

《Original》 Preparation of High Specific Activity ^{51}Cr

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Abstract

High specific activity ^{51}Cr is mainly prepared by Szilard-Chalmers process from K_2CrO_4 target. Usually the recoil atom, $\text{Cr}^*(\text{III})$, is coprecipitated with $\text{Fe}(\text{III})$ as a scavenger to be separated from K_2CrO_4 . A new preparation method has been developed, by adding 0.1N NaOH and $\text{C}_2\text{H}_5\text{OH}$ to the irradiated target solution, to precipitate $\text{Cr}^*(\text{III})$ without any scavenger such as $\text{Fe}(\text{III})$. The new method gives the product of higher specific activity and better yield than that of other methods, in the shorter processing time.

This method is compared with the conventional method and the French method, and following results are obtained: the new method gives specific activity more than twice that of the conventional method and better yield than the conventional method; the French method and the new method give similar specific activity, but yield of the new method is almost twice that of the French method.

요 약

높은 비방사능의 ^{51}Cr 은 주로 K_2CrO_4 를 표적으로 하여 Szilard-Chalmers process에 의해 제조되고 있다. 종래에는 그 화학적 분리과정에서 recoil된 $\text{Cr}^*(\text{III})$ 를 담체 $\text{Fe}(\text{III})$ 와 같이 공침시키는 방법을 쓰고 있다.

담체 없이 0.1 N NaOH와 $\text{C}_2\text{H}_5\text{OH}$ 를 사용하여 침전시키는 방법을 씀으로써 그 화학적 조작을 간편하게 하고 소요시간을 단축할 뿐만 아니라 보다 높은 비방사능의 제품을 높은 수율로 얻을 수 있었다. 실제 이 방법을 제래방법 및 프랑스방법과 비교해 보았을 때 다음의 결과를 얻을 수 있었다. 즉 제래방법보다는 훨씬 단시간내에 두 배 이상의 비방사능의 제품을 보다 높은 수율로 얻을 수 있었고, 프랑스방법과의 비교에서는 제품의 비방사능이나 소요시간은 비슷하나 수율은 새 방법 쪽이 2배에 가까웠다.

1. Introduction

^{51}Cr is produced when ^{50}Cr is irradiated with thermal neutrons, as in the following equation.
 $^{50}_{24}\text{Cr}(n, \gamma)^{51}_{24}\text{Cr}$, $\sigma: 17.0$ b., Abundance of ^{50}Cr : 4.31 %

Because ^{51}Cr has a lower energy γ -ray and no

β (as seen in Fig. 1), it is used more favourably than others for diagnostic work.

But, because of chromium's toxicity, the product of a high specific activity is required.

In order to obtain a product of high specific activity, two ways of proceeding ¹⁾ are generally used: by the (n, γ) reaction with enriched ^{50}Cr target and by the (n, γ) reaction followed by

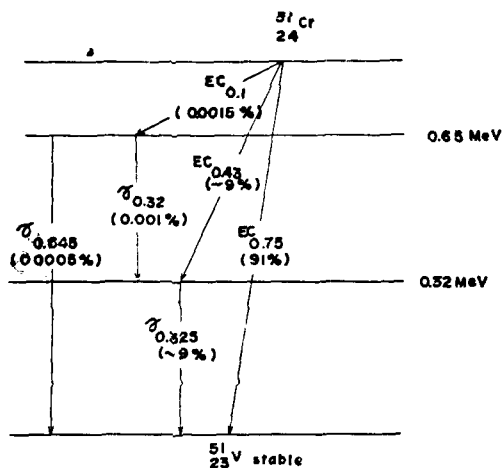


Fig. 1. Decay scheme of ^{51}Cr .

enrichment by the Szilard-Chalmers effects. The latter method is more popular than the former because the enriched ^{50}Cr is too expensive. The principle of the latter method is as follows. When ^{50}Cr is transformed to ^{51}Cr by the thermal neutron-capture, ^{51}Cr is no longer in a hexavalent state as is ^{50}Cr in the target material, but it is in a trivalent state. The difference of oxidation state is used to separate the radioactivated from the nonradioactivated.

The usual method to separate $\text{Cr}^*(\text{III})$ from the chromate is to coprecipitate $\text{Cr}^*(\text{III})$ with $\text{Fe}(\text{III})$ as a scavenger and then the $\text{Fe}(\text{III})$ - $\text{Cr}^*(\text{III})$ mixture precipitate is dissolved in 6 N HCl and $\text{Cr}^*(\text{III})$ is separated from $\text{Fe}(\text{III})$ with the aid of an anion exchange resin, because $\text{Fe}(\text{III})$ is changed to a chloride complex anion and is absorbed in the resin. This method is not only cumbersome and time-consuming but also low in specific activity. The method developed in France which uses a saturated solution of KCl to enhance precipitation of chromium hydroxide is not good in yield.

The new ^{51}Cr preparation method which is simple and gives a good yield by using alkali and $\text{C}_2\text{H}_5\text{OH}$ without a scavenger such as Fe

(III) is developed as follows.

2. Experiment

2-1. Reagents

K_2CrO_4 : E.P., Kanto Chemical Co., Inc.

KCl: E.P., Wako Pure Chemical Industries, Ltd.

$\text{C}_2\text{H}_5\text{OH}$ (99.5 vol. %): E.P., Hayashi Pure Chemical Industries, Ltd.

H_2O_2 (30%): E.P., Kanto Chemical Co., Inc.

2-2. Instruments

2"×2" NaI (Tl) well type scintillation detector P-20-D and Versa Matic V scaler (Tracerlab)

Paper chromatograph

Spectronic 20 spectrophotometer (Bausch & Lomb)

Gammascope II Model 102 100-channel pulse height analyser (Technical measurement Corporation)

2-3. Irradiation method

Target, K_2CrO_4 , is irradiated at the rotary specimen rack (neutron flux = $1.5 \times 10^{12} \text{ n cm}^{-2} \text{ sec}^{-1}$) of the TRIGA MARK II reactor. Each day's irradiation time is as follows:

	Irradiation time (h)
Monday	7.2
Tuesday	7.2
Wednesday	7.2
Thursday	7.2
Friday	0
Saturday	2.2
Sunday	0

There is a slight variation in the above irradiation time. The one week or two week period of irradiation mentioned in this paper means the duration between the time the target is put in the reactor and the time the chemical treatment is begun. The net irradiation time in the case of this paper is written below:

Elapsed time (week)	Net irradiation time (h)
1	26.1
2	49.5
3	92.1
4	123.6

2-4. Preparation method

10 gr of K_2CrO_4 irradiated for two weeks is permitted to stand for three days to reduce the radioactivity of ^{42}K ($t_{1/2}=12.4$ h, 98% decay) which is formed along with ^{51}Cr . The target is dissolved in 30 ml of water. 3 ml of 0.1 N NaOH and 6 ml of $\text{C}_2\text{H}_5\text{OH}$ are added to the solution and it is left to stand for 10 minutes, being stirred occasionally. The solution is then passed into a filtration apparatus fitted with a medium pore sintered-glass filter. Filtration is performed at ordinary pressure. Washing is then carried out with a small quantity of water (about 5 ml) under reduced pressure by an aspirator. This washing process is repeated four times (when yellow coloring remains in the filtrate, the number of washing time is increased.) The precipitate on the fritted disc is redissolved with 5 ml of 3N HCl. This process is repeated three times at ordinary pressure and the residue is washed in the same manner mentioned above. The combined solution is heated to dryness with an infrared lamp to expel HCl. The residue is redissolved in 2 ml of water. $\text{Cr}^*(\text{III})$ is oxidized to Cr^*O_4^- by adding 1 ml of 1 N NaOH and 5 ml of H_2O_2 and it is left to stand for 10 minutes with occasional stirring. The excess peroxide is expelled by heating carefully to dryness under an infrared lamp. After cooling, the solution is adjusted to PH 7–7.4 and centrifuged to separate any foreign material, if it exists.

2-5. Measurement of activity and determination of chromium

The radioactivity is measured by a well type NaI (Tl) crystal scintillation counter of which

the counting efficiency for γ -ray of ^{51}Cr is 0.415.

The ratio of the activity of $\text{Cr}^*(\text{III})$ to that of Cr^*O_4^- in the target and the product are determined by E. Kowalski's paper chromatography²¹ (developing solvent; $\text{H}_2\text{O}:\text{C}_2\text{H}_5\text{OH}:\text{NH}_4\text{OH}=5:2:1$, R_f values: $\text{Cr}^*(\text{III})=0.0$, $\text{Cr}^*\text{O}_4^-=0.9$).

The chromium content is determined colorimetrically by comparing the absorbance at $366\text{ m}\mu$ with the calibration curve.

The radioactive purity is confirmed by γ -spectrometry with the aid of a multichannel pulse height analyser.

Peroxide test of the product is checked by KMnO_4 spot test.

2-6. Investigation of optimum irradiation time

To find the optimum irradiation time that gives maximum specific activity to the product obtained by the new method, the effect of irradiation time to the specific activity of the product was investigated. The followings were also investigated in relation to the irradiation time: variations of total activity formed from 10 gr of K_2CrO_4 ; variations of activity of the product produced by the new method from 10 gr of irradiated K_2CrO_4 ; variations of enrichment factor.

The activity of $\text{Cr}^*(\text{III})$ in the target was determined from the solution of the target before the chemical procedure was carried out. Its variations to the increase of irradiation time were plotted in Fig. 3. The measurement of the specific activity of the product, activity of the original and product, and enrichment factor was carried out uniformly after four days from the time the chemical treatment for separation of $\text{Cr}^*(\text{III})$ was begun. The variations of the above items to the increase of irradiation time was drawn in Fig. 2, 3, and 4, respectively.

2-7. Comparison of each method

30 gr of K_2CrO_4 irradiated for two weeks

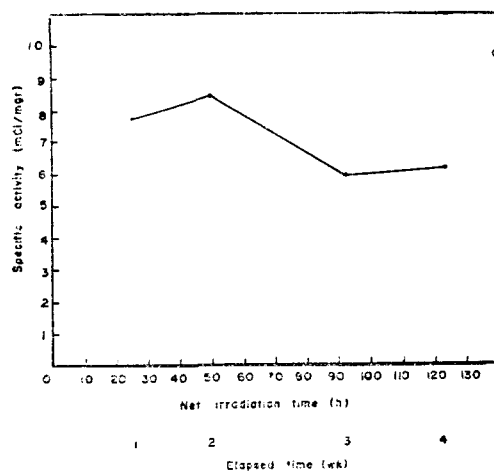


Fig. 2. Specific activity of the product.

was divided into three parts and they were treated, respectively, in three different ways, namely the conventional method, the French method and the new method. Results are given in Table 1.

3. Results and Discussion

Fig. 2 shows that the irradiation for two weeks (as the elapsed time) gives maximum specific activity to the product. The increase of the irradiation time increases the activity of the product, as seen in Fig. 3, but the specific activity decreases after two weeks.

Comparison of the data in the below table leads to the following conclusion:

1) The product made by the new method shows much higher specific activity than that

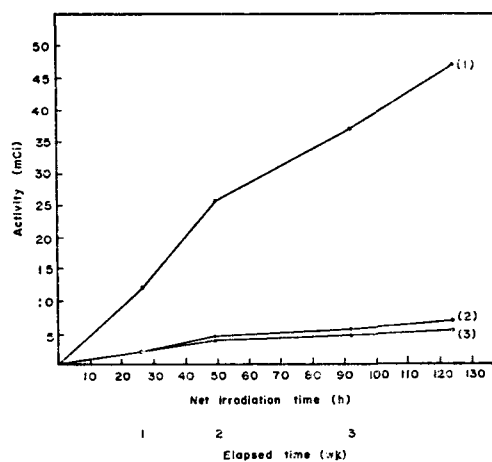


Fig. 3. Activity of the original produced from 10 gr of K_2CrO_4 (1), Activity of $Cr^{*}(III)$ in 10 gr of irradiated K_2CrO_4 (2), Activity of the product obtained from 10 gr of irradiated K_2CrO_4 (3).

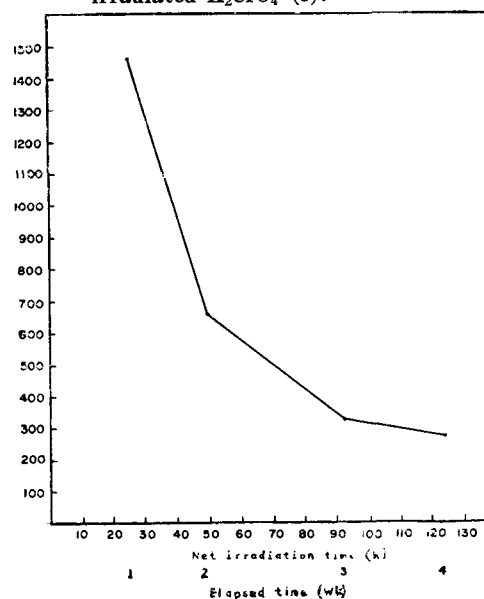


Fig. 4. Enrichment factor.

Table 1. Comparison of each method.

methods	net irradiation time(h)	separable $^{51}Cr(III)$ content (%)	original		product		enrichment factor	yield (%)
			activity obtained (mCi)	specific activity (mCi/g)	activity obtained (mCi)	specific activity (mCi/mg)		
conventional method	50	15.5	25	12.5	2.3	3.75	300	9.2
French method	52.0	15.8	26.4	12.3	2.06	8.53	694	7.78
new method	1+	49.5	25.6	12.8	3.68	8.45	660	14.4
	2+	49.5	25.2	11.7	3.72	8.12	695	14.8

+ Experimental number

made by the conventional method.

2) Both specific activities of the product made by the French method and the new method are similar but yield of the new method is almost twice that of the French method.

3) Processing time required for one batch by the French and the new methods is similar and it is much shorter than that of the con-

ventional method.

References

- 1) Manual of Radioisotope Production, p. 125 (1966), I.A.E.A.
- 2) Kowalski, E., Nucl. Instruments Meth. 33, 29 (1965).