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Synthesis of uranium-di-oxide nanoparticles by pulsed laser ablation in ethanol and their characterisation

Aniruddha Kumar [a,*], Manisha Prasad [a], Shailini Shail [a], Chetan Baghra [a], B.N. Mohanty [a], R.B. Bhatt [a], P.G. Behere [a], Mohd. Afzal [a], Arun Kumar [a], Suman Neogy [b], Rajib Kar [c], D.J. Biswas [c]

[a] Advanced Fuel Fabrication Facility, Bhabha Atomic Research Centre, Tarapur, Maharashtra, 401504, India
[b] Material Science Division, Bhabha Atomic Research Centre, Mumbai, 400085, India
[c] Laser & Plasma Technology Division, Bhabha Atomic Research Centre, Mumbai, 400085, India

HIGHLIGHTS

- UO_2 nano-particles were generated by Pulsed laser ablation in ethanol.
- Particle size was found to be dependent on laser fluence values.
- The phase of the particles was found to be FCC.
- The particles showed higher absorption in UV wave length.

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ABSTRACT

The importance of actinide based nano-structures is well known in the area of biology, nuclear medicine and nuclear industry. Pulsed laser ablation in liquid is recognised as an attractive technique for production of nano-structures of different metals and metal oxides with high purity. In this paper, we report synthesis of uranium-di-oxide nanoparticles by pulsed laser ablation in ethanol. The second harmonic emission of an electro-optically Q-switched nano-second Nd-YAG laser was used as the coherent source here. The structural and optical properties of the fabricated uranium-di-oxide nanoparticles were investigated by X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), energy dispersive analysis of X-rays (EDX) and UV–Vis–NIR spectrophotometry. The mean size of the particles was found to be dependent on the laser ablation parameters in particular with laser fluence. XRD and TEM analysis confirmed the phase of the synthesised material as pure crystalline uranium-di-oxide with face centred cubic structure. UV–Vis–NIR absorption spectra of the colloidal solution revealed high absorption in the UV regime.

1. Introduction

Considerable efforts have been expended of late towards synthesis and characterisation of nanoparticles and structures. Development of different techniques for synthesis of nano scale materials received a growing interest due to the unique properties of the nano-materials, that are widely different from the bulk, have left indelible mark in almost all branches of science including engineering and biology. This has led to the emergence of a variety of techniques for the synthesis of nano-materials with controlled size, shape and composition. There now exists numerous reports on the synthesis of nano-structures of varying morphologies in case of pure metals [1–5], alloys [6,7], metal oxides [8–11], carbides [12–14] and a variety of other compounds [15–17]. The actinides on the other hand are generally radio-toxic and understandably therefore, literature on synthesis of nano-structures of these class of elements is scanty. The importance of actinide based nano-structures is well known in the area of biology [18], nuclear medicine [19] and nuclear industry [20]. Actinide nanoparticles are expected to play a significant role towards fabrication of advanced nuclear fuels in the future [21]. Engineered nanoparticles of different actinide materials with varying sizes can also be used as models for studying migration of radio nuclides in atmosphere in the event

* Corresponding author.
E-mail address: nontee65@yahoo.com (A. Kumar).

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of any nuclear emergency [22]. Uranium-di-oxide nanoparticles are also known to catalyse a number of chemical reactions such as gas phase destruction of volatile organic compounds [23] or selective reduction of NO at moderate temperature [24]. Cao et al. [25] reported synthesis of nanoparticles of uranium-di-oxide by thermally decomposing uranyl acetyl acetonate-a compound comprising uranium ions and organic ligands. This study revealed that the size of the nanoparticles could be controlled by changing the concentration of the organic additives in the reaction. Roth et al. [26] reported on the synthesis of uranium-di-oxide nanoparticles by irradiating U(VI) solutions with both electrons and γ radiation. Irradiation with electron was found to be more advantageous as the yield of nanoparticles with a narrow size distribution is higher. Hasan et al. [27] reported a method for preparation of uranium-di-oxide nanoparticles using a surfactant templating crystal growth technique. The size and shape of the particles could be controlled by selecting appropriate surfactant micelles. Wang et al. [28] reported a facile hydrothermal route to phase controlled synthesis of uranium-di-oxide nano-structures using organic amines as reducing reagents. We note here the recent work of Nenoff et al. reporting the room temperature synthesis of UO_2 nanoparticles, uranium metal and uranium alloy nanoparticles by gamma ray irradiation of aqueous uranyl nitrate solution [29,30]. Their experimental result suggested a promising method towards reuse of dissolved actinides from spent nuclear fuels. Chemical synthesis always involves handling of aggressive chemicals and generated nanoparticles may therefore retain residues of chemical precursors. Laser ablation of solid targets in a liquid medium has been recognised as an attractive technique for production of nano-structures with varying compositions and morphologies [14,31–34]. Further modification of the size and shape can be achieved by subjecting post laser treatment to the suspended nanoparticles [35]. This technique of production of nano-structures is simple and "green" which normally operates in water or organic liquids under ambient conditions without any by-products. In PLAL (Pulsed Laser Ablation in Liquid) process a high power laser beam is focussed on to the surface of a solid target that is immersed in a liquid. The interaction of the laser pulse with the target causes the material to vapourise and, in turn, a plasma plume from the solid target is generated on the interface between the solid target and the confining liquid. The laser induced plasma adiabatically expands at a supersonic velocity to create a shock wave under the confinement of the liquid. The shock wave thus generated pushes the laser induced plasma and transforms it into a thermodynamic state of higher temperature, pressure and density. In this state, different chemical reactions between the species from the target and molecules of the liquid could take place in the laser induced plasma forming different materials [36]. Condensation of the plasma under suitable conditions results in nucleation and growth of the plasma species into a desired nano-structure in the liquid medium. In this paper, we report for the first time to the best of our knowledge, synthesis of uranium-di-oxide nanoparticles by laser ablation of bulk uranium-di-oxide pellet in ethanol. The second harmonic emission of an electro-optically Q-switched nsec Nd–YAG laser was used as the coherent source here. The structural and optical properties of the fabricated uranium-di-oxide nanoparticles were investigated by X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), energy dispersive analysis of X-rays (EDX) and UV–Vis–NIR spectrophotometry. The mean size of the particles was found to have a dependence on the laser parameters. In particular, the diameter of the generated nanoparticles was seen to bear a direct proportionality with the laser fluence. XRD analysis confirmed the phase of the synthesised material to be pure uranium-di-oxide. UV–Vis–NIR absorption spectra of the colloidal solution revealed increased absorption in the UV regime.

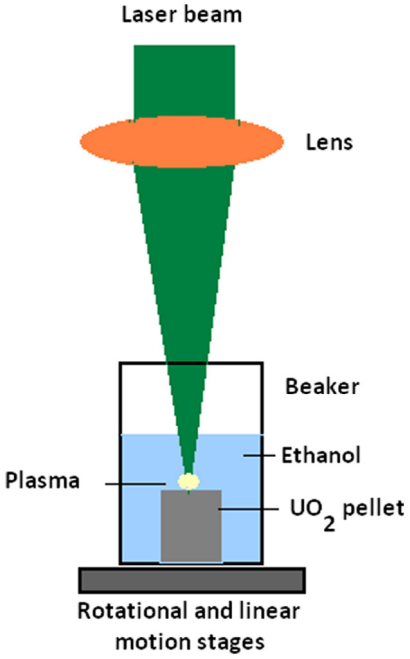

Fig. 1. Schematic of the experimental set up.

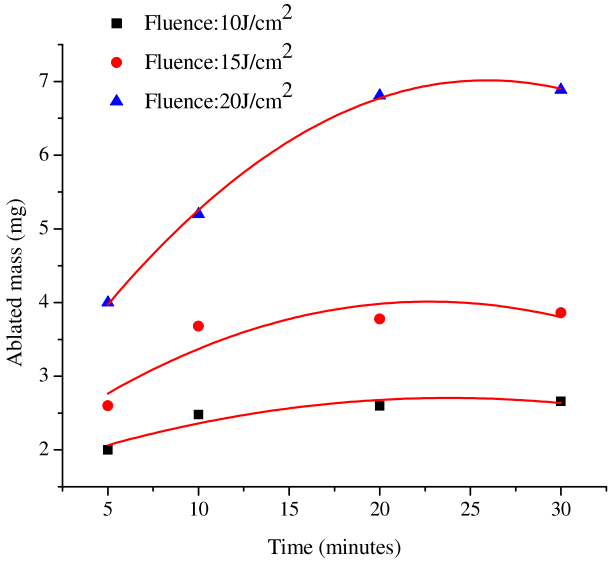

Fig. 2. Ablation rate of UO_2 as a function of time.

2. Experimental

The schematic of the experimental setup for synthesis of colloidal solution of uranium-di-oxide nanoparticles by using laser ablation is shown in Fig. 1. An electro-optically Q-switched Nd–YAG laser capable of delivering pulses of duration 6–8 ns with a maximum of 800 mJ in the second harmonic (532 nm) @ 10 Hz repetition rate was used for carrying out the experiments. The energy and duration of the pulse were monitored respectively by a pyro-electric joule metre (Coherent J-MB-50YAG) and a fast photo detector (Thorlab DET 10A) in conjunction with a digital storage oscilloscope (Tektronix TDS 380). The laser beam was focussed on the target surface to a diameter of ∼1 mm using a plano-convex lens of focal length of 100 mm. The target, which was a solid natural UO_2 pellet was placed in a small beaker containing absolute ethanol (purity > 99.9%) and was totally immersed in the liquid. The beaker was placed on a "Z" axis motion stage with a