Microstructural evolution and thermophysical property evaluation of Th-U alloys
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Thoria has been used for flux flattening and as blanket in nuclear reactors. In the early history of nuclear reactors, thorium metal has been used in combinations with other fissile counterparts [1]. However, only in limited number of cases, attempts have been made to use thorium metal in nuclear fuel [2]. Although thorium metal has many attractive properties (e.g. high melting point, isotropic crystal structure, higher thermal conductivity, good irradiation stability etc.) it needs to be alloyed with a fissile metal (e.g. uranium) to be used as fuel. Thorium-uranium fuel cycle offers many advantages (e.g. possible use in thermal as well as fast reactors, inherent proliferation resistance, high temperature of operation etc.). In Indian context of abundance of thorium against lean reserve of uranium, it makes sense to pursue explorative work on thorium-uranium alloy system. Moreover, thorium-uranium is most important subsystem of the thorium-uranium-zirconium ternary alloy system which we aim to use in CNPP in DAE, India [3].

Thorium-uranium alloy fuel has not received much research attention mainly because of easy availability of uranium and military incentive offered by U-Pu cycle. Moreover, (i) lack of a consistent systematic effort to develop the alloys and define the limitations of these fuels, (ii) dearth of initiatives to define its microstructures that can result from composition and fabrication variables are prime reasons for this system not having witnessed much developmental research endeavour [4]. Hence, it seems prudent to explore few compositions selected from thorium-uranium phase diagram keeping two primary objectives in view viz. (i) establishing its microstructural features and to study the variations in those, if any, brought about by processing variables etc. and (ii) to assess few thermal properties relevant to fuel applications. This experimental work aims at addressing gap in research on thorium-uranium alloys. Selected compositions of thorium-uranium alloy have been taken for microstructural study and evaluation of thermophysical properties.

As-cast microstructures are presented in Figure 1. The as-cast microstructures change from single phase (in Th-3U) to eutectic (in Th-96U). Uranium is present as small spheroids (~ 70 nm) in Th-3U and as islands (~ 2 to 6 µm) in Th-7U & Th-10.7U. In Th-10.7U, it starts occurring at the grain boundary junctions or triple points. With increasing amount of uranium, thin layers (Th-30U) surrounding grains, are formed and this eventually becomes a well-developed interconnected network of uranium in Th-52U. This network is woven inside another network of thorium rich phase. In Th-80U, as the amount of thorium becomes lesser, it no more remains a network and instead it is present in the form of dendrites. These dendrites are spread over uniformly in the matrix of uranium rich phase. Th-96U is a different structure from these and it is eutectic in nature where fibrous eutectic products form colonies inside the microstructure. As-cast grains are refined substantially with increasing uranium addition.

Room temperature hardness increases sharply in the beginning and up to Th-10.7U and follows gradual increase in hardness with further increase in uranium content. The hardness of Th-80U is similar to that of unalloyed uranium. Room temperature hardness of biphasic Th-U alloys has been found to be more influenced by that of uranium. However, Th-96U is less hard as it has a eutectic structure having softer thorium phase inside the fibrous colonies of eutectic product.
Figure 1. As-cast Th-U alloys; (a) TEM image of Th-3U alloy, (b) SEM (BSE mode) image of Th-7U alloy, (c) optical micrograph of Th-30U alloy, (d) SEM image (BSE mode) of Th-30U alloy - continuous network of uranium is seen, (thorium, having low atomic number has appeared darker in BSE image and uranium, harder at room temperature remains elevated after mechanical polishing), (e) SEM image (BSE mode) of Th-52U alloy - fully developed and continuous uranium network has interpenetrated in to the thorium network; (f) Th-96U alloy – eutectic colonies with a fibrous eutectic product.
The variations in room temperature thermal conductivity ($\lambda_{RT}$) and average (27-670 °C) linear thermal expansion coefficient (CTE, $\alpha_{(27-670 \, ^\circ\text{C})}$) of Th-U alloys with increase in uranium content are shown in Figure 2. The $\lambda_{RT}$ of Th-U alloys decreases with increasing addition of uranium in thorium and arrives at its lowest when uranium content is ~52 wt.%. $\lambda_{RT}$ does not change significantly beyond this point and up to that of unalloyed uranium. The experimental data has been found to follow a logarithmic decrease with increase in the amount of uranium. A model explaining this behaviour has been proposed based on the effect of interfacial area between adjacent phases.

![Figure 2](image)

**Figure 2.** (a) Room temperature thermal conductivity values of Th-U alloys are shown against uranium content. Model fitted to the experimental data is also shown in the figure. (b) variation of linear coefficient of thermal expansion (27-670 °C) with uranium content in Th-U alloys. CTE of uranium (27-670 °C) in [010] direction is (-)7×10^{-6} °C^{-1}, negative value is not shown in the plot.

The $\alpha_{(27-670 \, ^\circ\text{C})}$ is not substantially influenced by addition of small amount uranium (<15 wt.%) in thorium. The thermal expansion behaviour remains nearly unaffected (≤7% increase only) till ~15 wt.% uranium alloying in thorium. However, the change in $\alpha_{(27-670 \, ^\circ\text{C})}$ is significant when uranium content is >30 wt.% and it is seen to be progressively influenced by increasing presence of uranium in the alloy.

Based on the microstructural features and thermophysical property evaluation it is seen that high thorium Th-U alloys have appreciable thermal conductivity and low thermal expansion coefficient. It can reasonably be concluded that high thorium Th-U alloy can be used for possible nuclear fuel application in reactors provided other factors (e.g. reactor physics, post irradiation examinations etc.) are also seen to be favourable.

**References:**

3. Department of Atomic Energy, India.