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Residual radioactivity of silicon doped by transmutation

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Résumé. — Nous avons étudié la double réaction nucléaire ($\text{Si-30} \rightarrow \text{Si-31} \rightarrow \text{P-31} \rightarrow \text{P-32}$) qui produit l'isotope radioactif P-32 par irradiation du silicium par des neutrons thermiques. Une équation a été établie afin de relier la radioactivité de P-32 à la section efficace de capture de P-31. La section efficace ainsi déterminée à partir des mesures de la radioactivité de P-32 est en bon accord avec la valeur classique. Il est ainsi possible de prédire la radioactivité du silicium dopé par transmutation nucléaire pour toute dose et à tout moment. Une équation a également été établie pour relier la radioactivité de P-32 au nombre de donneurs créés.

Abstract. — The complex nuclear reaction ($\text{Si-30} \rightarrow \text{Si-31} \rightarrow \text{P-31} \rightarrow \text{P-32}$) leading to the formation of radioactive P-32 by irradiation of silicon with thermal neutrons, has been studied. An expression has been derived to correlate the activity of P-32 with the neutron cross-section of P-31. The neutron cross-section thus computed from the experimentally measured activity of P-32 is in good agreement with the classical value. It is then possible to predict the radioactivity of silicon doped by nuclear transmutation for any neutron dose at any time. An expression has also been derived to correlate the activity of P-32 to the number of donors created.

1. Introduction. — Neutron transmutation doping using the following reaction :

is being used to convert ingots of high resistivity silicon into striation free, precisely doped n-type material [1, 2]. This neutron transmutation doped material is being successfully used in the production of high voltage thyristors and diodes [3, 4]. Recently, a comparatively large neutron flux

$$(\sim 10^{19} \text{ neutrons.cm}^{-2})$$

has been employed to produce precisely and homogeneously doped n-type epitaxial layers [5].

In an earlier publication [6], it was shown that the phosphorus concentration thus produced by neutron irradiation is uniform even at a microscopic level (500 Å). The neutron irradiation not only produces phosphorus but also activates the background impurities present in silicon. This was utilized to study the distribution of gold in silicon from the surface into the bulk of the crystal which was found to be non-

uniform. The neutron irradiation also gives rise to another nuclear transmutation :

The aim of the present work is to study this secondary transmutation. The half-life time of P-32 is 14.3 days as compared to 2.62 h of Si-31. Thus this can be studied as residual radioactivity of neutron irradiated silicon samples. We shall first derive the expression for the radioactivity of P-32 to evaluate the neutron cross-section. We shall then give the numerical expression relating the residual beta-radioactivity of the neutron doped silicon to the number of carriers produced.

2. Theory : radioactivity of P-32 atoms. — Several processes occur inside the reactor :

a) the production of Si-31 from Si-30, b) the transmutation of Si-31 into P-31, c) the activation of P-31 to P-32 and d) the decay of P-32 to S-32. Besides these reactions, radioactivation of impurities and activation of Si-28 and Si-29 also take place but are not of interest in the present case.

The total activity, A , per g of silicon in 4π solid

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angle due to Si-31 at the instant the sample is removed from the reactor is conveniently given by [6] :

$$A = \frac{36 \times \sigma \times J}{M} \times [1 - \exp(-\lambda t_1)] \times AI \times PM \text{ cpm per g} \quad (3)$$

where

σ = neutron cross-section of Si-30 = 0.102 barn,
 J = neutron flux in neutrons/cm²/s,
 λ = radioactive constant of Si-31 = $7.35 \times 10^{-5} \text{ s}^{-1}$,
 AI = isotopic abundance of Si-30 (= 0.030),
 PM = the mode of decay (= 1, for this case),
 M = atomic mass of silicon,
 t_1 = time of irradiation.

Time t is taken as zero at the instant samples are removed from the pile. The activity as given by the counter at any time t_2 in counts per minute per gram of silicon is

$$A_{\text{counter}} = A \times RC \times \exp(-\lambda t_2) \text{ cpm per g} \quad (4)$$

where RC stands for the efficiency of the counter.

Because of the short half-life time of Si-31, this radioactivity A quickly tends to zero. On the contrary, the radioactivity of P-32 decays much more slowly and after a few days it is the principal contributor to the radioactivity. Let A' denote this radioactivity that we are now going to relate to the neutron dose and finally, the number of carriers.

To a good approximation, the number of P-31 atoms which appear is equal to the number of Si-31 atoms which disappear. A few hours after putting the samples in the nuclear reactor, one has

$$1 - \exp(-\lambda t_1) \simeq 1.$$

Then, the number of P-31 atoms produced between time t and $t + dt$ is :

$$dN = \frac{36 \times \sigma \times J}{M} \times AI \times dt \quad (5)$$

having the infinitesimal mass (per gram of silicon) :

$$dm' = \frac{36 \times \sigma \times J}{M} \times AI \times \frac{M'}{N_A} dt \quad (6)$$

where M' denotes the atomic mass of P-31 and N_A the Avogadro's number.

These dN atoms are subject to the neutron flux during the remaining time $= t_1 - t$ (Fig. 1). These will give the infinitesimal contribution to the radioactivity of P-32 at the time t_2 :

$$dA' = \frac{36 \times \sigma' \times J}{M'} \times \frac{[1 - \exp\{-\lambda'(t_1 - t)\}]}{\exp(\lambda' t_2)} dm' \times RC \quad (7)$$

Fig. 1. — Schematic variation in the activity of P-32 with time inside and outside the reactor.

where σ' is the neutron cross-section of P-31 in barn. PM and AI are unity for P-31 activation. Substitution of dm' from (6) into (7) gives

$$dA' = \frac{(36 J)^2 \sigma \sigma'}{N_A \cdot M} \times \frac{[1 - \exp\{-\lambda'(t_1 - t)\}]}{\exp(\lambda' t_2)} \times AI \times RC \times dt. \quad (8)$$

The total activity due to all P-32 atoms produced in time t_1 is thus obtained by integrating between $t = 0$ to $t = t_1$ as

$$A' = \frac{(36 J)^2 \sigma \sigma'}{N_A \cdot M} \times \frac{\left[t_1 - \frac{1}{\lambda'} \{ 1 - \exp(-\lambda' t_1) \} \right]}{\exp(\lambda' t_2)} \times AI \times RC. \quad (9)$$

3. Experimental results. — *Hyper pure* silicon crystals supplied by *Wacker* were used. They are float-zone, dislocation free, grown in the 1-0-0 orientation, p-type containing boron, the concentration of boron ranging from 10^{13} up to 10^{16} atoms/cm³. The neutron activation analysis confirms the high purity if one excepts the gold which is

$$3 \times 10^{13} \text{ atoms/cm}^3.$$

The samples, carefully washed and cleaned, were normally irradiated in a thermal neutron flux of $3.5 \times 10^{13} \text{ neutrons cm}^{-2} \text{ s}^{-1}$ for a period of 7 days. Longer irradiation time helps to increase the measurable radioactivity of P-32.

The samples were again given a wash

$$(5\% \text{ HF} + 5\% \text{ HNO}_3)$$

to remove any accidental surface contamination. The radioactivity was measured in a low background (1 cpm) anti-coincidence beta-counter 14 days after removal from the pile. This ensures a complete decay of Si-31 and Au-198. For some wafers a *severe wash* in 3 parts conc. HNO_3 + 1 part conc. HF to dissolve a part of the Si was also given, to remove also a substantial part of the gold present mostly at the surface [6]. Sufficient number of counts were obtained

Fig. 2. — The experimentally obtained decay curve of a typical silicon crystal as a function of time.

in crystals of different masses ranging from 0.1 to 0.4 g in order to reduce the statistical error down to 2 %. The radioactive decay was then followed for 2 months (Fig. 2). The half-life time ranges from 13.8 to 19.6 days for the 6 samples where it is measured. Taking into account the statistical error of 2.2 days, one may conclude that there is good agreement with the half-life time for P-32 which is 14.3 days [7]. After a longer time, a small residual beta radioactivity is observed due to the traces of longer lived impurities such as Sb-124. So, in a good approximation, the beta radioactivity measured between $t_2 = 2$ weeks and 2.5 months can be attributed solely to the P-32 atoms produced by transmutation of silicon after the double nuclear reaction.

As the samples are radioactive in the bulk, correction for self-absorption of the beta-rays is to be taken into account. The absorption coefficient for the quite energetic beta-rays emitted by P-32 (1.71 MeV) and absorbed by silicon is measured. Because of the light mass of the silicon atoms, this correction is negligible for the very thin wafers under consideration (300 m).

Figure 2 shows a typical decay curve corresponding to P-32 activity. Equation (9) can thus be used to relate the residual beta-activity to the number of phosphorus atoms. However, the neutron cross-section σ' for the second nuclear reaction is needed. It can be determined from the present results and equation (9) where we use the cross-section for the first reaction $\sigma = 0.102$ barn, as measured for the particular channel in TRITON reactor.

The counter efficiency is monitored for every counting with a standard beta-ray source. The average value is $RC = 0.30$. One finally finds $\sigma' = 0.17$ barn with a standard mean deviation for the different samples of 0.02 barn. This value agrees well with the value of 0.19 ± 0.01 barn [7] obtained from irradiating pure phosphorus.

Therefore, equation (9) can be used to determine the mass m' of transmuted phosphorus atoms per gram of silicon. This is :

$$m' = \frac{A' M'}{36 \sigma' J \times RC} \times \frac{t_1 \exp(\lambda' t_2)}{\left[t_1 - \frac{1}{\lambda'} \{ 1 - \exp(-\lambda' t_1) \} \right]} \quad (10)$$

Expressing as number of carriers, C , per cm^3 of silicon after substituting for M' , σ' and the specific mass of silicon :

$$C = \frac{2.28 \times 10^{23}}{J} \times \frac{t_1 \exp(\lambda' t_2)}{\left[t_1 - \frac{1}{\lambda'} \{ 1 - \exp(-\lambda' t_1) \} \right]} \times \frac{A'}{RC} \quad (11)$$

4. Conclusions. — When silicon is submitted to a thermal neutron flux for the purpose of doping, a second nuclear reaction occurs which produces radioactive phosphorus. We have measured the neutron cross-section for this reaction. We have then derived the equations which offer the following possibilities :

- 1) Equation (9) predicts the residual radioactivity of commercial samples at any time when the irradiation conditions are known.
- 2) Equation (11) relates this residual activity to the number of phosphorus atoms.
- 3) Equation (11) can be used to determine unknown irradiation conditions in commercial samples.

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