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《**Original**》

The Uptake and Loss of Strontium-90 by the Seaweed *Undaria pinnatifida*

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미역에 의한 스트론튬-90의 濃縮 및 殘留

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Abstract

The uptake and retention of strontium-90 from seawater by the seaweed *Undaria pinnatifida* (sea mustard) varied depending on the plant part, exposure time, salinity, contents of stable strontium and calcium, and presence of chelating agent in the seawater. The concentration factors attained at equilibrium were in the range of 50 and it was evident that the bioaccumulation was largely due to the adsorption of the radionuclide on the surface of seaweed.

요 약

해조류인 미역(*Undaria pinnatifida*)에 있어서 海水로부터 Sr-90의 濃縮 및 殘留현상을 실험실적 조건하에서 실험한 바 다음과 같은 결과를 얻었다.

해수로 부터 Sr-90의 濃縮係數는 식물체의 部位, 노출시간, 해수의 鹽度, 안정원소인 Ca, Sr의 농도에 따라 달리 나타났다. 해조체내에 한번 흡수된 Sr-90은 깨끗한 해수에 대한 노출시간, Ca의 농도와 킬레이트제의 존재여부에 따라 다시 流出되는 패턴이 달리 나타났다. 해조에 의한 Sr-90의 흡수는 表面吸着에 크게 의존하는 것으로 판단되었다.

1. Introduction

The operation of nuclear power plants inevitably causes the release of radionuclides into the marine environment. Radionuclides released into the marine environment are concentrated to varying degrees by marine organisms and transferred to man by means of marine food. In order to evaluate the potential radiation hazard to man and to ensure the safety of nuclear facilities, more information on the contaminat-

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ion of marine environment by radionuclides is required. The term concentration factor is frequently used to evaluate the contamination level of aquatic organisms and to predict the route and rate of radionuclide transfer from seawater to man. The available data on concentration factors based on uptake experiments with radionuclides were compiled by several authors[1~2].

Macrophytes, i.e. benthic macroscopic algae dominate as a primary producer in many shallow coastal areas and radionuclides taken up by macrophytes can be transferred directly to man as food. Several workers have studied the uptake and loss of radionuclides by marine benthic macrophytes in laboratory[3~5]. Nakahara et al.[6] investigated the accumulation of cobalt-60 by *Undaria pinnatifida*, where they reported the concentration factor of 210. However, only a few papers for *Undaria* have been found so far. The *Undaria*, sea mustard, is one of the most greatly consumed macrophytes in Korea and is cultivated on a large scale in coastal areas.

In this regard, the uptake and loss of strontium-90 by *Undaria* and the effect of some environmental factors on this radioisotope flux were investigated. Strontium-90 has long been recognized as the radionuclide of potential radiation risk to man through contamination of diet.

2. Materials and Methods

The seaweed (*Undaria pinnatifida*) was collected from the seashore of cultivation area located at Gijang, Dongnae-Gun, Gyungnam Province. Seawater collected from the same place was filtered through a membrane filter of 0.45 μm pore-size. The properties and composition of seawater used for this experiment are shown in Table 1.

Radioactive strontium ($^{90}Sr(NO_3)_2$ in 1 M HNO_3, carrier free, Radiochemical Centre, Amersham) was added to filtered seawater to give final concentrations of 0.1, 1 and 10 μCi/L. Solutions containing stable strontium of two or five times higher concentration than natural seawater were prepared by adding $Sr(NO_3)_2$ to seawater. Calcium ion was also added as $CaCl_2$.

Table 1. Properties of Seawater used for Experiment

pH	7.9
Salinity	29%
Calcium ion	400 mg/kg
Strontium ion	13 mg/kg

After the radioactivity of ^{90}Sr solution showed a steady value, blades of *Undaria* (2×15cm size) were introduced into the wide-mouth glass bottle containing 500 or 1000 ml of ^{90}Sr solution. During the uptake experiments, temperature was maintained at 10 ± 1°C and dark condition was maintained to retard the growth of *Undaria*. The medium was renewed several times during the experiments so that the variation of ^{90}Sr concentration did not exceed ± 10% of the average concentration. The radioactivity of the medium was monitored by using 1 ml aliquot and that of the seaweed was measured for samples (100~500 mg) cut from the blades in small portion at proper time intervals.

Following the uptake of ^{90}Sr by *Undaria* blades (2×10 cm) at the concentration of 10 μCi/L for 24 hours, each blade was transferred to a bottle containing 500 ml of filtered nonradioactive seawater. The seawater was exchanged several times during the loss experiment period. The temperature was maintained at 10±1°C. Samples (100~500 mg) of blades were taken at time intervals for the determination of retained radioactivity. To assess the effect of calcium concentration in medium on the loss pattern of ^{90}Sr in blades, the radioactivity was observed on blades transferred to seawater containing calcium ion two or three times higher than natural seawater after uptake of ^{90}Sr as stated above. In the loss experiment with chelating agent, ethylenedinitrilotetraacetic acid tetrasodium salt (EDTA Na_4), two blades (2×12 cm) were introduced into the 1,000 ml medium of ^{90}Sr solution with the concentration of 10 μCi/L. After the incubation period for 24 hours, each blade was transferred into nonradioactive seawater with or without EDTA (2 mM). The medium was shaken manually every ten minutes for one hour. Samples for radioactivity analysis were taken in small portion.

Seaweed samples taken in uptake and loss experiments were rinsed briefly with nonradioactive seawater and weighed after surface moisture was blotted with filter paper. The samples were then digested in 25 ml vials using concentrated HNO_3 and 30% hydrogen peroxide. After digestion, the solutions were dried on hot plate and dissolved again in 5 N HNO_3. One ml aliquot of the dissolved solution was poured into an aluminium planchet and dried under I.R. lamp. The radioactivity was measured by using a G.M. Counter (Aloka G.M. tube, window thickness 1.5 mg/cm^3, operating voltage 1,100 V). Counts per minute (cpm) were corrected for background.

3. Results

The concentration factors of radionuclide were calculated on a wet weight basis according to the following equation,

$$\text{Concentration factor} = \frac{\text{cpm/g of tissue sample}}{\text{cpm/g of medium}}$$

Fig. 1 shows the distribution pattern of ^{90}Sr in a single blade of *Undaria*. A higher concentration factor was found at the top portion of the blade and similar concentration factors at the rest portion. Therefore, a median portion of the blade was used in the following experiments.

The uptake pattern of ^{90}Sr by *Undaria* blade is shown in Fig. 2. Rapid uptake in the initial stage was followed by slow uptake, which ten-

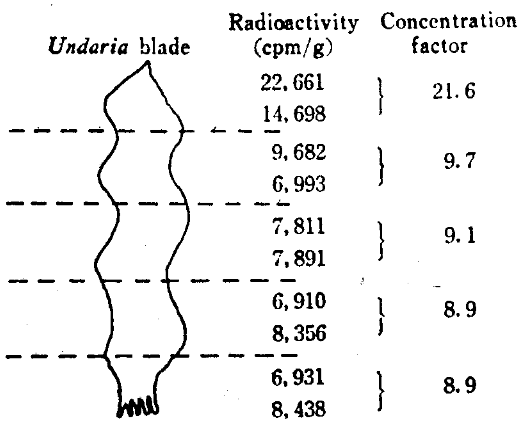

Fig. 1. Radioactivities and Concentration Factors in Various Parts of *Undaria* Blade after 24 Hour Uptake Period in ^{90}Sr Medium (1μCi/L)

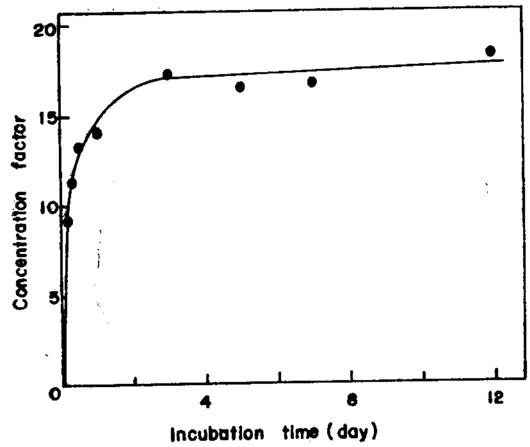

Fig. 2. Uptake of ^{90}Sr by *Undaria* Blade

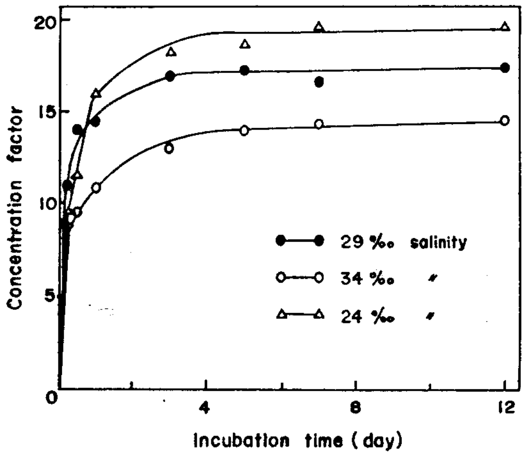

Fig. 3. Effect of Salinity on the Uptake of ^{90}Sr by *Undaria* Blade

Table 2. Radioactivities and Concentration Factors of ^{90}Sr in *Undaria* Blade at Different Stable Sr Concentrations

Sr^{++} in medium (mg/kg)	After 24 hrs			After 3 days		
	Tissue activity (cpm/g)	Medium activity (cpm/ml)	Concentration factor	Tissue activity (cpm/g)	Medium activity (cpm/ml)	Concentration factor
13	14,724	744	19.8	37,238	741	50.3
23	12,079	753	16.0	33,934	736	46.1
65	10,365	756	13.7	24,956	730	34.2

Table 3. Radioactivities and Concentration Factors of ^{90}Sr in *Undaria* Blade at Different Ca^{++} Concentrations

Ca^{++} in medium (mg/kg)	After 24 hrs			After 3 days		
	Tissue activity (cpm/g)	Medium activity (cpm/ml)	Concentration factor	Tissue activity (cpm/g)	Medium activity (cpm/ml)	Concentration factor
400	16,335	746	21.9	54,703	743	73.6
800	9,240	732	12.6	25,801	731	35.3
1,200	6,836	740	9.2	17,291	730	23.7

ded to reach an equilibrium state after about 3 days. Fig. 3 shows the effect of salinity on the uptake pattern of ^{90}Sr by *Undaria* blade. At an equilibrium state the concentration factor decreased with an increase of salinity in the medium. The effect of stable Sr on the uptake of ^{90}Sr by blades is shown in Table 2, where the uptake of ^{90}Sr tended to decrease with an increase of stable Sr ion concentration in medium. After 24 hours uptake the concentration factors in medium containing two or five times higher concentration of stable Sr than natural seawater were decreased to 81 or 70% of control, respectively. The effect of calcium ion on the uptake of ^{90}Sr was also similar to stable Sr shown in Table 3. The uptake of ^{90}Sr by blades in medium containing two or five times higher concentration of Ca ion than natural seawater showed that the concentration factors were decreased to 58 or 42% in 24 hours uptake and decreased to 48 or 32% in 3 days uptake, respectively.

The retention of ^{90}Sr in blade of *Undaria* was investigated on samples transferred to non-radioactive seawater after 24 hours uptake from seawater at a concentration of 10 μCi/L of radioactive Sr. Fig. 4 shows the loss of radioactivity from *Undaria* blade, where rapid loss of ^{90}Sr during the first 3 days was followed by slow decrease of radioactivity. About 90% of radioactivity was lost from blades on the first day. Loss of ^{90}Sr was slightly stimulated by increase of Ca ion in the medium as shown in Fig. 5. Fig. 6 shows the effect of EDTA, which is known to be a typical chelating agent, on

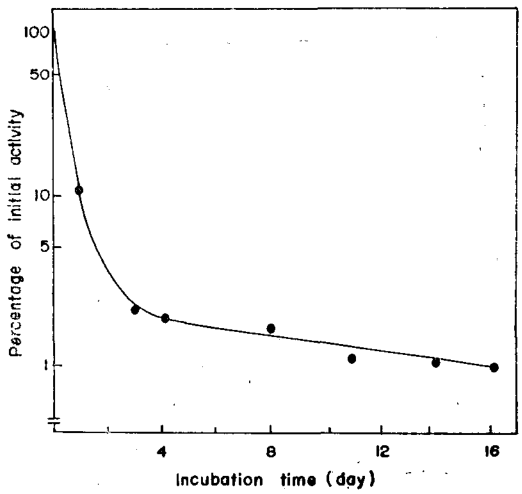

Fig. 4. Loss of ^{90}Sr from *Undaria* Blade

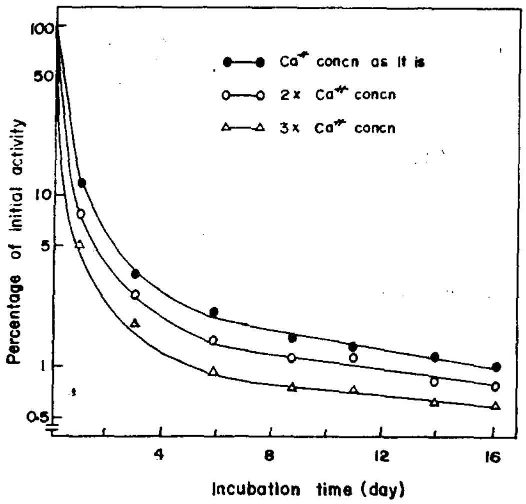

Fig. 5. Effect of Ca Concentration in Seawater on the Loss of ^{90}Sr from *Undaria* Blade

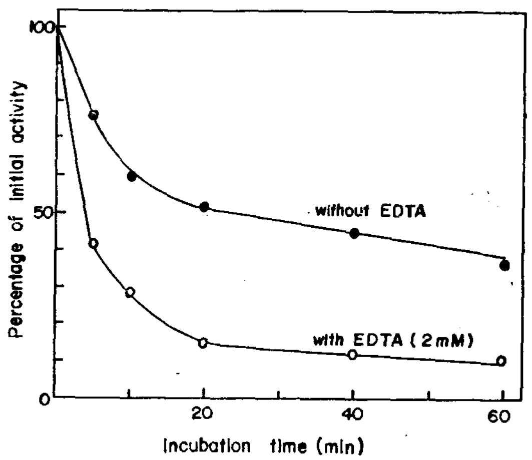

Fig. 6. Effect of EDTA on the Desorption of ^{90}Sr from *Undaria* Blade

the loss of ^{90}Sr. Addition of EDTA (2 mM) to medium after the blade was contaminated with ^{90}Sr enhanced the desorption of ^{90}Sr three times faster than occurred in medium without the chelating agent.

4. Discussion

The accumulation of radionuclides by benthic algae is known to vary according to the radionuclide and algal species[7]. As shown in Fig. 4, the higher concentration factor of top portion in the blade as compared with the rest portion seems to be related to metabolic activity of the seaweed. Environmental factors such as salinity, pH, and temperature might influence the metabolic activity and consequently the isotope flux. In general, the addition of stable carrier can be expected to reduce the accumulation of the corresponding radioisotope. Miyama et al.[8] reported that increasing the concentration of stable I, Fe and Ru resulted in the lowering of concentration factors of the corresponding radioisotopes in *Ulva*, *Enteromorpha* and *Codium*. The addition of stable iodine has also been found to cause a decrease in the ^{131}I uptake rate in *Laminaria*[9]. In addition, increasing amounts of stable Y slightly reduced the concentration factors of ^{91}Y in *Porphyra*[5].

In addition to stable carrier, elements which are chemically similar to the radioisotopes of interest can also reduce the accumulation of corresponding radioisotopes. For example, Scott[10] found that *Fucus* took up more cesium in potassium deficient seawater than in natural seawater. Likewise, it was reported that ^{90}Sr uptake can be affected by the substitution element Ca[11]. The results presented in Tables 2 and 3 show that an increase in stable Sr or substitution element Ca decreased significantly the uptake of ^{90}Sr, which was consistent with the results stated above. However, the decrease of ^{90}Sr uptake by increase of Ca ion concentration might be partly due to the change in salinity because the increase in salinity resulted in decrease of ^{90}Sr uptake as shown in Fig. 3.

Extracellular materials from benthic algae can also influence the isotope uptake. In Fig. 2 about 80% of ^{90}Sr uptake at equilibrium state occurred in the initial 24 hours, which seems to be due to adsorption by extracellular substances on surface. This was also demonstrated in the remarkable loss of ^{90}Sr in the presence of EDTA in the medium as shown in Fig. 6.

Therefore, it is likely that surface adsorption plays an important role in the uptake of ^{90}Sr by *Undaria*. From the result of retention experiment as shown in Fig. 4, the loss of ^{90}Sr after uptake of the radionuclide by *Undaria* blade can be described by two-exponential model having two compartments with different rates. The short-lived component might be due to the elimination of a large fraction of the radioactivity initially adsorbed on the surface layer, while the long-lived component could be accounted for by a fairly slow elimination of the radioactivity accumulated in the inner portion of *Undaria*.

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