

PLUTONIA-THORIA FUEL CYCLE AS STARTING  
SOLUTION FOR A WIDER THORIUM USE

Carlo Lombardi  
Politecnico di Milano  
Dip. Ing. Nucleare  
via Ponzio 34/3  
20133 Milano, Italy  
(0039) 02 23996397

Enrico Padovani  
Politecnico di Milano  
Dip. Ing. Nucleare  
via Ponzio 34/3  
20133 Milano, Italy  
(0039) 02 23996325

Marco E. Ricotti  
Politecnico di Milano  
Dip. Ing. Nucleare  
via Ponzio 34/3  
20133 Milano, Italy  
(0039) 02 23996360

Fortunato Vettraino  
ENEA  
Nuclear Fission Division  
via Martiri di Monte Sole 4  
40129 Bologna, Italy  
(0039) 051 6098485

ABSTRACT

This paper is focused on a description of thoria fuel option. Our opinion is that this option, beyond being a valuable way to exploit the energy content of plutonium without further breeding it, may be a starting point for introducing an Uranium-Thorium fuel cycle, based on a different strategic context with respect to past proposals. The option is based on the adoption of current or advanced PWRs, the latter characterised by a reduced fuel power density, always adopting conventional fuel rods and assemblies. A three-batches full core loading scheme is assumed. The thoria-plutonia composition is determined by the constraints to obtain at Beginning Of Life (BOL) a non positive void coefficient, and to reach a burnup as high as possible. Different fuel compositions and pellet radius are considered. The plutonium content is in the range of 4.5÷15%, mainly depending on the plutonium quality, namely Weapon Grade (WG) or Reactor Grade (RG). The results are in terms of dynamic coefficients, life duration, plutonium consumption and final isotopic compositions. These fuels show the capability to destroy about 40÷60% of total plutonium for RG, while this figure rises to 65÷70% for WG. These values are well above those obtained by MOX option. A variant to eliminate any proliferation concern foresees the addition of small quantities of  $^{238}\text{U}$  at the expenses of a reduction of the fuel burnup and its capacity in burning RG plutonium, while the opposite occurs for WG.

The low boron worth is not different from the MOX one, being related mainly to the plutonium content, and much less to the chosen fertile isotope. Therefore, modifications of control devices for a full core strategy is not expected to be different in the two cases.

The results confirm the viability of this proposal, apt to future variants, including those connected to accelerator

actinide burning solution. An irradiation experiment, expected to take place at Halden HWBR in the context of the ENEA participation to the Halden Project, is the main part of the Inert Matrix – Thoria fuel R&D activity presently underway as a Polytechnic of Milan–ENEA co-operation.

I. INTRODUCTION

The wide and successful operation of nuclear power plants, in addition to a very moderate recycle of the discharged fuel, has given rise to the accumulation of a plutonium stock largely exceeding 1000 tons world-wide. Besides this large amount of RG plutonium, at least 100 tons of WG plutonium are expected to come from the dismantling of nuclear weapons, as a result of the disarmament agreements between USA and the former USSR.

In lack of a clear strategy for a future use, for instance in FBRs, all this plutonium has to be disposed of. The option of burning it is being viewed under a novel standpoint: not as a mean for reaching a more efficient utilisation of resources, but as a possible option to reduce the plutonium stockpile, or at least to limit its further build-up.<sup>1-16</sup>

The use of MOX fuel in LWRs is the current reference solution, even though it is well known that it leads only to a reduction of the plutonium growing rate and its proliferation capability, and not to a stabilisation of its levels. As far as the WG plutonium is concerned, the MOX option does not aim at substantially destroying the plutonium stocks, but rather at reducing the proliferation potential of the discharged fuel down to the same level of the conventional one.<sup>17</sup> This is a technically sound solution, but cannot be considered fully satisfactory, due to the limited plutonium consumption. Then, we studied the

following uranium-free options for an efficient burning of WG- and RG-plutonium:<sup>18-21</sup>

- inert matrix fuel, based on stabilised zirconia;
- thoria doped inert matrix fuel, in which part of zirconia is substituted with thoria;
- thoria fuel.

For all these options a once-through cycle was considered, with direct disposal of the burnt fuel.

The main conclusions of our past works were:

- inert matrix fuels appear the best for plutonium burning in a PWR within a once through cycle scheme, however contrasting the high peaking factors is a major issue to cope with, which needs further computational efforts. In addition, the technology of this kind of fuel and its in-pile behaviour has to be validated by costly and time consuming R&D programs.
- a thoria based fuel appears as an interesting option. In a once through cycle the burning capability is good, particularly if a highly moderated lattice is adopted; alternatively, if a reprocessing strategy is foreseen, this can be an effective way to start a closed U-Th cycle.

In the past, we focused our attention on the plutonium burning capabilities of these fuels. The present work is devoted to the neutronic analysis of some thoria-plutonia fuels, with emphasis on the reactivity coefficients and fuel burnup. The different imposed constraints bring to new fuel compositions and neutronic behaviours.

## II. CALCULATION METHOD

The neutronic analysis was performed with the WIMSD-5 code<sup>22</sup>, considering an infinite lattice of fuel pins. The cell dimensions, reported in Table 1, corresponds to a 17x17 fuel elements, with 264 active pins and 24 water rods. The selected linear power is 13.5 kW/m, that is the same value adopted in the AP600 reactor. The plutonium vectors for RG and WG plutonium are reported in Table 2. The RG composition corresponds to the plutonium coming from an enriched uranium fuel burnt till 33 MWD/kg in a standard 900 MWe PWR,

Table 1. Reactor design parameters

Reactor power, MWth	2035
No. of assemblies	156
Assembly layout	17x17
Assembly pitch, mm	214.2
No of rods per assembly	264
Fuel rod diameter, mm	8.2
Clad outer diameter, mm	9.5
Pin pitch, mm	12.6
Active fuel length, m	3.66
Average linear heat rate, kW/m	13.5

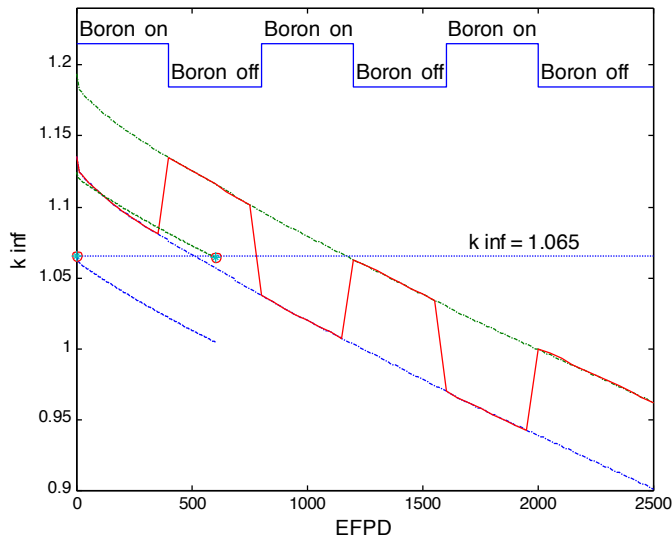
Table 2. Plutonium isotopic composition (wt%)

Isotope	Weapons-Grade	Reactor-Grade
<sup>238</sup> Pu	--	1.32
<sup>239</sup> Pu	93.3	60.32
<sup>240</sup> Pu	6.0	24.27
<sup>241</sup> Pu	0.6	8.33
<sup>242</sup> Pu	0.04	4.95
<sup>241</sup> Am	0.06	0.81

reprocessed after 10 years of cooling, then immediately used to fabricate a plutonium bearing fuel, and placed in the reactor 2 years later.<sup>23</sup>

The convergence value for k-inf was 1.065: when this value is attained it is assumed the corresponding k-eff value is 1; it was deduced from previous calculations on an infinite lattice of 3.2% enriched uranium pins, with the same cell dimensions. The difference between k-inf and k-eff were ascribed to leakage and not included parasitic absorptions (grids, instrumentation, etc.). When fuels with high plutonium content are inserted in the core, the flux greatly hardens, so that parasitic thermal captures in the above mentioned structural materials are decreased; on the other hand, when a fuel reaches a very high burnup, as in many of the cases considered in this work, an improved fuel assembly design, with higher capture cross sections, is more appropriate than the usual one, adopted in our calculations. These two effects partially compensate each other, than the k-inf value corresponding to criticality is not expected to differ much from the present 1.065 value. However, the decrease of the k-values versus the burnup is much slower for these fuels than for the enriched uranium one, than a little variation of the assumed convergence k-inf value leads to a consistent variation of the fuel life. Other approximations are inherent to the cross section libraries and resonance shielding treatment, which are, in general, much less accurate when the fuel is thorium-plutonium in respect with enriched uranium. In conclusion we can state that the reported results are affected by approximations much larger than the ones relevant to a conventional fuel; the reader should focus his attention to the rankings, which are more reliable than the single figures.

Figure 1. Reactivity curves for MOX with RG-Pu



### Depletion analysis

For depletion calculations the following procedure was applied:

- tentative fresh fuel composition and boron concentration in the water at BOL were first assumed;
- the boron was assumed as a not burnable material. It was alternatively present/absent for periods of 400 EFPDs, then, from a single depletion calculation, two reactivity values can be deduced at each time step (see dots and dashes lines in fig. 1);
- from the two reactivity curves so obtained, the mean values over three adjacent time intervals of equal length, corresponding to a tentative batch life in a zero-dimensional approximation, are calculated (see dashed lines in fig. 1).

If both the  $k_{\text{inf}}$  value at begin of batch life with boron and the  $k_{\text{inf}}$  value at end of batch life without boron happen to be equal to 1.065, then the set of the three chosen parameters (fuel composition, boron content and batch life) forms a coherent set, but it is not yet considered acceptable, unless the two following constraints are satisfied:

1. the dynamic coefficients are to be negative
2. the fuel life is to be as long as possible.

This two constraints operate on the choice of the initial plutonium content in opposite ways: the second constraint pushes towards high plutonium contents (and high boron contents, as the set of parameters has to be coherent), till a limit is reached when one of the dynamic coefficients becomes positive, so that the first constraint limits the plutonium contents. Then a sequence of depletion trials has to be performed for selecting an acceptable set of parameters. The adopted convergence rule was:

- stop the trials when a coherent set of parameters has been selected, for which all dynamic coefficients are negative and one void coefficient is included in the range  $[-10, 0]$  pcm / %void.

### III. RESULTS

In table 3 the composition of the analysed fuels and the boron content in water (ppm weight of natural boron), as resulting from the above procedure, are reported. MOW and MOR refer to MOX fuels, while the others are thorium based fuels. THW7, THR7, THW9, THR9 are variants, in which the pellet diameter is reduced or increased by 1 mm, while the pitch and the linear power remain the same. As it was not possible to obtain an acceptable set of parameters for the case THW7, then it was not considered any longer in the following tables.

The symbol THWB refers to a variant of the THW fuel, in which a burnable poison is inserted in the form of Integral Fuel Burnable Absorber (IFBA), which is a thin layer (thickness = 30  $\mu\text{m}$ , in this case) of  $\text{ZrB}_2$ , coated on the surface of the fuel pellets.

The W30, R30, W20, R20 symbols indicate thorium based fuels, in which part of the thorium is substituted by depleted uranium (enrichment = 0.25%), so that the uranium fissile isotopes fraction ( $^{233}\text{U} + ^{235}\text{U}$ ) /  $U_{\text{tot}}$  at EOL is reduced to 30% or 20%, so eliminating any proliferation concern of the breded  $^{233}\text{U}$ . This means that a further constraint has been imposed, and another composition parameter has to be determined; then the trial and error procedure can become somewhat long for these cases.

The dynamic coefficients were calculated for the fresh fuel, at 20 EFPDs (poisons' equilibrium) and at 2400 EFPDs (an indicative EOL). In all the considered cases the

Table 3. Fuel composition (w% HM), boron content (ppm)

Fuel	Description	Pu	Th	U	B
MOW	MOX with WG-Pu	4.5	—	95.5	2500
MOR	MOX with RG-Pu	10.5	—	89.5	2500
THW	Thoria and WG-Pu	4.5	95.5	—	1950
THR	Thoria and RG-Pu	15.2	84.8	—	3400
THW7	Thoria and WG-Pu $\varnothing 7.2$	—	—	—	—
THR7	Thoria and RG-Pu $\varnothing 7.2$	12.0	88.0	—	2100
THW9	Thoria and WG-Pu $\varnothing 9.2$	8.75	91.25	—	5200
THR9	Thoria and RG-Pu $\varnothing 9.2$	14.8	85.2	—	4300
THWB	Thoria WG-Pu IFBA	12.0	88.0	—	2450
W30	Ufiss/Utot=30%. WG-Pu	7.7	88.1	4.2	3100
R30	Ufiss/Utot=30%. RG-Pu	15.0	81.0	4.0	2800
W20	Ufiss/Utot=20%. WG-Pu	8.4	84.3	7.3	3350
R20	Ufiss/Utot=20%. WG-Pu	15.0	78.5	6.5	2600

Table 4. Dynamic coefficients

Fuel	Void05 pcm/%	Dopp pcm/K	Void05 Pcm/%	Void95 pcm/%	B worth pcm/ppm
MOW	- <b>0.3</b>	- 3.0	- 64.	- 276.	- 3.2
MOR	- 37.	- 2.4	- 59.	- <b>7.9</b>	- 2.0
THW	- <b>2.0</b>	- 3.4	- 70.	- 444.	- 3.7
THR	- <b>1.0</b>	- 2.5	- 26.	- 9.4	- 1.7
THR7	- <b>0.9</b>	- 2.4	- 52.	- 128.	- 3.2
THW9	- <b>1.0</b>	- 3.6	- 52.	- 216.	- 1.6
THR9	- 6.7	- 2.7	- 12.	- <b>4.7</b>	- 1.0
THWB	- <b>4.3</b>	- 2.4	- 36.	- 46.	- 1.8
W30	- <b>2.5</b>	- 3.8	- 64.	- 275.	- 2.6
R30	- 31.	- 2.9	- 52.	- <b>9.0</b>	- 1.7
W20	- <b>1.1</b>	- 3.9	- 60.	- 238.	- 2.4
R20	- 40.	- 3.1	- 58.	- <b>6.2</b>	- 1.7

Table 5. Fuel life, plutonium balance, Pu fiss/Pu tot (EOL)

Fuel	EFPD	Burn-up MWD/kg	Pu BOL kg/GWye	Pu EOL / Pu BOL	Pu fiss/ Pu tot
MOW	1630	45.3	1087	67.4 %	63.9 %
MOR	1790	49.6	2299	74.5 %	59.6 %
THW	1070	33.0	1492	39.1 %	56.4 %
THR	2440	74.1	2228	50.1 %	47.3 %
THR7	1780	70.5	1850	42.1 %	39.6 %
THW9	2900	70.7	1355	34.8 %	56.5 %
THR9	2240	50.1	2972	62.1 %	54.1 %
THWB	3380	104.7	1273	26.3 %	46.1 %
W30	2150	65.7	1282	35.6 %	53.9 %
R30	2080	62.9	2589	59.5 %	52.6 %
W20	2350	71.6	1285	37.6 %	55.2 %
R20	1980	59.7	2727	62.4 %	54.0 %

limiting dynamic coefficient resulted to be a void coefficient at 20 EFPDs. It is worth noticing that the 5% void coefficients were computed not only with reference to operational conditions for the water (15.5 MPa, 300 °C), but also for refuelling conditions (ambient pressure, 50 °C). It happens, particularly when WG Pu is used, that this coefficient is the limiting one.

Table 4 reports the dynamic coefficients: in the second column the 5% void coefficient for refuelling conditions of the water, and the subsequent columns the Doppler coefficient, the 5% void coefficient, the 95% void coefficient and the boron worth, all for operational water conditions. Only the coefficients computed at 20 EFPDs are reported; the limiting one is showed in bold type.

Table 5 shows the fuel life, the burn-up, the amount of plutonium charged in the core per GWe year of produced energy, and the plutonium survival fraction.

In the table 6 the fractions of energy produced by thorium and uranium isotopes and by plutonium and americium isotopes are reported. As the values were dedu-

Table 6. Energy from fissions of Th-U and Pu-Am

Fuel	Energy from Th-U	Energy from Pu-Am
THW	18.7 %	81.3 %
THR	13.4 %	86.6 %
THR7	13.0 %	87.0 %
THW9	23.7 %	76.3 %
THR9	14.0 %	86.0 %
THWB	20.3 %	79.7 %

ced from the fuel composition at EOL, it was not possible to evaluate the same figures for thoria-urania-plutonia fuels.

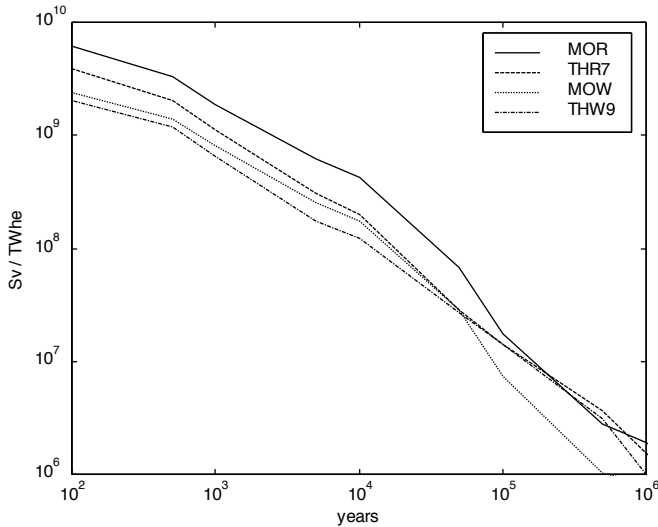
#### Comments

It is evident from the results that the choice of using WG or RG plutonium leads to very different fuel behaviours. Let us examine what happens when passing from MOX fuels to THW and THR fuels. In general substitution of thoria for urania leads to a decrease of the multiplication value, mainly due to the much fewer thorium fast fissions than the uranium one. This decrease can be balanced by an increase of the plutonium content in the case of RG, but not in the case of WG, for which a major penalisation of the fuel life results. The extreme case is THW7, for which no acceptable fuel composition was found. The boron worth gives an idea of the importance of the thermal component of the neutron spectrum. Generally, WG plutonium is associated to a greater importance of the thermal neutrons, and the limiting void coefficient occurs for high water densities. On the contrary RG plutonium-bearing fuels exhibit a lower thermal neutron importance, and the limiting condition is when a high degree of voidage occurs. The different behaviour of WG and RG plutonium is evident also for the fuel life: for RG the introduction of some denatured uranium (R30 and R20 vs. THR) translates in a life penalisation, while the contrary occurs for WG plutonium.

#### IV. RADIOTOXICITY CURVES

The radiotoxicity for some burnt fuel, expressed as Sievert per TWhe are showed in figure 2. Up to some ten

Figure 2. Radiotoxicity



thousands years, the toxicity is mainly due to the residual plutonium and the breeding of americium and curium; the ranking from the higher toxicity to the lower is: MOR, THR7, MOW and THW. During the period 10<sup>5</sup>-10<sup>6</sup> years the radiotoxicity of the thorium-bearing fuels, due to the breded <sup>233</sup>U, meets the MOR one, then afterwards becomes lower again.

## V. THE IRRADIATION EXPERIMENT SCHEDULED IN HALDEN

An R&D activity aimed at assessing the in-reactor response of Inert Matrix and Thoria fuel has been undertaken, through a first step irradiation experiment based on HEU oxide as fissile. This experiment consists in a representative inert matrices and thoria fuels test rig to be irradiated in the HWBR-Halden.<sup>24</sup> Planned design, fuel fabrication and test rig assembly actions are underway through the ENEA-HP co-operation with the aim of starting the irradiation, early next year. A second irradiation experiment based on Pu-bearing fuel is expected to follow later. The results are expected to be valuable also for Minor Actinides transmutation via inert carriers such as the calcia stabilised zirconia that we have selected for these experiments.

An IFA-Type III test rig for the Halden HWBR is proposed with a 6 rods fuel bundle and active length of about 400 mm (Fig. 3). The experiment will focus specifically on fuel, so that clad side parameters and phenomena are judged to be of minor influence to the end of fuel behaviour. A maximum linear heat rate in the range 300-350 W/cm of commercial LWR reactor is foreseen. Taking into account the low thermal conductivity of zirconia and expected peaking factors, the performance parameters will be tuned however in such a way to comply with the test rig performance limits.

The test rig which will aim at simulating, at least for the fuel, the commercial LWR conditions, will consist of a rod bundle as follows:

- 2 rods based on calcia-stab. zirconia+HEU oxide
- 2 rods based on calcia-stab. zirconia+thoria+ HEU oxide
- 2 rods based on thoria+HEU oxide

The zircaloy clad fuel rod will have standard diameter as in commercial PWRs. The minimum expected equivalent burn-up is 33 MWD/kg, which will correspond to about 600 EFPD (6 cycles of the Halden HWBR) during a 3 year irradiation time.

The test rig will be instrumented so as to keep the following main parameters tracked: fuel centreline temperature, fuel stack elongation, Fission Gas Release. For possible swelling assessment, off-line examinations in conjunction with PIE will be needed.

About the foreseeable problems of peaking factors and burn-up reactivity swings in the pure inert matrix fuel rods, a careful preliminary assessment will be done in order to either confirm burnable poisons need or/and in which measure is possible to cope with them through the appropriate safety margins, by maintaining the expected burn-up performances.

HEU containing fuel will be fabricated at Kjeller labs during the next few months for the three rod variants which will be assembled in the experimental rig: inert matrix; thoria-doped and all thoria matrix. The completed test rig is expected to be ready by the end of the year in order to be loaded in the HBWR-Halden at beginning of year 2000 during the winter outage.

## VI. INTERNATIONAL COOPERATION

International co-operation is a compulsive condition for the programme to achieve final objectives. Strong co-operation efforts will be needed for the programme to progress and an enlarged-harmonised action at European level would be desirable. In this perspective it would be highly desirable to follow up the present joint irradiation experiment in Halden with a second more challenging plutonium-bearing experiment to be carried out within the HP common programme. Also the EU 5-th Framework Programme is expected to bring a significant support to R&D experiments devoted to Trans-Uranium (TRU) and long lived fission products transmutation.

## VII. CONCLUSIONS

In this work several (Th,Pu) fuels were analysed and discussed placing the main emphasis on dynamic coefficients, burnups, plutonium consumption, and proliferation issues. The utilisation of these fuels appears advantageous due to the following reasons: a) the lack of <sup>238</sup>U prevents the production of new plutonium thus increasing the fraction of total plutonium burnt; b) the Doppler coefficients are higher than those of the standard UO<sub>2</sub> fuel. The percentage of burnt plutonium is

substantially increased by the reduction of the pellet diameter, allowing to better exploit in-situ the produced  $^{233}\text{U}$ . Notwithstanding these attractive factors, some drawbacks are also evident in the (Pu,Th) fuel cycle: i) the high thermal cross sections of plutonium isotopes significantly reduce the worth of control mechanisms such as control rods and soluble boron, thus requiring the adoption of enriched boron and, likely, a different control rod design; ii) the high fissile  $^{233}\text{U}$  is generated during the fuel irradiation and this might pose proliferation problems, although mitigated by the presence of  $^{232}\text{U}$ . For this latter problem, the addition of a limited amount of depleted uranium (4-7%) eliminates the proliferation concern at the expense of a reduction in fuel burnup for RG plutonium.

Thorium fuels show good plutonium annihilation capabilities: more than 50% of the total loaded RG plutonium is consumed at EOL. The fissile content of the discharged plutonium is in the range of 40-55%, then further less attractive for any attempt of recovery and improper use than the MOX discharged plutonium, whose fissile content is around 60%. The appealing advantage of the thorium fuel is that the existing experience, although limited, indicates an excellent behaviour under irradiation, even better than standard fuel, and a very stable behaviour in deep disposal conditions. In general, the thorium fuel cycle was thoroughly studied in the past, but the strong reduction in nuclear programmes around the world resulted in a practical stop of the development of the thorium cycle, except in India. After the past studies and the realisations performed in the world, thorium based cycles should be reconsidered, also taking into account their low actinides production. Perhaps it is worth recalling M. Lung's opinion that "in view of its potential advantages, the thorium fuel cycle has to be considered again as a promising energy source in, and after, the next century. (...) Burning some of the weapons plutonium could be one interesting way to enter the thorium fuel cycle." <sup>25</sup>

These fuels are also of interest for other applications, namely those involving Accelerator Driven Systems (ADS) which carry a strong interest in the present debate about nuclear power. In this context the above mentioned irradiation activity, managed by ENEA, can be viewed as a common program between the plutonium burning framework and the ADS technology, devoted to actinide burning by accelerators.

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Fig.3 Proposed Test Rig for Inert Matrix-Thoria Fuel Test

