FABRICATION OF MIXED URANIUM PLUTONIUM CARBIDE FUEL PELLETS FOR FBTR BY DIRECT PROCESSING OF SINTER REJECTS

U. Basak, P.S. Kutty, S. Mishra, S.K. Sharma, K.S. Bhatnagar, R.S. Mehrotra, S. Majumdar and H.S.Kamath
Nuclear Fuels Group
Bhabha Atomic Research Centre

Abstract

Fast Breeder Test Reactor at Kalpakkam uses plutonium rich mixed uranium-plutonium carbide as driver fuel in the form of sintered cylindrical pellets encapsulated in stainless steel cladding tube.The process flowsheet followed for the fabrication of sintered pellets is well established and involves preparation of carbide clinkers by vacuum carbothermic reduction of oxide- graphite mixture followed by processing of crushed clinkers into sintered pellets by powder metallurgical route. The sintered pellets rejected due to physical defects only are crushed and oxidized before further processing into fuel pellets.The present paper highlights a direct processing approach for the fabrication of fuel pellets from sinter rejects. This process does not involve vacuum carbothermic reduction and offers many advantages including substantial reduction in radiation exposure to personnel.

Introduction

Plutonium rich mixed uranium-plutonium carbide fuel of two compositions namely (30%UC-70%PuC) as Mark I and (45%UC-55%PuC) as Mark II are being used as driver fuel in the Fast Breeder Test Reactor (FBTR) at Kalpakkam. Fabrication of these fuel pellets involves the following two major steps :

(i) preparation of mixed uranium-plutonium carbide clinkers by vacuum carbothermic reduction of oxide – graphite mixture

(ii) crushing of clinkers and milling of powder followed by pre – compaction, granulation, final compaction and high temperature sintering in argon – 8% hydrogen gas mixture

In order to fabricate fuel pellets suitable for use in FBTR, process control parameters are required to be followed strictly for maintaining chemical quality of fuel pellets particularly the specifications on carbon, oxygen, nitrogen, Pu/U, M_2C_3 (M = Pu + U) and impurities. Finally, powder batches qualifying the chemical specifications are further processed for the fabrication of fuel pellets. The flowsheet already developed[1,2] and followed for the fabrication of mixed carbide fuel pellets incorporating process control parameters is shown in Fig. 1. The accepted sintered pellets are used for encapsulation and the pellets rejected due to off - dimensions and physical defects such as cracks, chips, surface defects etc. are normally crushed and oxidized before further processing into fuel pellets. By this re-cycling approach, rejects have to undergo the entire process steps (Fig.1) from mixing of oxide – graphite mixture to high temperature sintering once again and this leads to decrease in production throughputs and increase in radiation exposure to personnel.

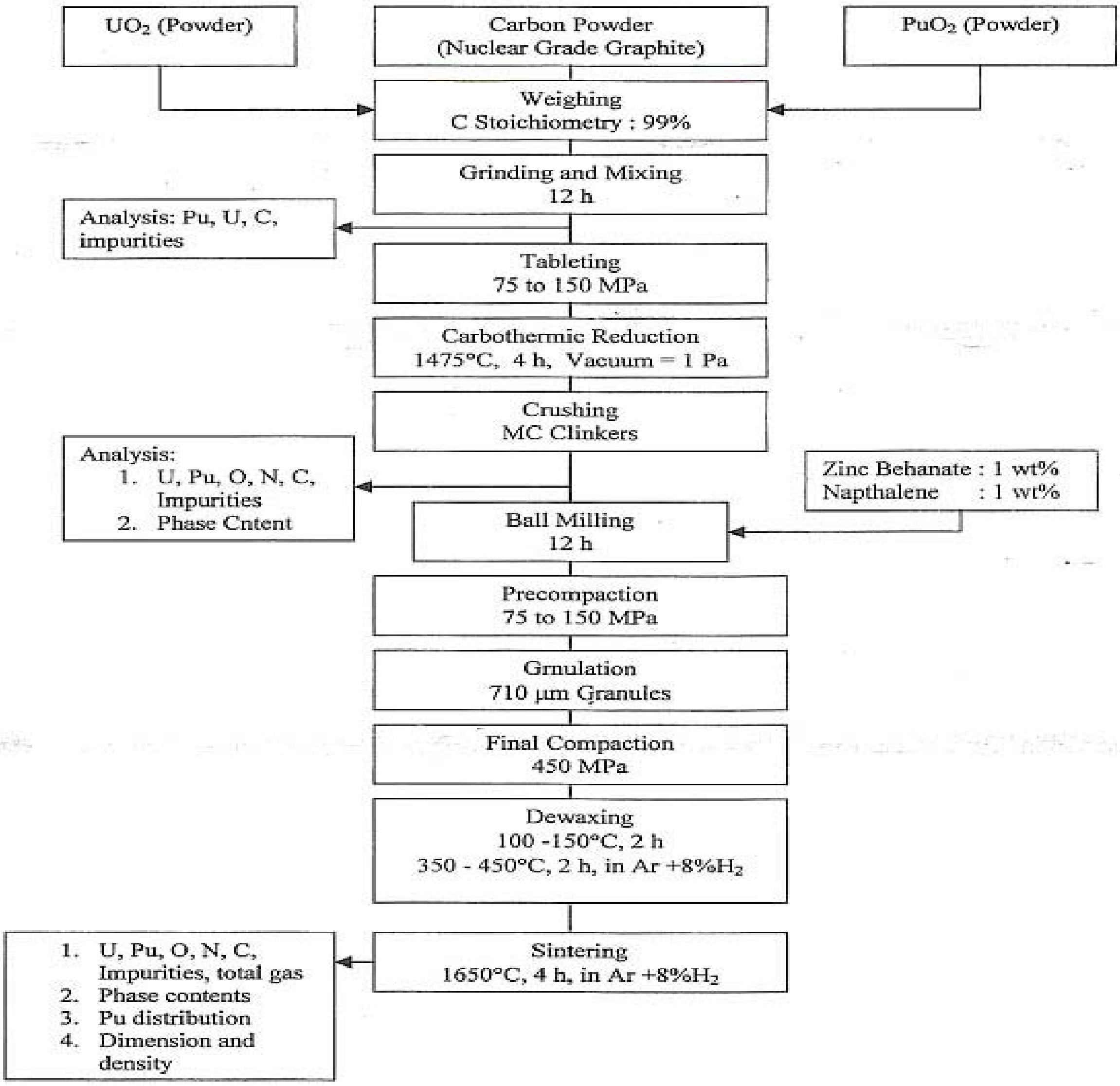


Fig.1 Flow sheet for fabrication of (U,Pu)C pellets for FBTR

Direct processing of chemically accepted but physically rejected sintered pellets has the following advantages:

- number of process steps is minimized .
- mixing & milling of oxide – graphite mixtures, tabletting and vacuum carbothermic reduction are not required leading to almost 30% savings in time & energy.
- loss of plutonium is minimized
- production throughputs is increased
- radiation exposure to personnel is reduced

Experimental

The rejected sintered pellets were directly crushed into powders and milled for 10 hours in planetary ball mill.The apparent density of powders was measured by Hall's Flow meter after every two hours of milling. Milled powders were divided into three batches (size: 200gms) and each batch was mixed with binder & lubricant for additional one hour in the following combinations :

Batch A : 1% zinc behanate and 1% naphthalene
Batch B : 1.5% zinc behanate and 0.5% naphthalene
Batch C : 2% zinc behanate

These powder batches were subjected to pre – compaction at 5 tsi followed by final compaction at ~ 20 tsi. The green pellets were dewaxed at 500° C for two hours with intermittent soak at different temperatures for adequate time in order to ensure complete removal of binder / lubricant. The dewaxed pellets were sintered at 1620ºC for 4 hours in argon – 8% hydrogen atmosphere, maintaining the heating and cooling rates at ~5ºC / min.

Results & Discussions

With progressive milling of powders , the apparent density decreased from 5.97g/cm^3 after two hours to 4.26g/cm^3 after ten hours and the specific surface area measured for the 10 hour milled powder by BET method was 0.32 m^2/g. The characteristics of pellets in terms of linear mass (mass/height), green densities and sintered densities made from different batches are shown in table 1. Densities of sintered pellets obtained from powders mixed with 1% zinc behanate and 1% naphthalene (Batch A) were found to satisfy FBTR fuel pellet specifications. Hence, a large batch (size : 600g) was processed under Batch A condition for the fabrication of sintered pellets for FBTR. Analysis of the critical parameters of sintered pellets before and after re-cycling is shown in table 2. The pellets made from direct processing and satisfying the fuel pellet specifications were used for fuel pin fabrication.

Table 1: Characteristics of (45%UC-55%PuC) fuel pellets fabricated from sinter rejects by direct processing

Batches	Green pellet linear mass(m/h),g/cm	Green density, %T.D	Sintered pellet Linearmass(m/h),g/cm	Sintered density, %T.D
A	1.49	65.90	1.58	87.4
B	1.56	69.03	1.69	93.5
C	1.55	68.83	1.68	94.4

Table 2: Analysis of (45%UC-55%PuC) sintered pellets

Chemical analysis	Before re-cycling (sinter rejects)	After direct processing	FBTR fuel pellet specification
Plutonium (wt%)	(i) 55.41 (ii) 54.82	(i) 55.37 (ii) 54.74	Pu + U = 95 wt% max.
Uranium (wt%)	(i) 39.31 (ii) 39.59	(i) 39.26 (ii) 39.33	
Carbon (wt%)	(i) 4.92 (ii) 4.68	(i) 4.68 (ii) 4.65	$\leq$ 5.03
Oxygen (ppm)	(i) 4251 (ii) 4585	(i) 4281 (ii) 4921	$\leq$ 5000
Nitrogen (ppm)	(i) 693 (ii) 1196	(i) 1123 (ii) 1603	$\leq$ 2000
M_2C_3 (wt%) M = U + Pu (by XRD)	(i) 11.40 (ii) 8.0	(i) 11.40 (ii) 12.40	5 – 20%
Total impurities(ppm)	(i) 668 (ii) 707	(i) 849 (ii) 852	$\leq$ 3000

Conclusions

1. Milling of crushed sintered pellets for ten hours resulted in powder characteristics similar to that obtained from crushed clinkers prepared by vacuum carbothermic reduction of oxide – graphite mixtures .
2. 1% zinc behanate and 1% naphthalene as binder- lubricant mixture was found to be optimum for obtaining pellets meeting FBTR fuel pellet specification.
3. Direct processing of sinter rejects improves production throughputs, reduces radiation exposure to personnel and minimizes plutonium losses.
4. Several fuel pins made from pellets fabricated by direct processing of sinter rejects were delivered to IGCAR for use in FBTR.

References

1. Development and fabrication of 70% PuC - 30% UC fuel for the Fast Breeder Test Reactor in India, Nuclear Technology, 72(1), 1986, p. 59-69.C.Ganguly, P.V.Hegde, G.C.Jain, U.Basak, R.S.Mehrotra, S.Majumdar and P.R.Roy.
2. Process control in mixed carbide fuel pellet fabrication for FBTR , Proc.Fast Breeder Reactor Fuel Cycle, RRC, Feb. 10 – 11, 1986. G.C.Jain, P.V.Hegde, U.Basak, S.Majumdar and C.Ganguly.

This paper received the 2nd Best Paper award in 14th Annual Conference of Indian Nuclear Society, INSAC 2003, held at IGCAR during December 17-19, 2003

About the authors ...

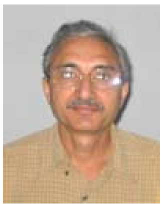
Mr U. Basak, *M.Sc.(Tech), Metallurgy, is a graduate from 21st batch of Training School,BARC. His expertise includes development of the processes for the fabrication of ceramic nuclear fuels for thermal and fast reactors.*

Mr P.S.Kutty, *AMIE.(Metallurgy), joined RMD, BARC, in the year 1976.He is involved in the fabrication of mixed carbide fuel for FBTR.*

Mr S. Mishra, *M.Tech.(Metallurgy) is a graduate from 34th batch of BARC Training School. He is involved in fabrication of mixed carbide fuel for FBTR and development of thorium-based fuels for thermal reactors.*

***Mr S.K.Sharma,** B.Sc, joined BARC in year 1981. He is working for fuel pellets and pin fabrication for FBTR.*

***Mr K.S. Bhatnagar,** B.Sc, joined BARC in year 1981. He is working for fuel pellets and pin fabrication of FBTR.*



***Dr.R.S. Mehrotra** is a graduate from 13th batch of Training School. His expertise is in the field of fast reactor fuel and fuel pin fabrication. He is presently Head of Fast Reactor Fuel Section, Radiometallurgy Division, BARC.*

***Mr S. Majumdar,** B.E., Metallurgy, is a graduate from 11th batch of BARC Training School. His expertise is in the field of fabrication and characterization of nuclear fuels, both for thermal and fast reactors. He is currently Head of Radiometallurgy Division, BARC.*

***Mr H.S. Kamath,** B.E., Metallurgy is a graduate from 13th batch of Training School, BARC. His expertise is in the area of nuclear fuel cycle activities. He is presently Director of Nuclear Fuels Group, BARC.*