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論　　文　　名　　Development of Real-time Visual Observation and Spectrophotometric Measurement System for High-dose Gamma-ray Irradiation Effect
（高線量ガンマ線照射効果に対するリアルタイム観察及び分光分析システムの開発）

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論文要旨

Radiation irradiation effects are used to alter material microstructures, compositions, properties, and functions; thereby, material performance can be degraded or enhanced. Understanding the evolution of materials under the radiation-induced process is important in the viewpoint of irradiation effect study. Post-irradiation examination is typically conducted to examine the impact of irradiation on materials, using a wide range of property testing platforms, from microscopes to spectroscopies. However, the post-irradiation examination is not ideal for understanding the dynamic evolution of materials exposed to radiation as it only offers two examination conditions: before and after irradiation. It is difficult to pinpoint the timing of the property change in irradiated specimens, which tends to progress rapidly. Furthermore, the conventional method requires considerable specimen preparation, the extraction of specimens from the irradiation site, and testing in offside devices. Such a process can be time-consuming, material-wastage, expensive, and may involve a fading/time delay effect. Therefore, a practical system that can measure the instantaneous effects would be advantageous. Elucidating the relationships between irradiation dose and property change in materials can be used to enhance our fundamental understanding of radiation effects to materials at intense dose and thus it leads to better design engineered materials.

In general, a heavy irradiation area will not be accessible during an experiment period. Therefore, a new alignment system using remote-controlled measurement has to be developed. Nevertheless, most electronic/optical devices are known to degrade over time with the effects of radiation, especially when such an operation takes place under a high dose condition. It is worthwhile to build a system that could withstand harsh radiation and could be used to observe irradiation effects under in-situ, dynamic conditions.

The heavy gamma-ray irradiation facilities of Osaka Prefecture University (OPU) is currently exploited for the study of irradiation effects in real time. The objective of this study is to capture full information about the interaction between the high-dose gamma rays and materials in situ. This thesis involved with the development of a system that has abilities for:

- dose monitoring at multiple positions simultaneously in intense field,
- spectrophotometric and colorimetric measurement, and
- direct visual observation of the irradiation effect during gamma-ray irradiation.

Chapter 1 introduced the irradiation effect and its importance in the revolutionary of science and technology towards the understanding and synthesis of new materials. Some typical characterization tools for optical property of material in wavelengths of visible light were presented. The disadvantages of the conventional approach were addressed and the necessity for real-time measurement was determined. The objectives and outline of the thesis were established.

Chapter 2 presented the development of a new gamma-ray sensor using a cadmium sulfide (CdS) photoresistor in combination with a small-sized cesium iodide doped thallium scintillator. The main characteristics of the sensor were experimentally clarified, and its performance was validated via Monte Carlo simulation. Gamma-ray exposure experiment was performed using a 591 TBq ^{60}Co source. The exposure rate was simultaneously measured with a small-sized ion chamber and was compared to the resistance of the sensor for calibration. The sensor exhibited radiation-resistant ability in an intense gamma-ray field in the order of several thousand gray, demonstrating a linear response with wide dose range. The sensor was able to respond well to dose rate change of a few Gy/h. A high spatial resolution was achieved owing to the compact size of the sensor afforded by the small area of the CdS photoresistor, as well as the flexible adjustment of the scintillator’s crystal size. The CdS photoresistor has a high current gain; and the sensor’s built-in structure and the detection setup were simple. Multiple sensors can be installed for multi-channel data acquisition simultaneously. The fabricated sensor has a number of advantages: compactness, robustness, inexpensiveness, high sensitivity, and high spatial resolution.

Chapter 3 described the construction of the real-time measurement system for emission and absorption of visible light at the irradiation facility of OPU with the intense ^{60}Co source. The radiation-induced degradation in performance of measurement devices was examined at primary step. The attenuation of light transmission through an acrylic optical fiber was addressed. The high noise level significantly influenced the photographic performance of a commercial digital camera. Next, radiation shielding configuration design was evaluated using Monte Carlo simulation. Absorbed dose of such instruments decreased considerably with the increase of the shielding thickness and/or the depth position of the devices inside the shielding tunnel. Experimentally, the optical fiber transmission technique was used to extract information about the irradiation effect from the intense radiation field successfully. A part of a long quartz glass optical fiber cable was inside the irradiation room, and the end of it was connected to an ultraviolet-visible spectrophotometer located outside the room for optical absorption measurement. A compact white light-emitting diode was used as a light source that offers two advantages, including the simplification of the measurement system and robustness against exposure. It produced a visible light beam that passed through an irradiated specimen and was measured by the optical fiber probe. The observation system constructed in this study, including a video camera, a zoom lens, a raspberry pi device, and the mirror, was successfully protected and afforded the direct visual observation of phenomena and simultaneous comparison between several specimens. The feasibility of the system for operating in the high dose gamma-ray field was validated experimentally with the source's radioactivity in the order of few hundreds terabecquerel in our experiment range. The stability of the system was achieved with the dose rate of few thousands gray per hour at irradiated specimen; and the consistency of the system in couple of hours of operation was confirmed. The combination of direct visual observation and optical absorption measurement is expected to provide powerful clues for elucidating various phenomena caused by irradiation effects.

Chapter 4 showed the results of the gamma-ray irradiation effects analysis (photographic, spectrophotometric, and colorimetric) with the developed system. Test specimens were subjected for gamma irradiation and their changes in optical absorption and color state were analyzed on the basis of the real-time measurement. The system allows the mechanisms during the dynamic radiation-material interaction to be deciphered at a dose fidelity, with minimal need for specimens. The successfully radiolytic synthesis of silver nanoparticles using silver metallic powder as an alternative to silver salts was newly reported. In order to study the formation of silver nanoparticles in detail, blue color dye was added to the radiolytic medium and its color change was examined as a function of irradiation dose to observe the interaction among radiation-introduced radicals in water, generated silver nanoparticles, and dye molecules. The formation/evolution of silver nanoparticles and the deformation of blue dye, which were interactive in a joint radiolytic medium, were characterized. These results relate to the application of gamma-ray irradiation to material modification and the development of a new dosimeter using color dye. The real-time measurement system has various advantages that are competitive with the conventional method: cost-effectiveness (minimal specimens), minimal time consumption, and the provision of timely and precise information. As a further step, PIXE (Particle-induced X-ray emission) analysis by low energy proton beam was proposed to examine the absolute concentration of synthesized silver nanoparticles. The silver nanoparticles yielded from gamma radiolysis process using powder precursor is high purity, and has a concentration in the order of few ten μg/mL in our experiment range.

Chapter 5 summarized the results of previous chapters and conveyed the conclusion of the thesis. The ability to perform characterization in situ offers a critical strategy to fully track the radiation-induced process. The technique is able to capture the transient phenomena which tend to happen suddenly and progress quickly. This will uncover information that has not been accessible by traditional characterization techniques and is used to identify the modes and kinetics of the evolution of materials/reactants in extreme radiation environments. They also provide a protocol to precisely control and optimize synthesis parameters. Integrating these techniques within irradiation facilities can enhance our understanding of the dynamics of radiation-material interactions to advance material design, and furthermore, the potential use of our facilities in research can be exploited thoroughly. The developed system could provide complementary capabilities to a wide range of facilities where irradiation effects during radiation exposure are studied.

審査結果の要旨

本論文は、高線量ガンマ線場における材料への照射効果を視覚的、分光学的にリアルタイムで観察、測定するシステムの開発に関する研究をまとめたものであり以下の成果を得ている。

(1) 高線量ガンマ線による照射効果を評価する際に照射試料と併置するガンマ線検出器は小型であることが求められるため、ヨウ化セシウムシンチレーターと硫化亜鉛フォトセンサーを組み合わせ、狭い空間で使える小型のガンマ線センサーを新たに開発した。本センサーが数 mGy/h～数 kGy/h に及ぶ広い線量範囲の測定に対応し、高い耐放射線性を有する特性を実験的に明らかにした。また、コバルト 60 線源の周辺空間の線量分布測定などの実験結果より、本センサーが高線量場における線量測定に極めて有用であることを示した。

(2) 照射効果のリアルタイム観察において、ビデオカメラや分光測定のための光ファイバーなど複数の機器に対する放射線による性能劣化が問題となる。そこで機器への影響を低減するために線源からのガンマ線の挙動をモンテカルロシミュレーションにて計算し、線源に対する測定機器や遮へい材の配置について最適化を行った。耐放射線性に劣るビデオカメラの CCD 素子を保護しつつ鮮明な画像を得るために鏡の反射を用いてノイズ影響を低減させながら画像を得ることに成功した。

(3)　開発された測定システムを用いてガンマ線照射下での銀微粒子水溶液からの銀ナノ粒子生成の様子をリアルタイムに観察し、同時に UV-VIS 分光スペクトルの測定を行う成果が得られた。取得したビデオ画像を RGB に色分解して照射効果を解析した。銀ナノ粒子生成では母材として銀金属微粒子を用い、界面活性剤と混合することでガンマ線照射下において直接的に微粒子から銀ナノ粒子が生成することを新たに示した。青色色素との混合試料の照射では、放射線によって誘起された活性ラジカルによる色素分解反応との競合により、銀ナノ粒子の生成や成長が複雑化する様子を色変化の直接観察や分光スペクトルの変化より明らかにした。

以上の諸成果は､高線量ガンマ線場における放射線照射効果の新たな科学的知見や応用技術を得ることに大きく貢献する。また、申請者が自立して研究活動を行うのに必要な能力と学識を有することを証したものである。学位論文審査委員会は､本論文の審査ならびに最終試験の結果から、博士（工学）の学位を授与することを適当と認める。