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Development of powder metallurgy technique for synthesis of U_3Si_2 dispersoid

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ABSTRACT

The Indian research reactor APSARA, which uses HEU based UAl_3 fuel dispersed in aluminium matrix, is to undergo core conversion to LEU based U_3Si_2 dispersoid in aluminium matrix fuel. Work has been initiated at BARC, to prepare uranium silicide (U_3Si_2) compound using powder processing route in place of vacuum induction melting or arc melting route. This paper describes the preparation flow-sheet of the U_3Si_2 dispersoid by an innovative powder processing route with uranium metal powder and silicon powder as starting materials.

1. Introduction

In the late 70s the Reduced Enrichment for Research and Test Reactor (RERTR) programme was initiated in many countries to address the concern related to proliferation and diversion issues. Till that time, most of the research and test reactors all over the world were using compact core containing highly enriched uranium (HEU), with uranium enrichment greater than $85\%U^{235}$. The fuel was in the form of a plate containing UAl_x (mainly UAl_3) or U_3O_8 dispersed in aluminium matrix and clad in an Al-alloy [1]. Since then, the research is mainly focused all over the world on the use of such fuel cycles which can utilize low enriched uranium ($LEU < 20\%U^{235}$) instead of HEU. This can be achieved either by increasing the fuel loading or by using higher uranium density compounds. The compounds of uranium and silicon have high densities and their use in the reactor fuel was considered primarily because of high uranium content per unit volume. Among the seven phases in the uranium–silicon system (Fig. 1) only U_3Si and U_3Si_2 have received the most attention as reactor fuel [2]. Among the above U_3Si_2 has been considered as a better fuel material because of its friability and low swelling rate, though the U_3Si has higher uranium density in comparison with U_3Si_2 (see Table 1). The major problem with U_3Si as a reactor fuel is that it develops extraordinarily large voids at medium burn-up that leads to unacceptable breakaway swelling at high fuel loadings [3–6].

U_3Si_2 has been found to perform extremely well under irradiation tests in comparison to U_3Si even with uranium densities up to 5 gU/cm^3. France has mastered the manufacture of plate type fuels containing U_3Si_2 with densities up to 6 gU/cm^3 [7]. Uranium silicide obtained by arc or induction melting normally consists of a mixture of intermetallic compounds (i.e. U_3Si_2, U_3Si, USi, etc.) [2,8,9]. Development work earlier done at Bhabha Atomic Research Centre (BARC) [10–12] also used the induction melting route for the preparation of U_3Si_2 for fabricating mini plates.

It is evident from the U–Si phase diagram (Fig. 1) that the U_3Si compound is formed from the peritectic reaction while the U_3Si_2 is a congruent melting compound. In most of the cases synthesis of such compounds was carried out either by arc melting in argon or induction melting in vacuum of elemental buttons. In both these cases reaction takes place in the liquid phase and the temperature required for synthesis is of the order of ~1700 °C. Further in both the cases, two to three remelting operations of the product is required in order to get good homogeneity, which suggests that there is always a chance for the product to get contaminated by the crucible and/or electrode material. To avoid these problems, a powder metallurgy route has been selected for the fabrication of U_3Si_2. In this method the entire operation takes place below the melting point of the reaction product (U_3Si_2).

The APSARA research and test reactor, which is located in BARC, India, is under up-gradation and the project of developing LEU ($<20\%U^{235}$) based fuel is a part of the programme. At present this reactor uses plate type fuel of HEU ($>85\%U^{235}$) in the form of UAl_3 dispersed in aluminium matrix. With a view to develop an alternate LEU fuel, work has been initiated in BARC to develop plate fuels with U_3Si_2 as dispersoid in aluminium matrix. This new fuel will enable to increase the reactor power from 1 MWth to 2 MWth. The dimensions of the fuel plates are given in Table 2.

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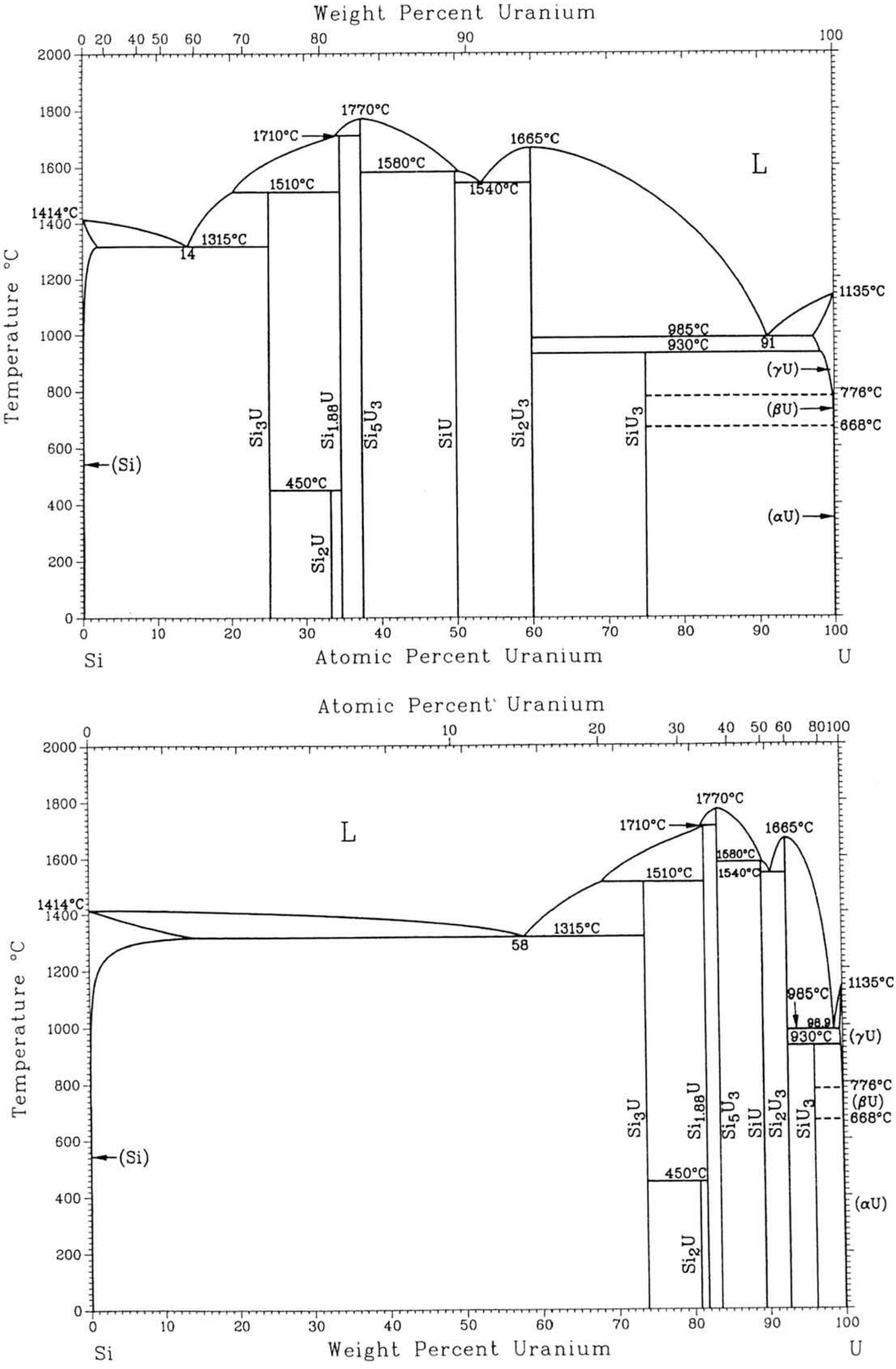

Fig. 1. Uranium–silicon phase diagram.

Table 1
Density of various fuel compounds

Uranium compound	UAl_4	UAl_3	UAl_2	U_3O_8	USi	U_3Si_2	U_3Si
Theoretical density (g/cm^3)	6.1	6.8	8.1	8.3	11.0	12.2	15.4

Table 2
Proposed fuel plate dimension for modified APSARA core

Description (fuel plate)	Length (mm)	Width (mm)	Thickness (mm)
Proposed fuel plate dimensions for APSARA reactor	640	72.5	1.5

The present paper covers the flow-sheet for U_3Si_2 compound preparation by powder processing route with uranium metal powder and silicon powder as starting materials. The results of characterization of the product using X-ray diffraction, optical and electron microscopy are also presented.

2. Experimental work

2.1. Preparation of U_3Si_2 compound

The starting material for this study is uranium metal powder (see in Fig. 2(a)) and silicon powder. Silicon chunks (see in Fig. 2(b)) were crushed and sieved to get silicon powder (see in Fig. 2(c)). The purity of U and Si powders used for this study was 99.6 and 98.6 wt%, respectively. Chemical characterization of uranium metal powder is shown in Table 3.

Extreme care was taken throughout the powder handling operation to avoid any oxidation of powder. For this purpose the entire powder handling operation was carried out inside the glove boxes flushed with once through high purity argon gas. The oxygen and moisture content in the high purity argon gas was 10 ppm and 5 ppm, respectively. The batch size for the initial trial runs was 200 g. Batch size was increased to kg level once the preparation parameters are optimized. To avoid oxidation of uranium during heating operation, glovebox housing the furnace was continuously purged with high purity argon gas.

The flow-sheet for the fabrication of U_3Si_2 fuel granules is shown in Fig. 3. For preparation of U_3Si_2, uranium metal powder and silicon powder were weighed in the required stoichiometry and were mixed in a cubical blender for 4 h. The powders were mixed thoroughly because this step has a large influence in the homogeneity of the product. The mixture was then compacted in a hydraulic press in the form of cylindrical pellets. During the pressing operation no admixed binder/lubricant was added to the mixture. Only die-wall lubrication was used. The pellets were compacted at 600 MPa pressure in a 20 mm diameter die and L/D ratio was kept at around 0.5–0.75. The green pellets thus compacted were loaded in molybdenum trays coated with plasma sprayed alumina. The charge carrier was then loaded in a 10 kW resistance heating furnace with graphite as heating element and synthesis was carried out under vacuum of ~1.33 Pa at 1550 ± 10 °C for 4 h. During the heating cycle, intermittent soaking was carried out at 1100 °C and 1400 °C for 1 h each. The synthesis temperature was kept well below the melting point of U_3Si_2. Hence liquid formation was avoided in the process which makes the product more clean as compared to the one synthesized by the conventional route. After cooling the furnace to room temperature, the synthesized U_3Si_2 clinkers were taken out for crushing. Due to the pyrophoric nature of the silicides, particularly in powder form, crushing was done in a glovebox flushed with once through high purity argon gas. Crushing was done using a jaw crusher followed by sieving in a sieve shaker. U_3Si_2 particles of below 125 μm were used for further processing into fuel meat after blending it with required quantity of aluminium powder.

Table 3
Chemical characteristics of uranium metal powder

Element	U	C	N	B
Concentration (wt%)	99.6 ± 0.1	0.063	0.050	0.0001

2.2. Characterization of U_3Si_2

For phase analysis, sample was prepared by compacting U_3Si_2 powder into about 31 mm diameter tablet. The XRD pattern was generated with the scan rate of 2°/min. Tube voltage was kept at 40 kV and the current was 3 mA. The X-Ray diffraction patterns of the pellets were obtained by using Cu Kα radiation monochromatized with curved graphite monochromator. For metallography, U_3Si_2 pellet was first cold mounted in 25 mm diameter mold and then it was ground using SiC papers. The sample was then polished

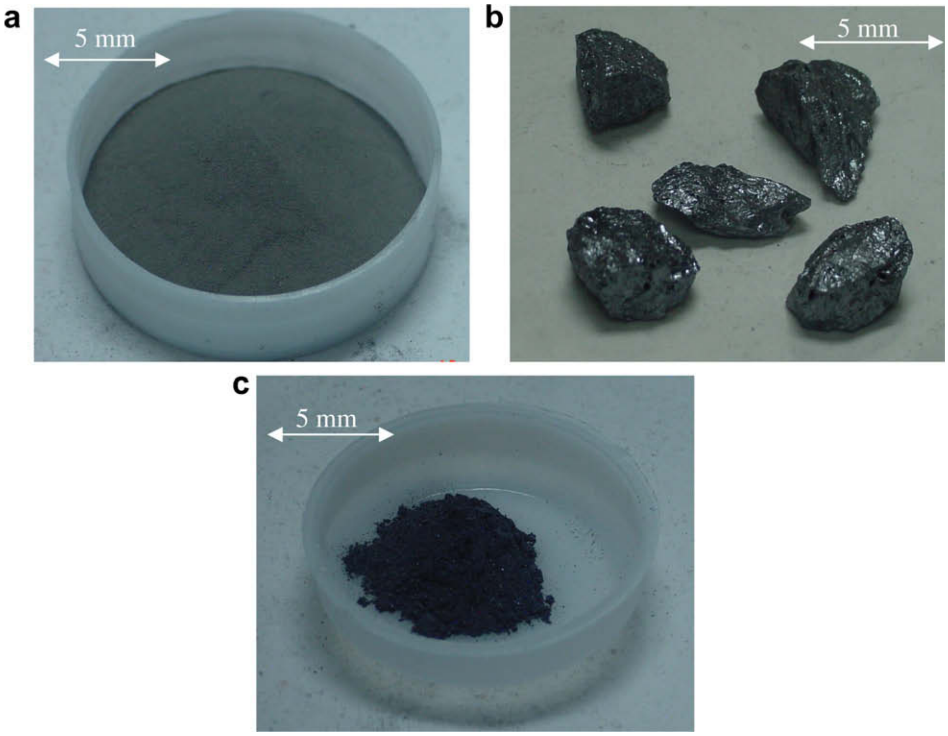

Fig. 2. Photographs of raw materials: (a) uranium metal powder; (b) silicon chunks; (c) silicon powder.

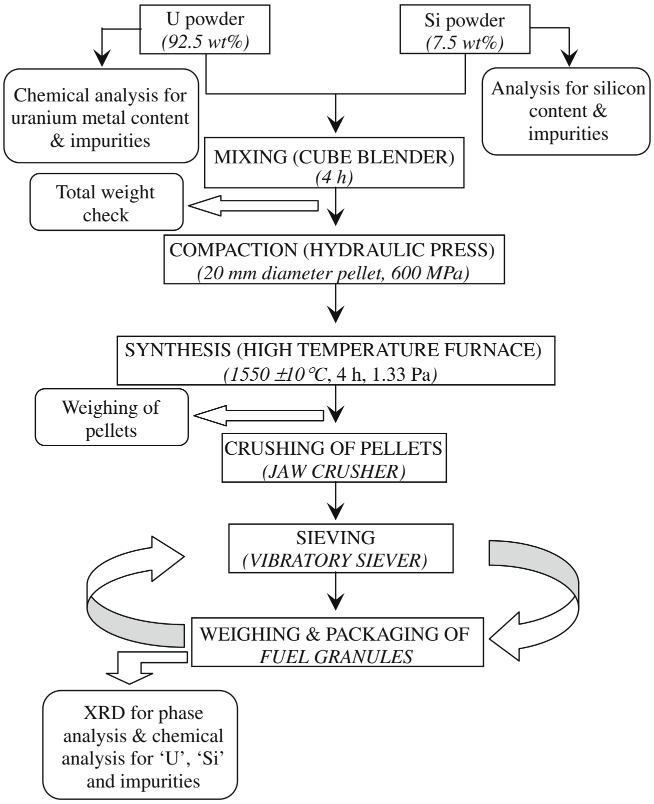

Fig. 3. Flow-sheet for fabrication of U_3Si_2 fuel granules by powder processing route.

mechanically using 2 µm diamond paste and then etched with freshly prepared etchant of very dilute phosphoric acid. The etched sample was then observed under microscope. This sample was further deep etched and examined under SEM.

3. Results and discussion

The U_3Si_2 pellets prepared by powder metallurgy route are shown in Fig. 4 which indicates that the pellets were physically intact after synthesis and no major cracks can be seen with the naked eye. The entire operation was carried out in well ventilated alpha tight glove boxes because of the pyrophoric characteristics of the powder. The XRD pattern of the synthesized U_3Si_2 is shown in Fig. 5. The pattern confirms the presence of U_3Si_2 as a major phase and other silicides are not present in detectable quantities. In other conventional techniques, it is very difficult to get single phase product (i.e. U_3Si_2) in a single step. Repeated melting is necessary to get a single phase compound [2,8,9]. The XRD pattern also suggests the presence of UO_2 in minute quantities. This UO_2 has been formed due to oxygen pick-up during handling of uranium metal powder.

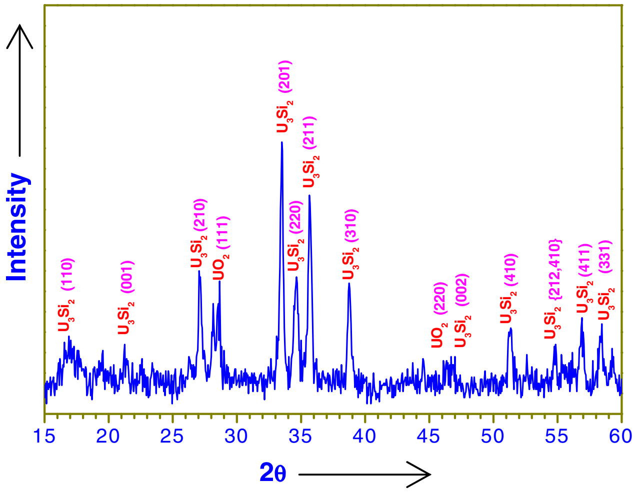

Fig. 5. XRD pattern of synthesized product (U_3Si_2) prepared by powder processing route.

The product formed by powder processing route does not require any further homogenizing treatment as in the case of conventional techniques [2,13]. Reason for this is that during mixing stage, the starting material, i.e. uranium metal powder and silicon powder are mixed intimately so that the uranium metal powder and silicon powder are in close contact with each other on micro-level because of their smaller particle size. Further compaction into pellets leads to a firm contact between uranium and silicon particles and facilitates solid state reaction at elevated temperature.

The optical photomicrograph of the as synthesized U_3Si_2 clinker is shown in Fig. 6 which indicates the presence of only U_3Si_2 phase. The grain size of U_3Si_2 clinker was in the range of 500–700 µm. Fig. 7 shows the SEM picture of U_3Si_2 clinker/pellet. The figure clearly reveals that cleavage fracture took place in the synthesized U_3Si_2 pellets during crushing. The product formed by the powder

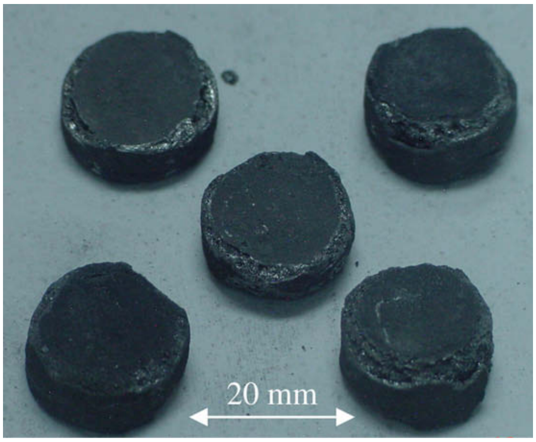

Fig. 4. Photograph of U_3Si_2 pellets after synthesis.

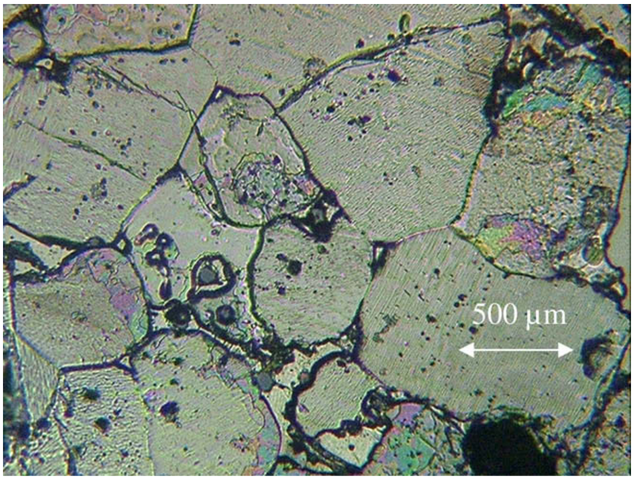

Fig. 6. Microstructure of U_3Si_2 clinker (as etched).

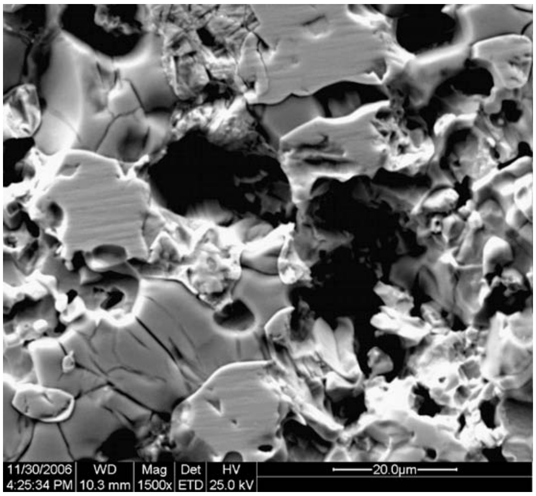

Fig. 7. SEM photomicrograph of U_3Si_2 clinker.

processing route has a density of around 75–80% T.D. The presence of porosity in the grains of U_3Si_2 is also shown in Fig. 7.

4. Conclusions

U_3Si_2 was synthesized by using powder processing route with uranium metal powder and silicon powder as the starting materials and a flow-sheet could be established for the production of U_3Si_2 granules. By this technique, it was possible to produce single phase U_3Si_2 compound. The U_3Si_2 clinker formed by this technique is porous in nature and has a density of around 75–80% T.D. This clinker could be easily crushed to fuel particles of required size (<125 μm) for further processing.

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