

References

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▲ **Fig. 3:** View of the experimental set-up for a non-destructive dating measurement of the wine. The Ge crystal is kept in vacuum inside the aluminium cap, in the center of the picture. With this device up to 4 identical bottles can be measured at the same time, and the sensitivity in the ^{137}Cs measurement is around 20 mBq/l.