KTH Royal Institute of Technology
Nuclear Energy Engineering

Neutron Lab

Radiation, Protection, Dosimetry and Detectors SH2603 HT24

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1 Introduction

This experiment aims to investigate the activation of materials and their decay. When a material is exposed to a sources of neutron, it may happen that one of them is absorbed by the nuclei of the target. The resulting isotope could still be stable or becomes radioactive. In case of an unstable isotope, this process is called *neutron activation* and it undergoes radioactive decay.

The time scale needed for the activation will be dependent on the cross-section, but also the half-life plays an important role for the following detection. An high cross-section translates in a short activation time, and it necessary in order to observe it within the timescale of the laboratory session. Moreover, also the half-life should be long enough to allow a proper detection.

In order to delve deeper into these topics, it will be analyzed the aluminum activation and decay; it will be measured the half-life of silver and also investigate an unknown material, trying to understand its composition by the characteristic energy spectrum.

2 Theory

2.1 Neutron Activation and Decay

During the activation process from a stable to an unstable nuclide, the number of new radioactive nuclide has two contribution:

- A source due to neutron capture;
- A sink due to radioactive decay.

The resulting formula for the evoution in time of the number of activated nuclides per unit volume:

$$N_a(t) = \frac{\sum_c \phi}{\lambda} \left(1 - e^{-\lambda t}\right); \tag{1}$$

As time approaches infinity, the maximum number of activated nuclides per unit volume under specific neutron irradiation conditions is reached:

$$N_{a_{max}}(t) = \frac{\sum_c \phi}{\lambda} = \frac{\sigma N \phi}{\lambda}; \tag{2}$$

Where:

- σ is the microscopic cross-section,
- N is the number of silver nuclides per unit volume.

To achieve maximum activation of the material, it is necessary to maximize the ratio:

$$\frac{N_a}{N_{a_{max}}} = \left(1 - e^{-\lambda t}\right). \tag{3}$$

After the target materials has been irradiated, the activated nuclei keep to decay, without any source term. This process can be expressed with equation 4.

$$N_c(t) = N_a\left(t_a\right) e^{-\lambda t}; \qquad 0 \leq t \leq t_c \tag{4}$$

Where t_a refers to the activation time, t_c to the cooling time over which the sample material is removed from the neutron bath and brought to the detectors.

Using a detector, we can measure the decay of this activated material. These decays can be described using Eq. 5

$$N_m(t) = N_c(t_c) e^{-\lambda t}; \qquad 0 \leq t \leq t_m \tag{5}$$

The measured counts N_m are expressed in units of decays per unit volume.

2.2 Physics of Detection

To understand what we measure when we insert an irradiated material inside a gamma detector, we must explain with what physical processes the detection works.

2.2.1 Photon Interactions With Matter

Gamma ray detection is the measured results of gamma ray interactions with matter. There are three distinct mechanisms in which this can occur; Compton scattering, pair production, and the photoelectric effect. Compton scattering occurs when the gamma ray collides with an electron causing an inelastic collision to occur as the electron absorbs some of the gamma energy. If both the electron and the scattered photon can be detected then the initial gamma energy can be inferred. Pair production occurs when the photon passes in close proximity to the nucleus causing the creation of an electron-positron pair. This electron-positron pair will eventually collide annihilating each other and creating a photon pair that can easily be detected to infer the gamma energy. The photoelectric effect is essentially the process that occurs when an atom absorbs a photon, in this case a gamma ray, and consequently emits an electron [1].

2.2.2 Sodium Iodide Detector

The Sodium Iodide Detector, which is an example of a Scintillator detector system, is highly efficient in gamma detection. Due to the high Z of iodine and the consequent large number of electrons to interact with Sodium Iodide detectors make excellent scintillation detectors. They are a cheaper option when it comes to detecting equipment but are generally only good for spectrometry [1].

It contains a NaI crystal, which takes the incoming gamma ray and turns it into visible light. The photocathode turns this into an electrical signal, which is amplified and then sent to processing electronics like the Multi-Channel Analyzer (figure 1) [1].

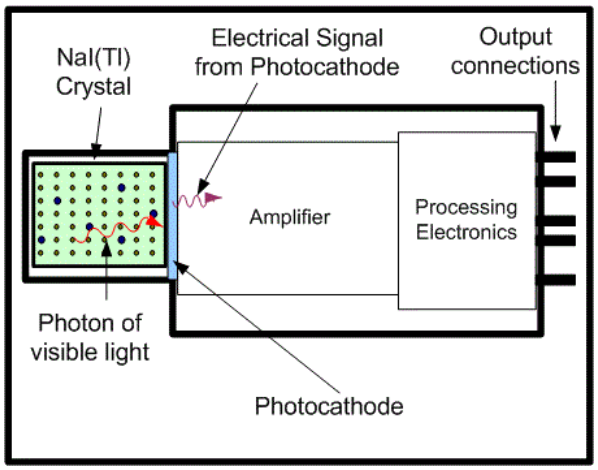

Figure 1: A depiction of the physical process that happens inside a NaI detector

3 Experimental Setup & Procedure

The laboratory exercise began with us calibrating the gamma detector using two common known sources, Cs-137 and Co-60. By undergoing β^- decay, their daughter nucleus is left in an excited state which then undergoes gamma decay, with characteristic energies that can be seen in table 1.

Table 1: Literature Peak Energy Values and their corresponding Channel Number.

Channel	**Energy [keV]**	**Nuclide**
1428.86	661.66	Cs-137
2488.05	1173.23	Co-60
2816.71	1332.49	

Then, we could proceed with the calibration, as can be seen in figure 2

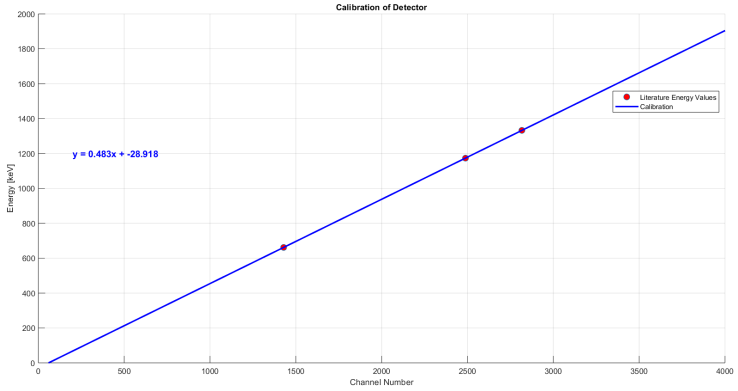

Figure 2: The Gamma Detector Calibration

In all of the experiments that were conducted, materials were irradiated inside a large bath of water which contained an alpha-neutron source, Am241-Be. The samples were irradiated while attached to long holders placed inside the neutron bath.

Afterwards, when the samples were activated we could remove them from the neutron bath and place them inside the gamma detector for spectrum measurements with the software Tukan8k.

Before we measured the energy spectrums of activated materials, we measured the background spectrum (figure 3) in order to subtract it from the resulting spectrums in our analysis.

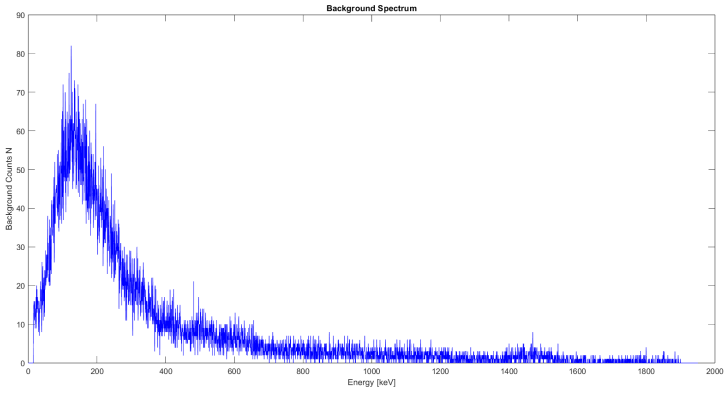

Figure 3: The measured background spectrum

4 Results and Discussion

4.1 Aluminium activation and decay

The first material irradiated by the neutron source is aluminium. According to the IAEA Live Chart of Nuclides, the only isotope available on the earth is ^{27}Al. During its exposure in the bath, the neutron emitted by the Am241-B source, may be absorbed by the nucleus, resulting in ^{28}Al.

After the absorption, the ^{28}Al has only one decay path: as we could expect when an excess neutron makes the nucleus unstable, it decay β-.

$$^{27}\mathrm{Al} + n \rightarrow ^{28}\mathrm{Al} \rightarrow ^{28}\mathrm{Si} + e^{-} + \overline{\nu_e} \tag{6}$$

The average energy associated to the electron is 1241.80keV while the maximum energy is 2863.22 keV. Moreover, as may happen after a *beta-* decay, the daughter nucleus is still unstable, and the excess energy is released by γ decay. The resulting photon will have an energy of 1778.987 keV.

For this reason, β or γ detector are able to observe the effect of this decay, but with different outcomes. With the beta detector we are supposed to see the distribution of the energy of the electron, while with the gamma detector we should be able to observe the discrete energy of the photon. In this way we can more clearly see that what is expected is really happening thanks to the unique peak.

According to Eq. 1, to reach the saturation, the exponential part should tend to zero. thus after an infinite time, the maximum number of activated nuclides should be reached, as reported in Eq. 2.

However, after 5 half-life the ratio between the number of nuclides and its maximum approaches the 99.3%.

Therefore,using the half-life $t_{1/2} = 134.7s$ from our reference [3]:

$$\frac{5}{\lambda} = \frac{5t_{1/2}}{ln(2)} = 971.66s \approx 16 \text{ min}$$

The aluminum sample, after the irradiation is moved from the bath to inside the γ detector and after a measurement time of 506 s, the result from Tukan are represented below:

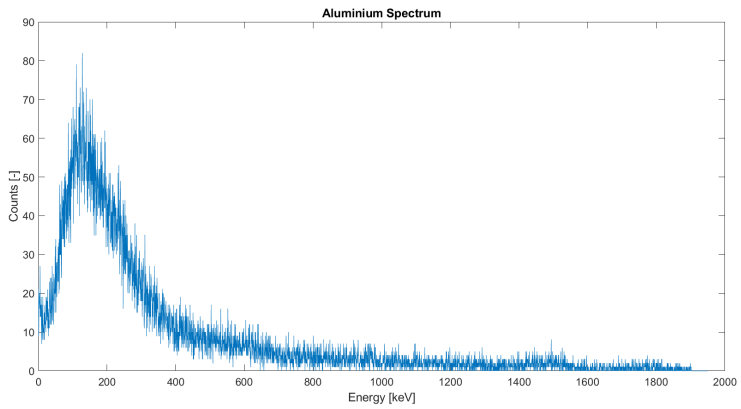

Figure 4: Energy spectrum of ^{28}Al inside gamma detector.

Differently from what was previously stated, there is no peak at a corresponding energy of 1778.987 keV, as we could expected from the gamma decay.

Considering that the time inside the detector was approximately 3.75 $t_{1/2}$, it may be supposed that it was long enough for the background spectrum "hide" the peak. For this reason, the counts of Fig.4 and Fig.3 are subtracted.

However, also in Figure 5 there is no noticeable peak. The main explanation could be that a too long interval of time has passed between the removal from the bath and the acquisition of data. Considering the relatively short half-life, one or two minutes longer than necessary, could compromise the result.

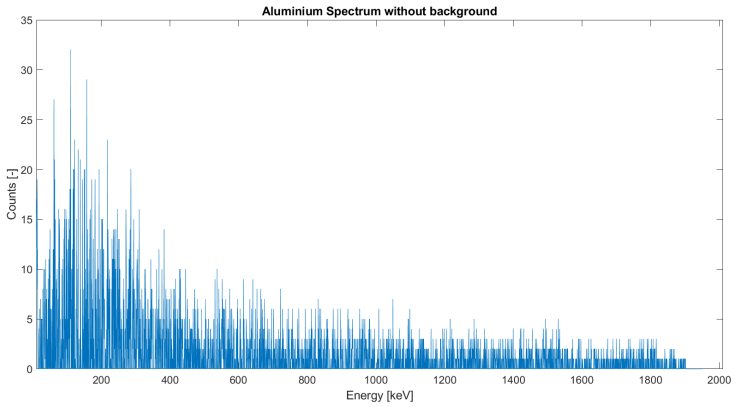

Figure 5: Energy spectrum of ^{28}Al without the background spectrum.

4.2 Half-life Estimation of Activated Silver

The sample was maintained in the bath for over 20 minutes, ensuring complete activation, considering the longer half-life corresponding to ^{108}Ag.

Recalling the Equation 3, examining the ratio between the maximum number of activated nuclides per unit volume for ^{108}Ag and ^{110}Ag yields:

$$\frac{N_{a_{max_{^{108}\mathrm{Ag}}}}}{N_{a_{max_{^{110}\mathrm{Ag}}}}} = \frac{\sigma_{^{107}\mathrm{Ag}}}{\sigma_{^{109}\mathrm{Ag}}} \cdot \frac{N_{^{107}\mathrm{Ag}}}{N_{^{109}\mathrm{Ag}}} \cdot \frac{\lambda_{^{110}\mathrm{Ag}}}{\lambda_{^{108}\mathrm{Ag}}} = 2.59.$$

This result indicates that when both natural silver nuclides are fully activated in the bath, the ratio of the respective activated nuclides will differ significantly from the natural abundance of their parent isotopes:

$$\frac{51.839\%}{48.161\%} = 1.08$$

This discrepancy is primarily attributed to the longer half-life of ^{108}Ag, in other words, since this decays slower than ^{110}Ag, the neutron flux is capable to mantain a larger number of these activated nuclides before reaching equilibrium.

4.2.1 Fitting method and determination of half-life

The experimental data was fitted using a two-term exponential model, representing the total counts of the decays of the two activated silver isotopes, ^{110}Ag and ^{108}Ag. The model applied includes scaling by the natural abundance percentages of the isotopes and is expressed as follows:

$$a \cdot e^{-\lambda_{110} \cdot t} + b \cdot e^{-\lambda_{108} \cdot t} \tag{7}$$

where:

- λ_{110} represents the decay constant of ^{110}Ag,
- λ_{108} represents the decay constant of ^{108}Ag,
- a b are scaling factor

The fitting process assigned weights as inversely proportional to the variance $(1/\sigma^2)$ of the data, thereby minimizing the influence of data points with higher uncertainty. The fitting curves is shown in Figure 6. Thanks to the fitting process, the decay constant has been found as long as the half-lives:

$$\lambda_{110} = 0.01909\,\mathrm{s}^{-1} \pm 0.0051\,\mathrm{s}^{-1} \tag{8}$$

$$\lambda_{108} = 0.00314\,\mathrm{s}^{-1} \pm 1.8127 \times 10^{-4}\,\mathrm{s}^{-1} \tag{9}$$

$$t_{1/2,110} = \frac{\ln(2)}{\lambda_{110}} = 36.31\,\mathrm{s} \tag{10}$$

$$t_{1/2,108} = \frac{\ln(2)}{\lambda_{108}} = 220.62\,\mathrm{s} \tag{11}$$

4.2.2 Derivation of uncertainties on the half-lives

The uncertainties in the half-lives were derived from the uncertainties in the fitted decay constants λ_{110} and λ_{108} using the formula for half-life:

$$t_{1/2} = \frac{\log(2)}{\lambda}$$

Since this is a non-linear relationship, the propagation of errors requires applying the general formula for error propagation in functions of a single variable:

$$\sigma_{t_{1/2}} = \left| \frac{d}{d\lambda} \left(\frac{\log(2)}{\lambda} \right) \right| \cdot \sigma_\lambda$$

Thus, the uncertainty in the half-life for ^{110}Ag is:

$$\sigma_{t_{1/2,110}} = \frac{\log(2)}{\lambda_{110}^2} \cdot \sigma_{\lambda_{110}} = \pm 9.70\,\mathrm{s} \tag{12}$$

Similarly, the uncertainty in the half-life for ^{108}Ag is:

$$\sigma_{t_{1/2,108}} = \frac{\log(2)}{\lambda_{108}^2} \cdot \sigma_{\lambda_{108}} = \pm 12.73\,\mathrm{s} \tag{13}$$

These uncertainties are propagated from the precision of the fit to the decay constants, ensuring the error on the half-lives accounts for the inherent uncertainty in the decay constants themselves.

4.2.3 Comparison with literature values

The computed half-lives were compared with known literature values:

- For ^{110}Ag, the reported half-life is approximately 24.6 seconds;
- For ^{108}Ag, the reported half-life is approximately 2.41 minutes (144.6 seconds).

The comparison between the fitted results and the reference values facilitates an assessment of the accuracy of the experimental data. The calculated intervals do not encompass the corresponding literature values. For ^{108}Ag, the deviation is minimal, while for ^{110}Ag, the order of magnitude is maintained despite a significant discrepancy. Any deviations can be attributed to experimental uncertainties or potential limitations in the fitting model.

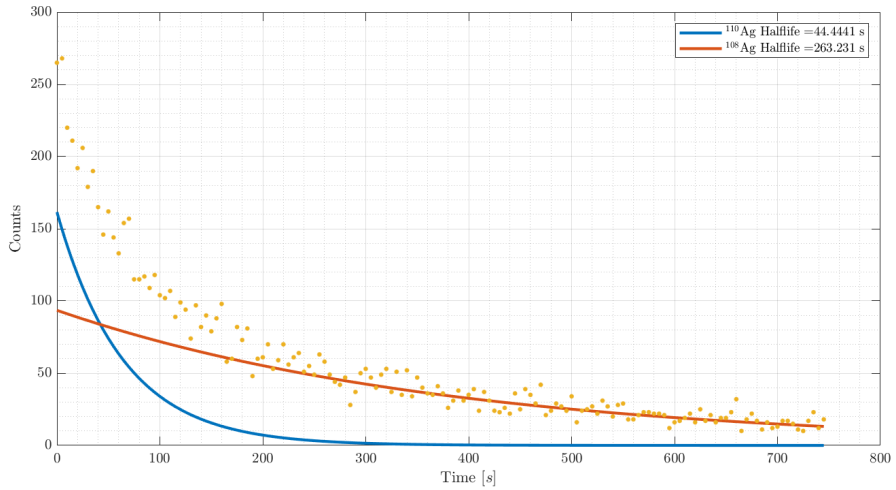

Figure 6: Fitting exponential models

4.3 Investigation of a Mystery Material

For the final part of the laboratory, our supervisor provided us an alloy of an unknown composition, after irradiating it in the neutron bath for a duration sufficiently close to the activation time, and instructed us to place it inside the gamma detector.

With the use of Tukan, we continued to measure a spectrum, in which we had to analyse the energy peaks to derive the composition of the mystery alloy.

The resulting gamma spectrum after removing the background, of the mystery alloy we measured can be seen in figure 7.

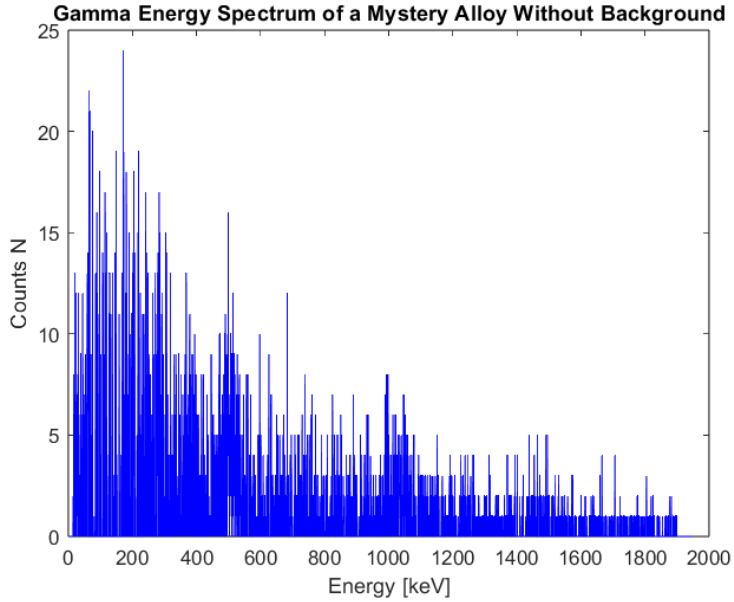

Figure 7: The gamma spectrum of the activated mystery alloy without the background

Before we made any assumptions based on the energy peaks of the spectrum, we took a good look on the alloy, and its color which had a gold-like hue. We assumed it consisted of Copper and another element. Since natural copper consists an abundance of 69.15% ^{63}Cu and 30.85% ^{65}Cu, its clear that

the activated sample would have the isotpes ^{64}Cu and ^{66}Cu. To check if it was indeed copper, we checked the energy of the gamma emissions from those unstable nuclides and the values can be seen in table 2

Table 2: Literature energy values of gamma photons after the decays [7].

^{64}**Cu** γ **energy [keV]**	**Intensity**	^{66}**Cu** γ **energy [keV]**	**Intensity**
511	35%	1039.2	9.23%

It must be noted that ^{64}Cu undergoes electron capture (E.C.) and β^+ decay with a rate of 61.5%, and the origin of the photon is from the annihilation of positrons and electrons that produce two 511 keV energy photons. The other gamma emissions are considered negligible because of their very low intensity.

The other unstable nuclei, ^{66}Cu decays by β^- emission 100% of the time and the most probable states that the daughter ^{66}Zn is left in, is the ground state (90%) and the 1039.2 keV excited state (9.01%) as can be seen in figure 8.

Then, we proceeded by locating energy peaks around those specific energies (table 2). The peak energies we found on the measured spectrum 7 where beteween 489.805 and 513.303 keV, as well as at 1046.35 keV. This heavily indicates that the alloy contains the decaying ^{64}Cu and ^{66}Cu isotopes.

Besides those clear energy peaks of copper on the measured spectrum 7, there isn't any other significant peak that can be considered as a gamma energy of decaying nuclei. One element that couldn't produce enough counts comparable to the ones of the copper isotopes, is Zinc. The unstable isotope of zinc, ^{65}Zn has a half-life of 244 days which within the context of the experiment duration, is very long. The other unstable isotope, ^{69}Zn may have a much shorter half-life, but the intensity of the gamma emissions is negligible. Thus, if the alloy also consisted of Zinc, after the activation it is justified that we couldn't observe any energy peaks besides the ones of copper.

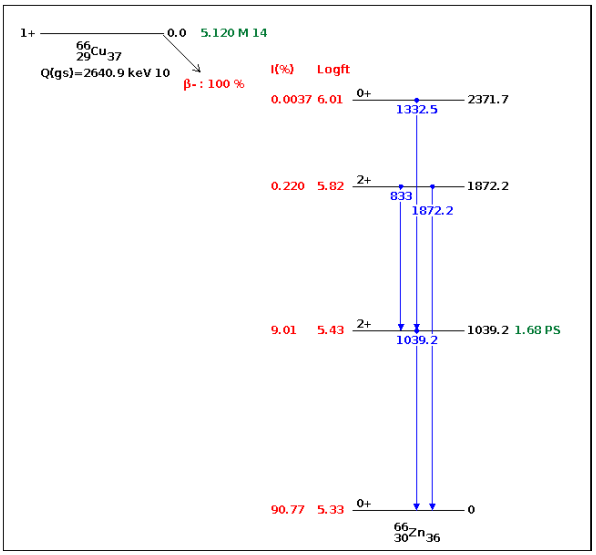

Figure 8: The decay scheme of the radioactive nuclide ^{66}Cu.

5 Conclusions

5.1 Aluminium Activation

To sum up, a sample made of aluminium is activated in a bath thanks a neutron source. The resulting radioactive nuclide, undergo a β- decay and its daughter a following γ decay. However, the discrete peak of the γ photon can't be appreciate from the energy spectrum measured. Our hypothesis is that the time spent to transfer the sample from the bath to the detector was too long and allows the decay of a significant amount of activated nuclei.

5.2 Half-life of activated silver

The measured half-lives are consistent with those found in the literature; however, they deviate from the reported values by non-negligible amounts. These discrepancies could be attributed to the time delay between the extraction of the silver from the tank and the start of the detection process. Additionally, the fitting method employed may not be entirely suitable for determining the parameters of the exponential models.

5.3 Mystery Alloy

Our conclusion that the mystery activated alloy was brass is a sound resolution after observing that only gamma photons of the copper's radioactive nuclei decay can be measured on the spectrum. Any other alloy that consists of copper and other elements would not only emit measurable gamma energies of the other isotope's decay, but it would have a different color.

Overall, the activation of the alloy and its spectrum measurement on a NaI gamma detector turned out to be an effective method of determining its composition. On the other hand, the use of a Germanium Detector would provide us with extremely precise energy measurements and higher certainty of our conclusion.

References

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