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学位論文題目	福島第一原子力発電所事故由来の放射性セシウムにおける形態と特性について
審査会	委員長　関東　康祐　　庄村　康人　　田中　伊知朗　　栗原　和男

論文内容の要旨

Great East Japan Earthquake occurred on March 11, 2011. Theearthquake caused atsunami, which has lost all the electric power sources of the reactor facilities (Units 1 to 5) inthe Fukushima Daiichi Nuclear Power Plant.

Since the function of the reactor cooling system was lost due to the loss of all the electric power sources, it became impossible to remove the decay heat of fission products generated in the reactor, and the fuel melted. These have caused alarge-scale core damage.

Furthermore, a large amount of hydrogen generated by the water-Zircaloy reaction in the fuel cladding and coolant has caused a hydrogen explosion,the damage ofthe reactor facilities, and the release of a large amount of radioactive materials, which have finally contaminated a wide area of northern Japan.

As a result of variousinvestigation, it was found that the radioactive cesium (cesium-137)madethe largest impact on the environment becauseithasa large amount of quantityin fission products,and emits gamma rays with high penetrating power, and it has relatively high radiation energy (662KeV), and a long half -lifeof about 30 years.

After that, many studies related toradioactive cesium and its decontamination including physical and chemical properties have beencarried out, and it has been consideredthat the emittedradioactive cesium was adsorbed on soil and other minerals,such as micain the form of ions.

On the other hand, some of the emittedradioactive cesium areaerosoland existas water-insolublematerials, and the radioactive cesium is incorporated into microparticle containing inorganic substances such as iron and silicon. In additionit is reported that the microparticles areamorphous.The microparticles have also been found in soil, and it has been reported that the radioactive cesium in the microparticlesis hardly incorporated into plants. However, it has been found that some plants have taken in avery smallamount of radioactive cesium.Then, how is the radioactive cesium taken up by plants?

The purpose of our investigationis to knowthe physicochemical properties of the radioactive cesium in the microparticlessuch as dissolution rates, mechanical strength, and thermal stability using litterbut not soil.The litter was collected in a bamboo forest located in Nihonmatsu in the Fukushima prefecture. The forest is approximately 45 km west-northwest from Fukushima Daiichi Nuclear Power plant.The reason for using the microparticles in the litter is that theyexist in the untouched state, that is, in a pure stateandthe main components of litters are dead branches and leaves.

The dissolution experiments were carried out, firstlyusing the raw littersamplewith a dialysis membrane as a filter, and secondlyusingthe charcoal litter samplewith a glass filter. The charcoal litter was made by putting raw litterin a heat-resistant container, heated in an electric furnace (about 300℃).In the dissolution experiment, the aqueous solution and the raw litter or charcoal litter were separated using a dialysis membrane or a glass filter, and the radioactivity of the filtrate was measured byaBequerelmonitor. After the radioactivity measurement, the filter, the residueand the filtrate solution were returned to the same container and after a certain period of time, the same operation was performed several timesto measure the time dependenceof the radioactivity ofthe filtrate.
We have assumed that the dissolution rate of the microparticle on the litter depends on the surface area.In order to increase the surface area ofthe microparticles, the charcoal litter was crushed using a ball mill. The dissolution rate ofthe crushed litter sample was measured in the same method and as a resultwe have found that the dissolution rate of the crushed sample hasincreased.
From the dissolution experiment using the dialysis membrane, it has been found that the radioactive cesium in the microparticlesdissolvesas a soluble radioactive cesium ion or a hydrated cesium ion intowater. This indicates thatthe radioactive cesium existing on the surfaceof the micropaticlesis easily dissolved when it comes into contact with water.Moreover, the fact thatthe dissolution ratesof themicroparticles in theraw litter and the charcoal litter were the same,has shownthat not only the surface but also the internal structureof the microparticlesdid not change. It also shows thati)the microparticlescontaining radioactive cesium have thermal stability at about 290 ° C, and ii)the dissolution rate of the sample after crushing with a ball mill was about 10 times higher than that of the other samples, that is, the average radiusof the microparticleswas about 1/10.Thesefactshaveshown not only to simulate the future contamination, but also to show that microparticlescontaining radioactive cesium are crushed by an external force such as a ball mill, and this property may contributeto the development of a method for decontaminating radioactive cesium.
We wish that our study should contribute the further development of the new decontamination method of Cesiumin future.
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論文審査の結果の要旨

本論文では、東日本大震災に起因する津波によって発生した、東京電力福島第一原子力発電所事故由来の放射性Cs (^{134}Cs及び^{137}Cs、以下 Cs*) の放射能汚染の原因であるCs*の存在様式を溶解性とその物性に関する研究を行っている。初期段階では、上記のCs*が土壌に吸着されたと考えられていたが、その後の先行研究で、ほとんどのCs*の周りを多数の金属元素が取り囲んでいるアモルファス状態の、顆粒状の粒子も存在することが示唆された。

本研究では、顆粒状放射性Cs*の詳細な性質等を明らかにすることを目的として、落ち葉を実験試料として用いて、上記の顆粒状放射性Cs*がどのように可溶化するのかの実験を、試料炭化前後のイメージングプレートを用いたオートラジオグラフィーによる放射性物質の分布や簡易ベクレル計による放射性物質の濃度測定とともに行った。また、走査型電子顕微鏡を用いたエネルギー分散型X線分析により、落ち葉に付着したCs*を含む顆粒状の粒子における元素分析も行った。さらに、顆粒状放射性Cs*の機械的特性を調べるために、ボールミルによる粉砕を行うことで、非粉砕試料との溶解特性の違いを見出し、水へ可溶化する割合を定量・比較したとともに、顆粒に含まれるCs*は300℃程度の温度には安定なことも考察した。この結果、風化や^{137}Csの崩壊等で顆粒状放射性Cs*が壊れると、表面積が増加し、溶解しやすくなることで、植物に取り込まれやすくなることが分かったとともに、逆に、人工的に顆粒状粒子を壊してやるとCs*の溶出を促進し除染につながる可能性も示唆された。

本学位論文では、研究の目的と当該分野での位置づけ、研究方法、結果が詳細かつ明確に記載されており、考察も論理的である。新規性、有用性のある独創的な内容であり、参考文献の引用も適切である。また、本学位論文の基礎となる原著論文1報を第一著者として査読付学術雑誌に発表済である。

以上を総合して，本博士論文は審査会全員の一致で、合格と判定した。