

## Preparation and Analysis of Tritium-Niobium Specimen

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I. 14 Preparation and Analysis of Tritium-Niobium Specimen

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I. Introduction

During the course of experiments to investigate the state of hydrogen in niobium (Nb) by an internal friction measurement, isotope effects have been studied using hydrogen and deuterium. To confirm the isotope effects observed in the experiments, the extension of the work to tritium (T) has become necessary. For this purpose, several methods to prepare Nb-T specimens have been tried, of which results will be described in the followings.

II. Experimental Procedure

Nb-N(O) ( $N(O)/Nb=5 \times 10^{-3}$ ) specimens have been prepared by a standard gas doping method. Two different reaction systems have been constructed for the T doping which are shown in fig. 1 (A) and fig. 2 (C). The system (A) consists of parts for ultra-high vacuum systems and set in a draft chamber in CYRIC. 1Ci of  $T_2$  gas was connected to the vacuum system via an UHV leak valve. After evacuating the system, all the valves to pumps were closed and then  $T_2$  gas was introduced in the system to the pressure of  $10^{-3}$  Torr. Subsequently the specimen was warmed up to 700~800°C and gradually cooled down to the room temperature during which T was absorbed to the specimen. After completing the reaction, the valve to the absorber part (activated carbon in a pyrex tube cooled by liquid nitrogen) was opened and the residual  $T_2$  gas was absorbed. Subsequently the absorber part was fused off from the system.

The system (C) is made of pyrex thick-wall tube, of which operational procedure is almost the same with the system A. Because of the larger amount of the  $T_2$ -gas (5Ci) used in the system, the operation was performed at Shinlohi K. K. and only the part of the specimens was sent to the author.

Hereafter, the specimen prepared in the system A (fig. 1) will be called as A and that in the system C (fig. 2) as C.

III. Results

i) Liquid Scintillation Counter (LSC) Measurement

In order to check the  $T_2$  gas release from specimen, it was set in a vinyl tube, of which one end is connected to the inlet and the other end to the outlet of a gas monitor. For two days of the gas monitor operation, no  $T_2$  gas release was detected within the sensitivity of the gas monitor ( $1\mu\text{Ci}/\text{cm}^3$ ). Therefore it is concluded that the Nb-T specimen can be safely handled if the temperature is kept below the room temperature.

Next, about 1 cm of the Nb-T specimen was dipped into a LSC vial and the activity of the specimen was measured. The results are summarized in Table 1. Because of the short range of the  $\beta$ -ray from T, only the activity in the surface

layer of the specimen can be measured by the method.

#### ii) Internal Friction ( $Q^{-1}$ ) Measurements

Figure 3 shows the results of the  $Q^{-1}$  measurement on the present Nb-N-T specimens together with a result on a Nb-N-H specimen. The curve (a) shows the result for the specimen A and the curve (c) for the specimen C. The curve (b) is for a Nb-N-H specimen prepared by an electrolytic method with ( $H_2SO_4+H_2O$ ) solution. A clear  $Q^{-1}$  peak is present at 60K in (a) and (c). The comparison of the results with the only H doped specimen (b) revealed that the 60K peak in (a) and (c) is due to the N-H pair, since they are present at the same temperature with the N-H peak in (b). Although no intentional H doping has been done on the specimen A and C, H contamination has taken place during the specimen preparation or handling procedure. The cause of the H contamination is now being investigated.

For the case of C, a new  $Q^{-1}$  peak is present at 85K, which is concluded to be due to the N-T pair, since the peak is only present in the T doped specimen. For the case of A, where a lower  $T_2$  partial pressure has been applied, a small hump is present between 80K and 90K which probably corresponds to the 85K peak of the N-T pair.

When both H(D,T) and impurities (N,O) are present in Nb, H(D,T) is bound to the impurity forming H(D,T)-N(O) pair. It has been well established that the jump of H around the impurity gives rise to an internal friction peak, of which magnitude is proportional to the pair concentration. For the case of low temperature ( $<100K$ ), almost all of H(D,T) are bound to O(N) and the concentration of H(D,T) in the specimen can be determined by measuring the peak magnitude. Also the temperature of the peak has been revealed to depend on the isotope (O-H(50K), O-D(70K), N-H(60K), N-D(75K)). Therefore, the  $Q^{-1}$  measurement on a specimen enables us to determine the concentrations of H and its isotopes separately even when both H and D(T) are present in the specimen at the same time. This is confirmed by an  $Q^{-1}$  measurement on a specimen doped by both H and D.

Guided by these results on H and D, the T concentration in the specimen C was determined as 0.30 at.% ( $T/Nb=3 \times 10^{-3}$ ) from the magnitude of the N-T peak in fig. 3 C<sup>1)</sup>.

#### IV. Discussion

The bulk T concentration has been determined as 0.30 at.% ( $T/Nb=3 \times 10^{-3}$ ) from the  $Q^{-1}$  measurement for the specimen C. On the other hand, the surface activity of it was determined as  $50 \mu Ci/cm^2$ . Although the range of the  $\beta$  from T is short in Nb, the surface activity must be from the T in the surface layer of a finite thickness  $t$ , of which magnitude should be the order of the  $\beta$ -range. In the followings we will estimate the magnitude of the thickness  $t$  with utilizing the  $Q^{-1}$  and LSC results obtained for the same Nb-N-T specimen.

The number of the T to give the activity may be estimated from the result in Table 1 with the relation  $-dN/dt=\lambda N$  where N is the number of T,  $\lambda$  is the decay constant for T,  $1.07 \times 10^{-7}$  (dpm), and  $dN/dt$  is the radioactivity. The N thus

determined is  $10^{15}$ . On the other hand, the number of Nb atoms in the unit surface layer of the thickness  $t$  is  $5.5 \times 10^{22} t$ . Therefore, the concentration of T in the layer is given as  $10^{15}/5.5 \times 10^{22} t$ . With assuming that the bulk concentration ( $3 \times 10^{-3}$ ) by the  $Q^{-1}$  measurement and that in the surface layer is the same, one can determine the thickness  $t$  as  $7 \times 10^{-6}$  cm or about 0.1  $\mu\text{m}$ . This value is comparable with the measured magnitude of the range of the low energy  $\beta$  in metals. ( $10^{-4}$  gr/cm<sup>2</sup> (=0.1  $\mu\text{m}$  for Nb) for 10 KeV  $\beta$ )<sup>2)</sup>

With using the value of  $t$  and the LSC result in Table 1, the T concentration in the specimen A is determined as about  $2 \times 10^{-4}$ . This concentration is too low to give a definite peak in the sensitivity of the present  $Q^{-1}$  measurement as shown in fig. 3 (a). On the other hand, the radioactivity from such a low concentration of T is quite high as shown in Table 1. This suggests that the LSC measurement provides us a new tool to analyse a quite low concentration of T which is far below the detection limit of the conventional analysing methods of H such as the  $Q^{-1}$  or the resistivity measurement.

#### V. Acknowledgements

The author is deeply indebted to the staff of CYRIC for their helps during the T specimen preparation. Also he is indebted to the staff of CYRIC-Katahira Subcenter for their helps and suggestions during the internal friction measurement and also the LSC measurement. Also he would like to thank to the staff of Shinlohi for performing the T-doping.

#### References

- 1) Hanada R., Script. Met., 15 (1981) 1121.
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Table 1. Results of the LSC and  $Q^{-1}$  measurement on the tritiated Nb-N specimens.

Specimen A: prepared in the system A  
 Specimen C: prepared in the system C  
 Specimen B: only H doped Nb-N specimen

Sp	TRET	weight (mg)	cpm	Sur. Act. ( $\mu\text{Ci}/\text{cm}^2$ )	Conc. (H,T/Nb), $Q^{-1}$	
					H	T
A	T-doped	5.7	$1.99 \times 10^5$	3.2	$2.4 \times 10^{-3}$	Trace
B	H	—	0	0	$2.5 \times 10^{-3}$	0
C	T	5.9	$3.18 \times 10^6$	50	$1.8 \times 10^{-3}$	$3.0 \times 10^{-3}$

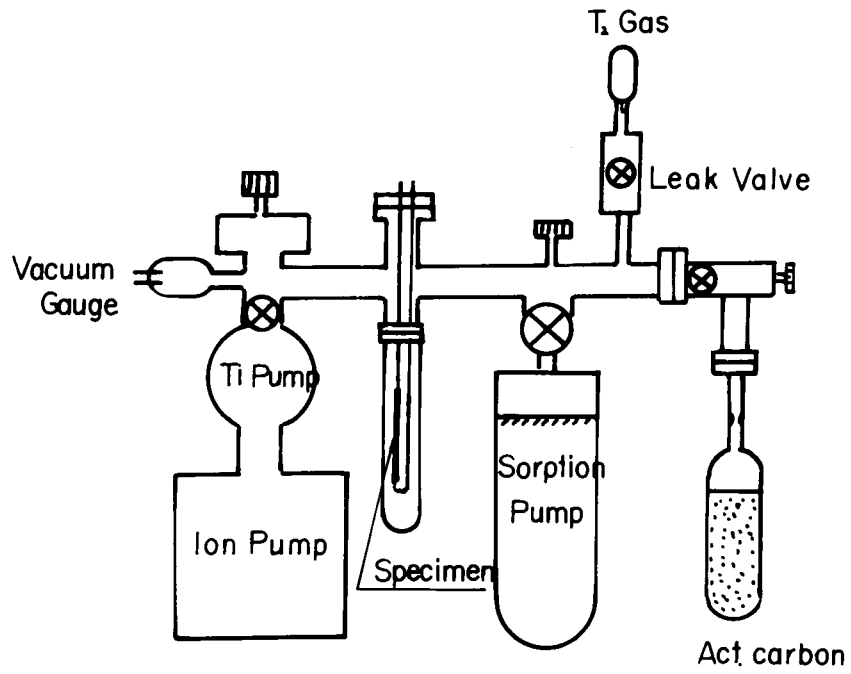


Fig. 1. Reaction system A for T doping.

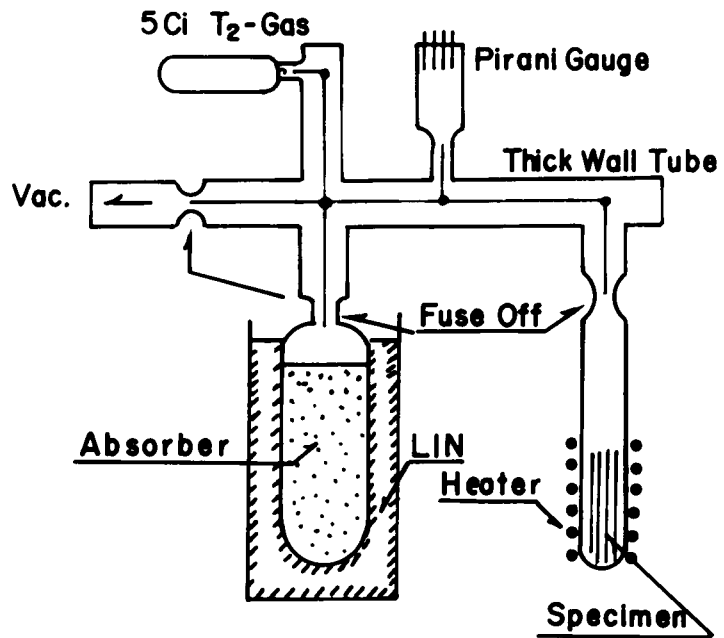


Fig. 2. Reaction system C for T doping.

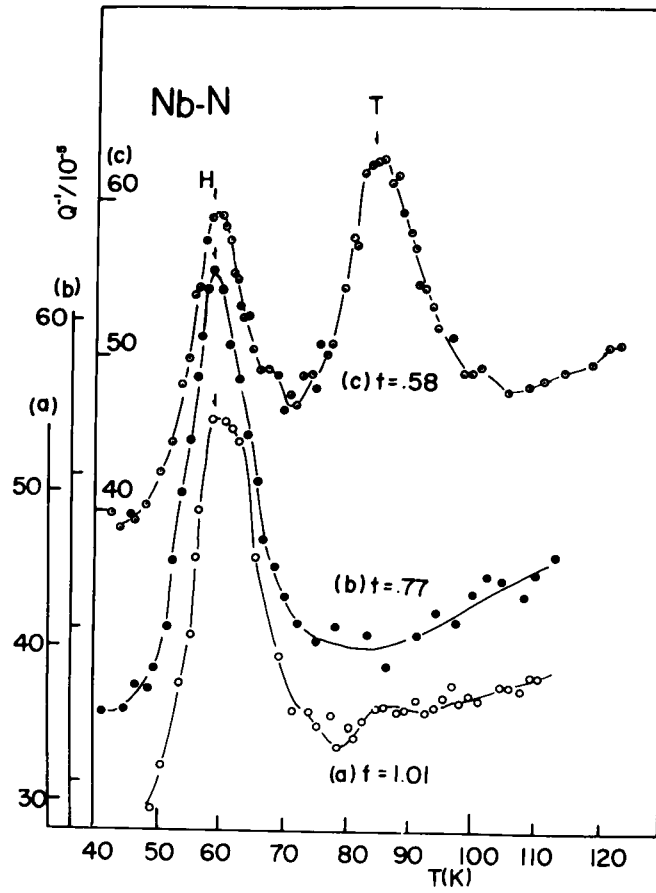


Fig. 3. Results of  $Q^{-1}$  measurements.  
 (a) Nb-N-T specimen prepared in the system A.  
 (b) Nb-N-H specimen (N:0.53 at.%, H:0.25 at.%).  
 (c) Nb-N-T specimen prepared in the system C.  
 f (Hz): measuring frequency.