Proliferation-Proof
Uranium/Plutonium Fuel Cycles
Safeguards and Non-Proliferation

by
Günther Keßler

SUB Hamburg
A 2012/7138
Contents

1 Nuclear Proliferation and IAEA-Safeguards .............................................. 1

1.1 Historical Developmenta .............................................................. 1
1.2 Safeguards Implementation .......................................................... 4
1.3 Arms reduction initiatives ............................................................ 5
1.4 Amounts of reactor-grade plutonium in the world .................................... 6
1.5 Amounts of reactor-grade americium and neptunium in the world ................. 6
1.5.1 Neptunium and americium .......................................................... 7
1.6 Nuclear fuel cycle concepts ............................................................ 8
1.7 New scientific results and further developments ...................................... 9
References Section 1 ............................................................................ 10

2 Technical applications of nuclear power reactors ........................................... 13

2.1 Nuclear reactors operating in the world in 2008 ........................................ 13
2.2 The nuclear fuel cycle ........................................................................ 15
2.3 Natural uranium ores ....................................................................... 17
2.3.1 Uranium resources ....................................................................... 17
2.3.2 Thorium resources ....................................................................... 18
2.4 Concentration of uranium ................................................................... 18
2.5 Purification of uranium ....................................................................... 19
2.6 Uranium conversion ........................................................................... 19
2.7 Natural uranium consumption and needs by the nuclear power industry .... 19
2.7.1 Natural uranium consumption by different reactor types .................... 19
2.7.2 Future need for natural uranium ..................................................... 20
References Section 2 ............................................................................ 22

3 Uranium enrichment ................................................................................ 23

3.1 Introduction ..................................................................................... 23
3.2 Enrichment technologies ..................................................................... 23
3.3 Enrichment and cascade theory .......................................................... 25
3.4 Ideal cascade .................................................................................... 27
3.4.1 Number of stages for an ideal cascade ........................................... 28
3.5 Inputs and Outputs of the enrichment process ....................................... 29
3.6 Separative work of the enrichment process ........................................... 29
3.7 Gaseous Diffusion Technology ........................................................... 32
3.8 Gas centrifuge .................................................................................. 33
3.9 Gas centrifuge technology ................................................................. 34
3.10 Russian centrifuge design .................................................................. 34
3.11 Rotor dynamics ................................................................................ 34
3.12 Laser enrichment ............................................................................. 35
3.12.1 The AVLIS enrichment technology ............................................... 35
4 Neutron and reactor physics ................................................... 39

4.1 Fission process .................................................................. 39
4.2 Neutron reactions .............................................................. 41
4.2.1 Reaction rates ............................................................... 41
4.3 Spatial distribution of the neutron flux in the reactor core ........... 43
4.4 Criticality factor $k_{eff}$ ..................................................... 44
4.5 Design of a reactor core .................................................... 45
4.6 Fuel burnup and transmutation during reactor operation ............ 47
4.7 The conversion and breeding process ................................... 51
4.7.1 Uranium-plutonium cycle .............................................. 51
4.7.2 Thorium-uranium cycle ................................................ 51
4.7.3 Conversion and breeding process ................................. 51
4.8 Fuel utilization ................................................................. 54
4.9 Radioactive Inventories in spent fuel ................................. 56
4.10 Inherent Safety Characteristics of Converter and Breeder Reactor Cores ...................................................... 57
4.10.1 Reactivity and Non-Steady State Conditions .................... 57
4.10.2 Temperature Reactivity Coefficients .............................. 59
4.10.3 Reactor Control and Safety Analysis ............................. 61

References Section 4 ................................................................. 61

5 Nuclear reactors with a thermal neutron spectrum ....................... 65

5.1 Introduction and historical development .............................. 65
5.2 European Pressurized Water Reactors (PWRs) ...................... 65
5.2.1 Core with fuel elements and control elements ................. 66
5.2.2 Reactor pressure vessel ............................................... 68
5.2.3 Primary coolant pumps, pressurizer and piping ............... 68
5.2.4 Steam generators ........................................................ 69
5.2.5 Safety injection and residual heat removal system ............ 71
5.2.6 In-containment refuelling water storage tank (IRWST) ....... 71
5.2.7 Emergency feed water system (EFWS) ........................ 71
5.2.8 Emergency power supply systems (EPSSs) .................... 72
5.2.9 EPR safety concept and containment system ................. 72
5.3 Russian Light Water Reactors .......................................... 72
5.3.1 Main design characteristics ........................................ 72
5.3.2 Safety concept of VVERs ............................................ 73
5.4 Boiling Water Reactors (BWRs) ....................................... 73
5.4.1 Core, Pressure Vessel and Cooling System .................... 75
5.4.2 The SWR-1000 inner containment system .................... 78
5.4.3 Safety relief valve system .......................................... 79
7.3.2 Solidification of the HLWC by vitrification

7.3.3 Conditioning of solid HLW from reprocessing plants

7.3.4 Conditioning of solid organic waste from reprocessing plants, re fabrication plants and nuclear reactors

7.3.5 Conditioning of liquid organic MLW

7.3.7 Transport and Storage of HLW and MLW

7.4 Long Term Waste Disposal

7.4.1 Low level waste disposal without long-lived α-emitters.

7.4.2 Repositories for low heat producing HLW/MLW

7.4.3 Repositories for HLW in deep geological formations

7.5 Direct Disposal of Spent Fuel

7.6 Mixed Oxide Fuel Fabrication

7.6.1 MOX fuel re fabrication capacity in the world

7.7 The Uranium/Plutonium Fuel Cycle of Fast Breeder Reactors

7.7.1 Ex-Core time Periods of FBR Spent Fuel

7.7.2 Mass Flow in an FBR Fuel Cycle

7.7.3 FBR Spent Fuel Reprocessing

7.7.4 FBR Fuel Fabrication

7.7.5 Status of FBR Fuel Reprocessing and Refabrication

7.8 The Closed Nuclear U/Pu MOX Fuel Cycle for PWRs

7.8.1 Plutonium Recycling as Plutonium Uranium Mixed Oxide (MOX) Fuel in the SGR mode.

7.8.2 Plutonium incineration in PWRs during several recycling steps

7.8.3 Balance of plutonium inventories and incineration of plutonium

7.8.4 Neptunium and Americium generation in the SGR plutonium recycle scenario

7.8.5 Plutonium incineration in a MOX-PWR or FR burner or ADS strategy

7.9 Chemical separation (partitioning) of minor actinides

7.9.1 Joint chemical separations of plutonium and neptunium

7.9.2 Aqueous chemical separation of americium and curium

7.9.3 Chemical separation of Americium/Curium from the Lanthanides

7.9.4 Chemical separation of Americium from Curium

7.9.5 Pyro-metallurgical methods for the separation of Uranium, Plutonium and Minor Actinides

7.9.6 Fuel re fabrication for incineration of actinides

7.9.7 Intermediate storage of Curium

7.9.8 Incineration of minor actinides in nuclear reactors

7.9.9 References Section 7

8 The IAEA Safeguards System

8.1 Material Balance Measurements

8.1.1 Significant quantities of fissile materials and timely detection

8.1.2 Methods of Safeguards Techniques

8.1.3 Material Balance Areas (MBAs)

8.1.4 Advanced Safeguards Approaches

8.1.5 Safeguards Measurement Technologies
9.12.7 Nuclear explosive yield of hollow reactor-grade plutonium HNEDs ........................................ 237
9.12.8 Discussion of this results compared to those of Mark ................................................................. 238
9.12.9 Conclusions from the analysis of the explosive yields of
  HNEDs based on reactor-grade plutonium .................................................................................. 239
9.13 Categorization of different isotopic compositions of plutonium .................................................. 239
  9.13.1 Integral pre-ignition probability and nuclear explosive yield .................................................... 241
  9.13.2 Numerical evaluation of the integral probability for pre-ignition
       for different isotopic compositions of plutonium ........................................................................... 242
  9.13.3 The US test of 1962 with reactor-grade plutonium ................................................................. 246
  References Section 9 .................................................................................................................. 247

10 Thermal analysis of HNEDs at different levels of technology ......................................................... 251
  10.1 Definition of different levels of technology .................................................................................. 251
  10.2 Geometric dimensions for different levels of technology ......................................................... 252
  10.3 High explosives for different classes of technology ..................................................................... 253
  10.3.1 Low technology high explosives ............................................................................................. 253
  10.3.2 Medium technology high explosives ...................................................................................... 253
  10.3.3 Very high technology high explosives ...................................................................................... 254
  10.4 The one-dimensional conservative approach for the thermal analyses .................................... 255
  10.5 Temperature profile within an HNED .......................................................................................... 257
  10.6 Outer temperature at the casing of the HNED ............................................................................ 258
  10.7 Radial temperature distribution within the HNED for constant thermal conductivity ............. 259
  10.8 Radial temperature distribution in a bare solid Pu-sphere .......................................................... 261
  10.8.1 Comparison with IAEA definitions ......................................................................................... 264
  10.9 Temperature profile in an assembled HNED ................................................................................ 264
  10.10 Results of thermal analyses ...................................................................................................... 265
  10.10.1 Radial temperature profiles in an HNED with reactor-grade plutonium
       with an alpha-particle of 0.144 kW ................................................................................................. 265
  10.10.2 Radial temperature profiles for reactor plutonium from spent fuel with
       an alpha-particle heat power of 0.240 kW .................................................................................... 267
  10.10.3 Radial temperature profiles for reactor plutonium with an alpha-particle
       heat power between 0.375 and 0.562 kW .................................................................................... 268
  10.10.4 HNEDs with other implosion geometries .............................................................................. 269
  10.11 Conclusions for the results of the thermal analyses ................................................................. 270
  10.11.1 Low Technology ..................................................................................................................... 270
  10.11.2 Medium Technology .............................................................................................................. 270
  10.11.3 Common assessment of the neutronic and the thermal analysis .......................................... 272
  10.11.4 Limits of alpha-particle heat power for proliferation-proof plutonium ................................. 273
  10.11.5 Additional remarks on the low and medium-technology cases ........................................... 274
  10.12 Outside cooling of the HNEDs ..................................................................................................... 274
  10.12.1 Coolability of HNEDs ............................................................................................................. 274
  10.12.2 Metal strips of high thermal conductivity .............................................................................. 274
  10.12.3 Coolability of very high technology HNEDs .......................................................................... 275
  10.12.4 Effects of cooling low-technology and medium technology HNEDs ........................................ 275
10.13 Solution of the steady state and transient heat conduction problem
with temperature dependent thermal conductivities and specific heats.........276
10.13.1 Formulation of the heat conduction problem ..................................276
10.13.2 Numerical solution for the transient temperature distribution ...............277
10.13.3 Thermal conductivity and specific heat at cryogenic temperatures .............278
10.13.4 Specific heat data at cryogenic temperatures ...................................280
10.13.5 Cooling of low technology HNEDs by liquid nitrogen or liquid helium .......281
10.13.6 Numerical results for medium technology HNEDs ..............................283
10.13.7 Conclusions for low and medium technology HNEDs ..........................284
10.13.8 Technical difficulties for cooling by liquid nitrogen or liquid helium .........285
10.13.9 Numerical results for high technology HNEDs ..................................286
10.14 Steady state and transient temperature distributions for cooling of the HNED by internal rods of high thermal conductivity.................................288
10.14.1 Outline of the approximate method for determining the steady state temperature distribution in an HNED with cooling by aluminum rods ..........288
10.14.2 Calculated results for low technology HNEDs (steady state temperature profile) ..........................................................289
10.14.3 Transient temperature distribution if the aluminum rods will be replaced by high explosive material .................................................290
10.14.4 Technical difficulties ........................................................................291
10.14.5 Installing the reactor grade plutonium sphere prior to detonation ..........291
10.15 Conclusions ......................................................................................292
References Section 10 ..............................................................................293

11 Proliferation Resistance of Americium Originating from Spent Irradiated Reactor Fuel ..........................................................297

11.1 Introduction ....................................................................................297
11.2 Some nuclear physics data of the three americium isotopes Am-241, Am-242m and Am-243 .................................................................297
11.3.1 Am-241 from the decay of Pu-241 .....................................................301
11.3.2 Am-242m production ...................................................................299
11.4 Considerations on pre-ignition, alpha-particle heat power and critical mass of americium .................................................................301
11.5 Critical mass of reactor americium metal based Hypothetical Nuclear Explosive Devices .................................................................302
11.5.1 Gun type HNED with metallic americium ........................................302
11.5.2 Spherical implosion type HNED with metallic americium ..................303
11.6 Critical masses for gun type HNEDs and spherical implosion type HNEDS ..............................................................303
11.6.1 Critical masses for gun type systems ..............................................303
11.6.2 Critical masses for spherical implosion type systems .................304
11.7 Pre-ignition for reactor-americium based gun type and spherical implosion type HNEDs .................................................................307
11.7.1 Pre-ignition of metallic americium based gun type systems .............307
11.7.2 Results of pre-ignition analysis for gun type systems ......................308
11.7.3 Results of pre-ignition analysis for spherical implosion HNEDs .........309
11.8 Geometric dimensions, alpha particle heat power and material characteristics for the thermal analysis of spherical americium based implosion type HNEDs...309

11.8.1 Geometric dimensions of a reactor-americium based spherical implosion type HNED for the thermal analyses 309

11.8.2 Material properties for high explosives 310

11.9 Outside temperature of the reactor americium based HNED 311

11.9.1 Temperatures of a metallic reactor americium bare sphere and gamma radiation problems 311

11.9.2 Outside casing temperature of americium based HNEDs 312

11.9.3 Inside temperature profile in the americium based HNED 312

11.9.4 Radial temperature profile for a reactor-americium sphere HNED (option G, PWR) 313

11.9.5 Radial temperature profile for a reactor-americium HNED (Option H LMFBR) 314

11.9.6 Radial temperature profile for a reactor-americium HNED (option K, ADS) 315

11.9.7 Radial temperature profile for a reactor-americium HNED (option L, 100% Am-241) 316

11.9.8 Radial temperature profile for a reactor-americium HNED (option M Am-242m breeding) 317

11.10 Discussion of the results of the thermal analyses and uncertainties 317

11.11 Characteristics of material data 318

11.12 Coolability of the reactor americium HNED 318

11.13 Conclusions 319

References Section 11 320

12 Fuel cycle options for the production of denatured, proliferation-proof plutonium 323

12.1 Introduction 323

12.2 Review of earlier research 323

12.3 Analysis of fuel cycle options for the production of proliferation-proof plutonium 324

12.4 Fuel cycle options for the production of denatured, proliferation-proof plutonium 325

12.5 Results of physics calculations for the selected fuel types 327

12.5.1 Results for fuel type A; UO2 from reenriched recycled uranium 327

12.5.2 Results for fuel type B 327

12.5.3 Results for fuel type C 328

12.5.4 Results for fuel type D 328

12.5.5 Results for fuel type E; MOX fuel with thorium, uranium, plutonium and minor actinides 329

12.6 Moderator density and Doppler reactivity coefficients for the fuel type A, B, C, D, E 330

12.7 Long term behavior of denatured, proliferation-proof fuel in PWRs and FRs 331

12.7.1 Long term behavior of denatured plutonium in LWRs 331

12.7.2 Long-term irradiation behavior of denatured plutonium in fast reactors 332

12.7.3 Destruction of denatured fuel type C in a PWR 333

12.8 Peculiarities of the fuel cycle and of the PWR design for production and recycling of denatured proliferation-proof plutonium 333

12.9 Conclusions 335
13 Neptunium as a proliferation problem and fuel cycle options for avoiding neptunium production

13.1 Neptunium as a proliferation problem

13.2 Neptunium-free nuclear fuel cycle

13.2.1 Model of a neptunium-free nuclear fuel cycle

13.2.2 Future proliferation-proof, neptunium-free fuel cycles

13.3 Initial fuel composition for proliferation-proof plutonium and neptunium-free fuel cycles

13.4 Selection of fuel composition for proliferation-proof plutonium and neptunium-free fuel cycles

13.5 Reactivity coefficients relevant to PWR safety

13.6 Peculiarities and technical modifications required for the PWR design

13.7 Conclusion for incineration of proliferation-proof plutonium in PWR cores

13.8 Fast reactor fuel cycle for utilizing americium as well as denatured proliferation-proof plutonium but avoiding neptunium production

13.8.1 Results of the FR core calculations

13.8.2 Plutonium incineration and breeding

13.8.3 Americium incineration and production of curium

13.9 Conclusion for FRs operating with proliferation-proof plutonium and americium, but avoiding neptunium

References

14 Future civil proliferation-proof fuel cycles

14.1 Introduction

14.2 Plutonium incineration by a multi-recycling strategy

14.3 Needed capacity of reprocessing and Pu/U refabrication plants

14.4 Fuel cycle plant capacity in the world in 2010

14.5 Transition phase for the production of proliferation-proof plutonium

14.6 Different levels for non-proliferation criteria of reactor-grade plutonium

14.6.1 Scientific proposal for level I criterion for non-proliferation

14.6.2 Scientific proposal for level II criterion for non-proliferation

14.6.3 Alpha-particle decay of Pu-238 in proliferation-proof reactor-grade plutonium

14.7 Can proliferation-proof plutonium be converted to weapon grade plutonium

14.7.1 Centrifuge enrichment technology

14.7.2 Decomposition of PuF6 by alpha-particle radiation

14.7.3 Atomic vapor Laser isotope enrichment

14.8 Future civil Pu/U fuel with proliferation-proof, reactor-grade plutonium

14.8.1 Incineration of proliferation-proof, reactor-grade plutonium in PWRs

14.8.2 Incineration of proliferation-proof reactor-grade plutonium in FRs

14.8.3 Future international proliferation-proof nuclear fuel cycles

14.9 Effect on Safeguards and Non-proliferation Issues in Future Civil Uses of Nuclear Power of the Proposed Concept of Upper Limits for Non-proliferation

14.9.1 Not Included In This Proposal

References Section 14