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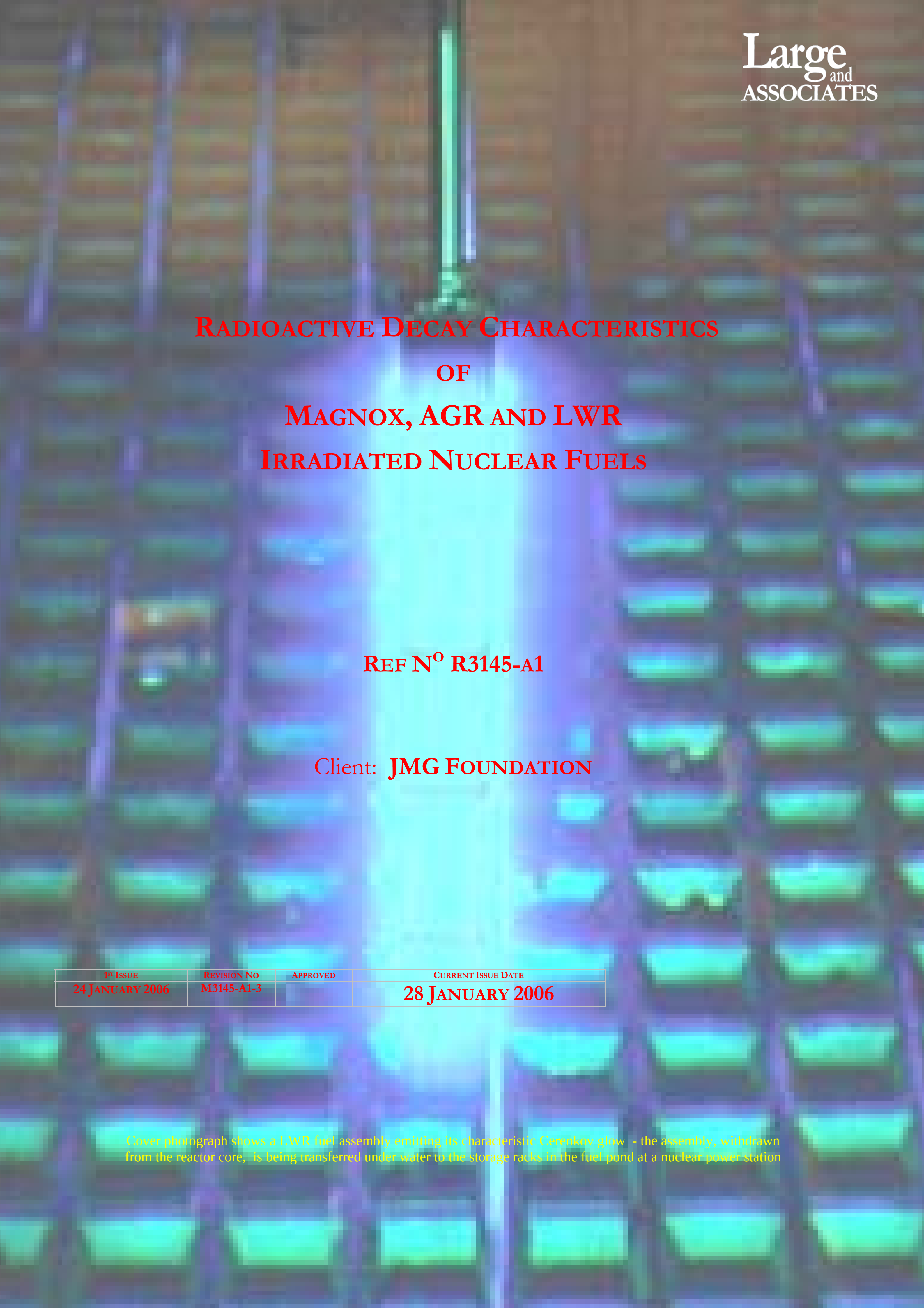

RADIOACTIVE DECAY CHARACTERISTICS OF MAGNOX, AGR AND LWR IRRADIATED NUCLEAR FUELS

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Cover photograph shows a LWR fuel assembly emitting its characteristic Cerenkov glow - the assembly, withdrawn from the reactor core, is being transferred under water to the storage racks in the fuel pond at a nuclear power station

RADIOACTIVE DECAY CHARACTERISTICS FOR MAGNOX, AGR AND LWR IRRADIATED FUEL

RADIOACTIVITY

The nuclear processes underway within the fuel core of a nuclear reactor bombard the atomic structure of the uranium fuel with neutrons. The collision and absorption of a neutron within a whole uranium atom, splits the uranium and creates a number of fission products and frees more neutrons which enable the nuclear chain reaction to prosper.

The isotope of uranium that is principally responsible for fission is uranium-235 which is present in naturally abundant uranium to the extent of 0.715%, with the remaining uranium being U-238 which is less fissionable. The fission process with U-235, which is also possible with another four different isotopic elements, commences with the absorption of a neutron in U^{235} forming U^{236} at an excited and unstable state. In some instances the U^{236} emits gamma radiation and returns to a stable state but the majority of absorptions result in the U^{236} nuclear splitting or fissioning, yielding two fission fragments whose mass numbers varies between about 70 and 160, giving a range of (radioactively) unstable elements (caesium, ruthenium, etc), together with a number of neutrons, beta particles and gamma radiation, neutrinos and energy. It is the majority of these fission fragments or products that are eventually separated out as high-level radioactive waste.

Increasing the amount of U^{235} present in the fuel increases the number of fissions available before the fuel becomes too depleted to function. The Magnox reactor fuel is not enriched and comprises just U^{235} at the natural level of ~0.7%, whereas the advanced gas-cooled reactor AGR and light water reactor fuels (pressurised water reactor PWR and boiling water reactor BWR) utilise enriched uranium where the content of U^{235} is increased to 2 to 3.5% with the level of enrichment differing in various regions of the reactor core.

Almost all of the species of fission fragments are unstable, losing energy by emitting from the unstable nuclei some combination of alpha, beta and gamma rays, and by internal conversion which emits X-rays. This process eventually leads to a stable or grounded isotope by a random process which characterises the individual radioisotope by half-life which, according to the particular fission fragment or radioisotope ranges from a few seconds (Radon Rn^{220} 55 seconds), to hours (Technetium Tc^{99m} 6 hours), days (Iodine I^{131} 8.05 days), years (Caesium Cs^{137} 30 years) and some actinides yield by the absorption of a neutron in U^{238} tens of thousands of years (Plutonium Pu^{239} 24,400 years). The half-life indicates for how long the initial (radio)activity of each radioisotope will take to halve, and thereafter to reduce to one quarter, one eighth and so on.

The radioactive decay is the measure of the number of nuclei decaying over a set time, taken as per second. This is measured in units of the Becquerel (Bq), or in the past, the Curie (Ci), with 1 Bq being one decay per second and a Ci being equal to 3.7E+10 Bq (3,7000 million Bq). The original Curie unit was set by the number of nuclei disintegrations occurring in 1 gram of radium, so 2 grams of radium will have 2 Ci of activity, and so on.

In summary, each fission product species is characterised by its elemental chemistry, its (radio)activity and its natural decay half-life. At any one time, the activities of any number of fission products can be summed together on the basis of their respective quantities, and the way in which any mix of fission products will decay over future time can be determine by account of the individual half-lives.

FUEL INVENTORY

Fission products are retained in the fuel whilst it is in the reactor core by the cladding or sheathing around the individual fuel rod or pellets. The longer the fuel stays in the core the greater the amount of energy in the form of heat liberated to the reactor cooling system, and the greater the amount of fission products held in the fuel pins.

The crude measure of this is fuel *burn-up* which essentially relates to the extent of irradiation (nuclear absorption), that is a product of the neutron flux in the reactor core and the total time that the fuel has been subject to irradiation. The unit of fuel burn-up is conventionally related to the electrical energy produced, being expressed as MWd/tU (MegaWatt.day per tonne of uranium) and which is usually given as an average burn-up for the whole fuel core. For the fuel inventories examined here the following *average* fuel characteristics and burn-ups have been assumed:

Table 1 Reactor and Fuel Characteristics

REACTOR TYPE	UNITS	MAGNOX	AGR	LWR
Fuel	U^{235} %	U^{nat}	2.25	3.05
Burn-Up	MWd/tU	3,600	18,000	33,000
Load Factor	%	80	75	75
Efficiency	%	26	41	32
U^{nat} per GW_eyear	tU/GW_eyear	400	180	200
Reprocessing Delay	year	4.5	4.5	4.5
Fuel Throughput 1 GW_eyear	tU	400	50	35

The burn-up and fuel utilisation figures given in Table 1 relate to fuels in operation for the 1980s through to the mid-1990s. Since that time, fuel enrichment and burn-up have increased with present day PWR fuel achieving 45,000+ MWd/tU, although for the UK Magnox and AGR reactor systems fuel utilisation improvement has not been that significant. The 3rd generation of light water reactors, the AP 600/1000 and the EPR, aim to achieve fuel burn-ups in excess of 65,000MWd/tU

In the UK high-level radioactive waste arisings from fuel reprocessing (a process that separates out fission products, depleted uranium and plutonium) and vitrification (where the fission product waste is mixed and solidified with borosilicate glass), now awaiting interim and longer term management/disposal, derives from the Magnox and AGR fuel consistent with the fuel characteristics of Table 1. Spent fuel from the Sizewell B PWR is presently maintained on-site at Sizewell and there is no contracted reprocessing option for this fuel. Overseas PWR fuel reprocessing HLW vitrified waste contracted post-1976 is committed for return to the overseas customer, and it is only the lower burn-up pre-1976 overseas PWR reprocessed fuel that will have to be managed and eventually disposed of in the UK, with this fuel being generally consistent with the characteristics given in Table 1.

RADIOACTIVE DECAY OF MAGNOX, AGR AND LWR SPENT FUEL

Figures 1, 2 and 3 are for the Magnox, AGR and LWR fuels specified in Table 1 respectively.

The decay characteristics are for the dominant and longer-lived fission products and actinides remaining in the fission product wastes derived from centrifuge and chemical separation stages of reprocessing.[1] The total reprocessing fission product yield for conversion to vitrified glass is also shown on each characteristic.

The fuel decay characteristics are expressed in terms of the amount of fuel (tonnes of uranium) required by each reactor type to generate 1 GWe year of electricity – there may arise some variation because of plant-to-plant efficiency and availability factors. The decay time in years (logarithmic horizontal scale) extending from the time that the fuel was withdrawn from the reactor core and includes the 4 to 5 year power station cooling pond dwell to reprocessing (usually much longer for the earlier fuels) and a one year delay until vitrification, thereafter the vitrified waste might be dry stored for tens or hundreds of years before final disposal to a deep geological repository. The logarithmic vertical scale gives the radioactivity of the dominant radioisotopes and for the overall reprocessed vitrified fission waste in TBq (Tera Bequerel or 1E+12 or a million million Bq).

As shown by Figure 4, generally, the overall decay characteristic for the vitrified waste arising from 1 GWyear of electricity generation is about the same for all three reactor types considered here, although the fuel mass varies considerably (see Table 1). Depending on the reprocessing and vitrification processes used on the fuel, the total mass (in tonnes U) of AGR and LWR fuels would be expected to reduce to an equivalent volume of 0.1 to 0.2m^3 of vitrified waste per 1 tonne of fuel (tU). Because of the high uranium content, it is likely that the early Magnox fuel HAL (high activity liquor) yielded by reprocessing will be not be vitrified so, approximately, 500 to 600m^3 of Magnox cannot at this time be considered for geological disposal.

The fission product vitrified wastes are held in stainless steel canisters of 430mm diameter and 1,350mm overall height, in account of ullage space each sealed canister contains about 0.150m^3 of glass mix and total weight about 500kg. Projection of canister containment life is between 300 to a somewhat optimistic maximum of 10,000 years, with a mean life assumed of 5,000 years with corrosion being the dominant failure mechanism. Canister life in the deep geological environment is expected to be extended by overpackaging of the vitrified waste canister with, for example as proposed by the Swedes SKB, with a stout copper cylinder which will accept both vitrified waste canisters or untreated fuel.

Following eventual canister failure, the leach rate for the (borosilicate) glass leads to total dissolution of the glass matrix and release of the fission product contents within a further 5,000 years. Canister failure times and vitrified waste dissolution may be accelerated by a number of long term thermodynamic processes, particularly if the container or overpack (if used) is slightly flawed. The range of canister failure and glass dissolution time scales (5,300 to 15,00 years) are shown on Figure 4 for comparison, indicating the timescales over which the nature geological barriers of a deep repository have to become effective and for which the radioactivity has to be maintained isolated from mankind's environment.

Also superimposed on Figure 4 are the levels of (radio)activity and times at which the vitrified waste forms would be expected to reach the today's UK specific activity limits for the mandatory clearance of radioactively contaminated land (400kBq/kg). In other words, at today's standard the fuel vitrified waste could only be left untreated at about 100 million years hence, although extreme caution should be adopted when applying such an extrapolation so far into the future.

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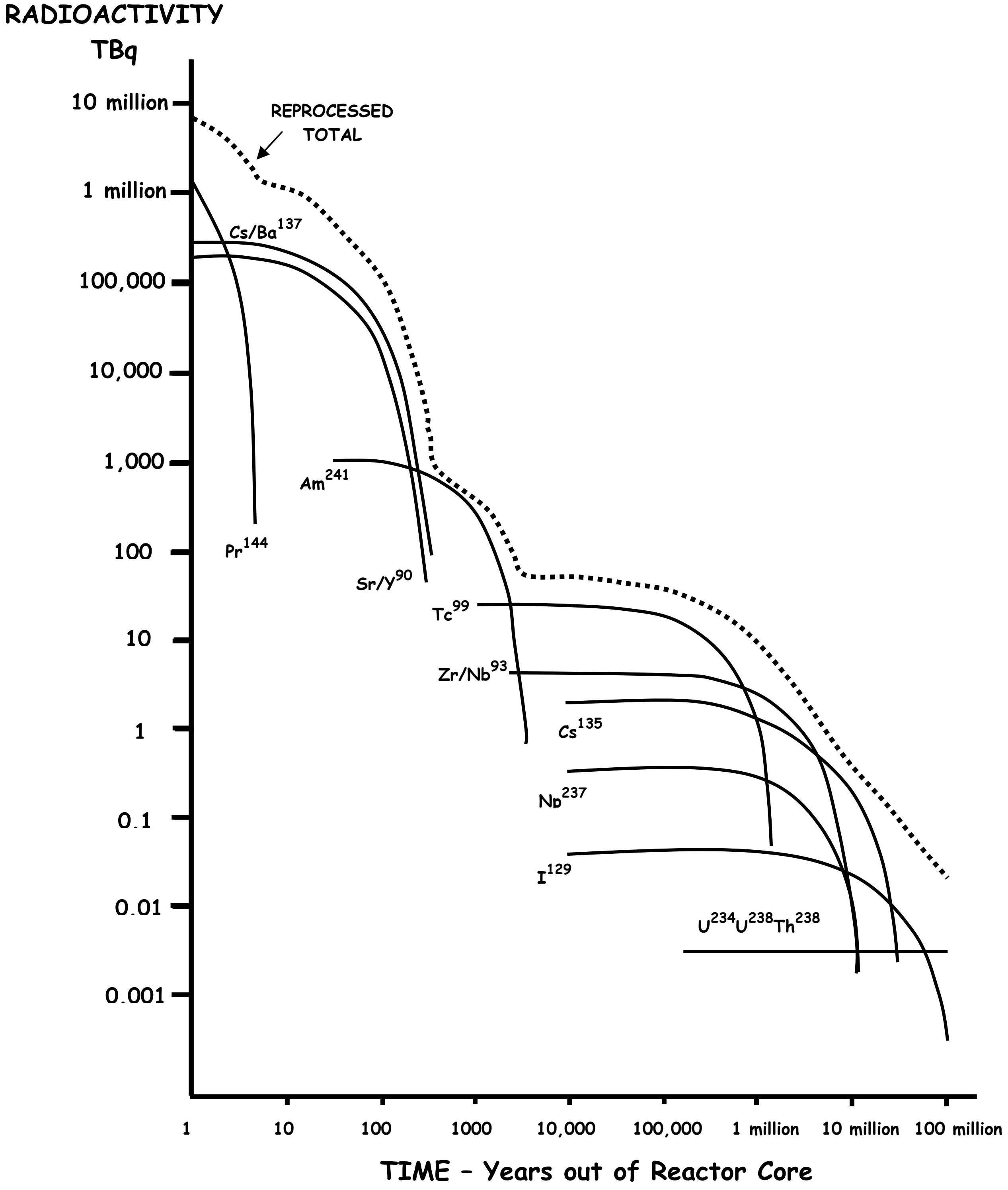
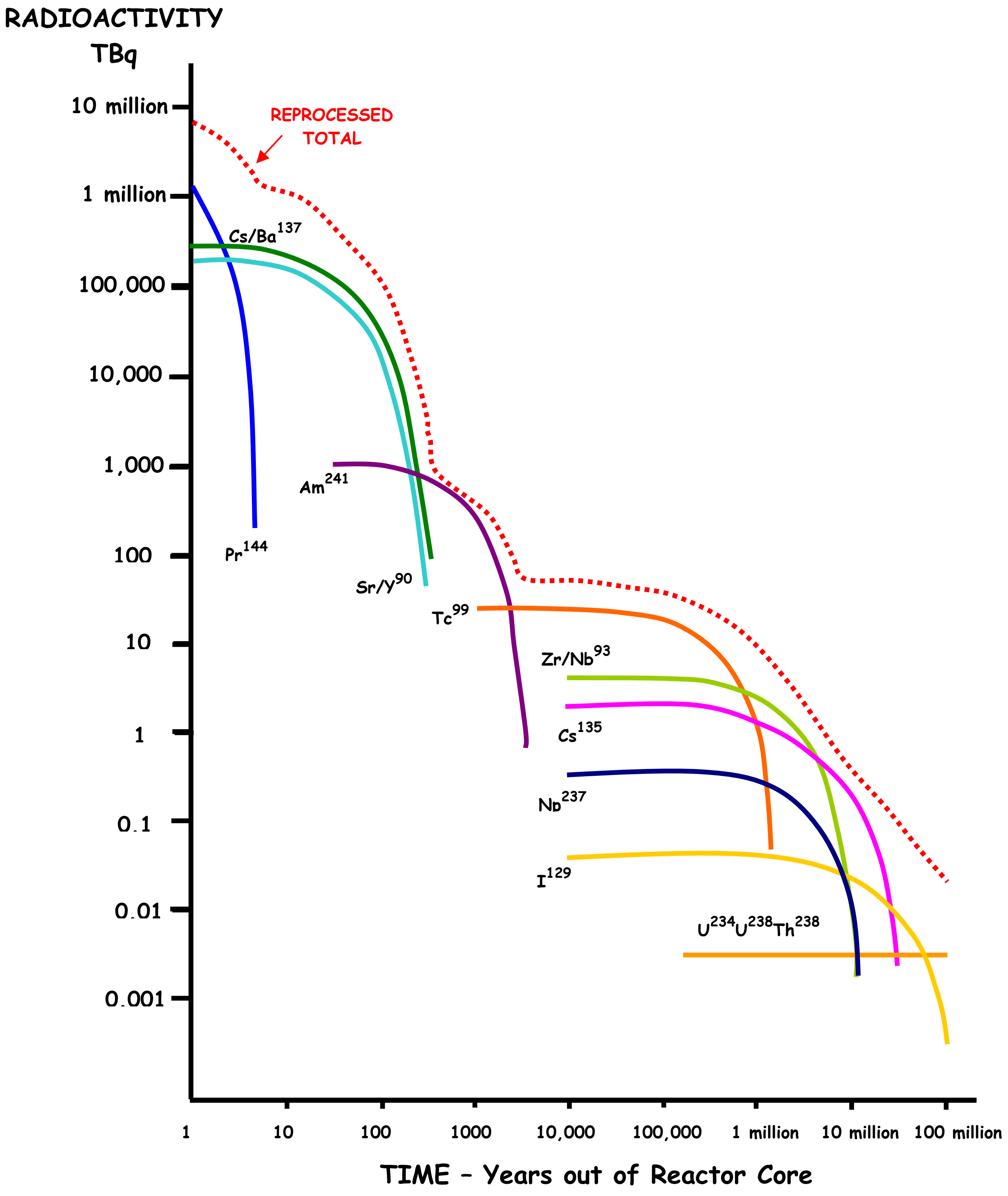
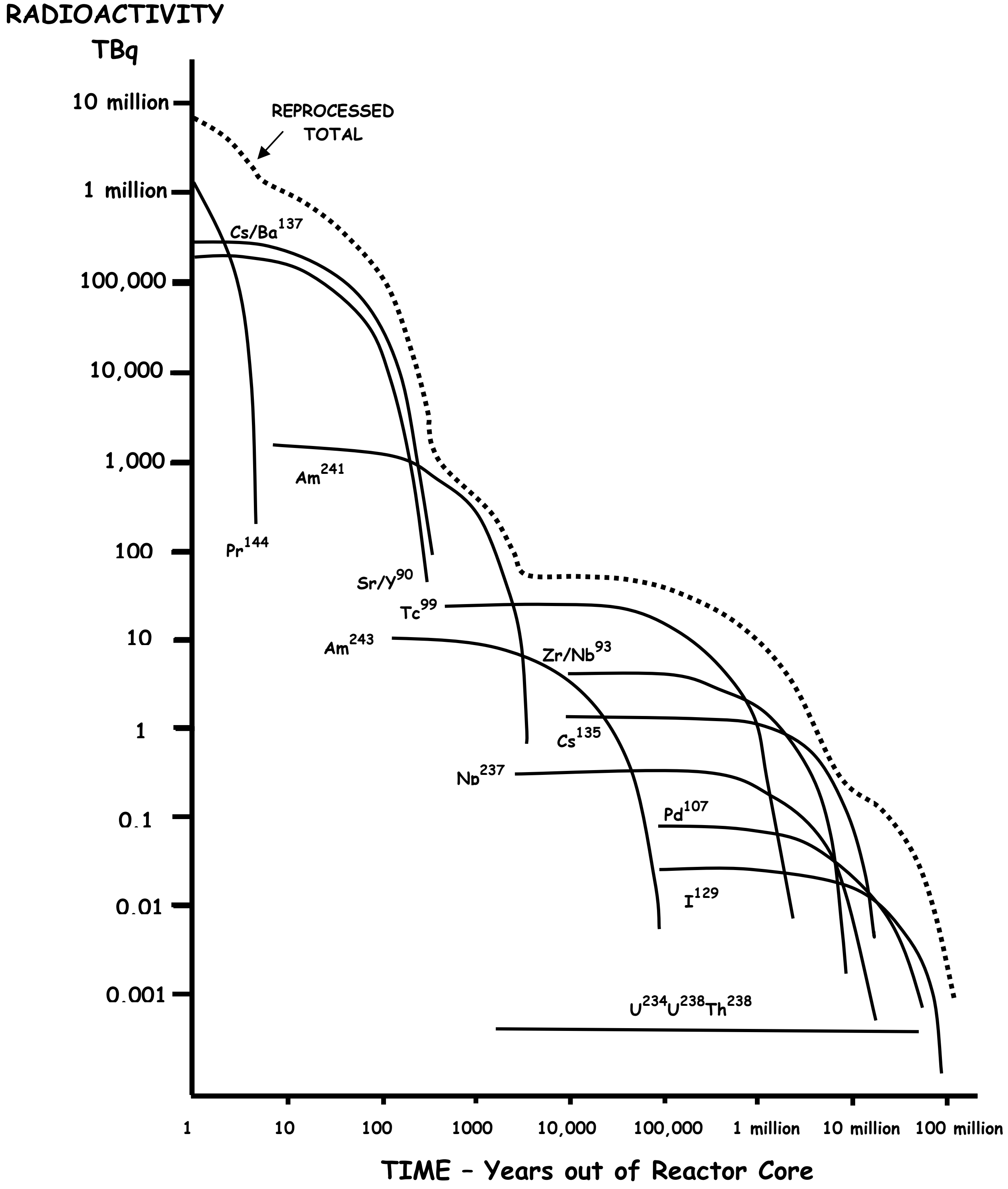
[1] Short-lived fission products (where the decay is significant during the 4.5 year defuel to reprocess dwell time assumed), activation products and, more generally, low and intermediate levels wastes arising during reprocessing, vitrification, etc are not included.



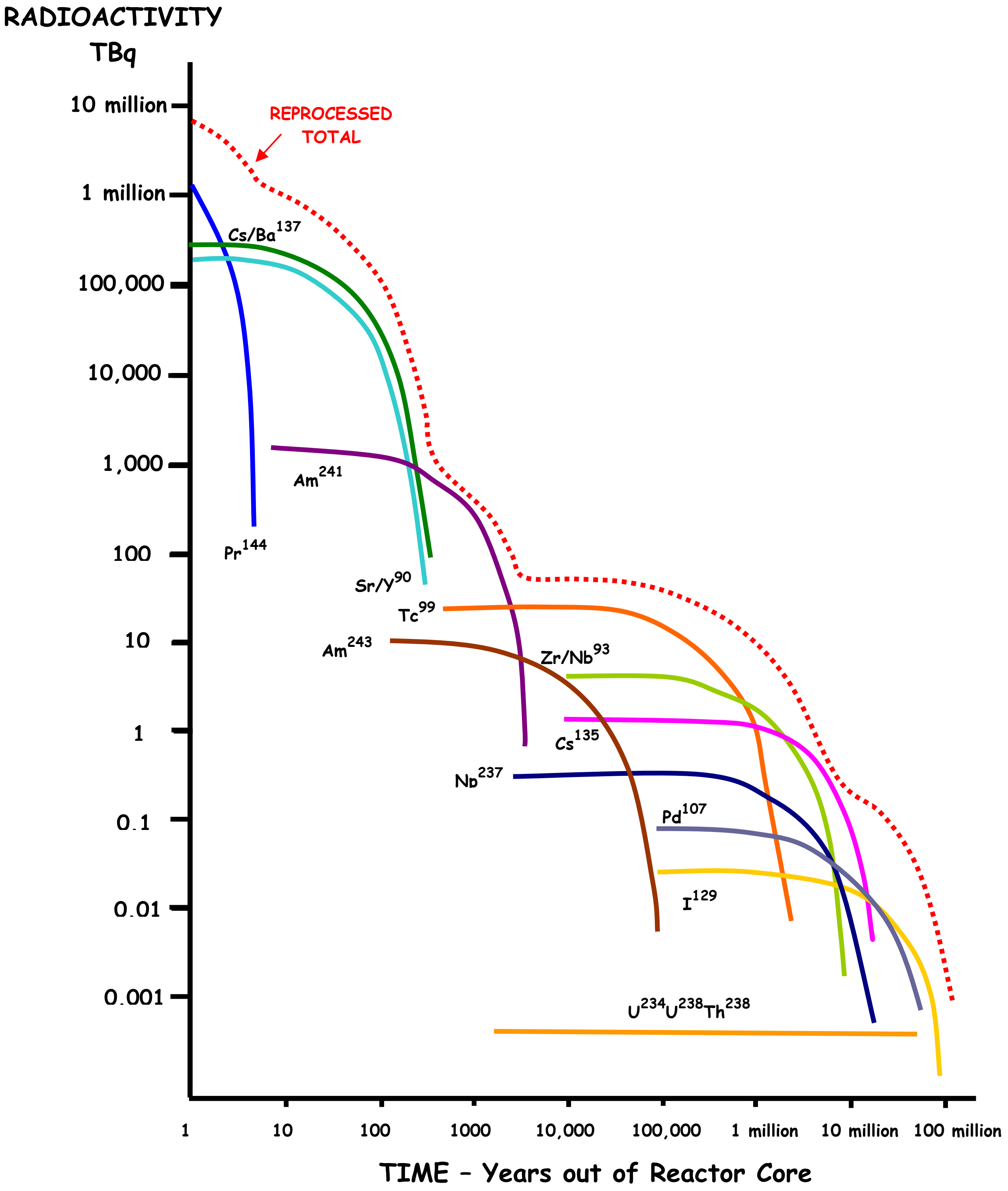
1) Total & Individual Fission Product Decay Characteristics – Magnox Reactor Fuel per 1 GWy(e) Generation – 3.6GWday/teU



1) TOTAL & INDIVIDUAL FISSION PRODUCT DECAY CHARACTERISTICS – MAGNOX REACTOR FUEL per 1 GWY(e) GENERATION – 3.6GWday/teU



2) Total & Individual Fission Product Decay Characteristics – AGR Fuel per 1 GWy(e) Generation – 18GWday/teU



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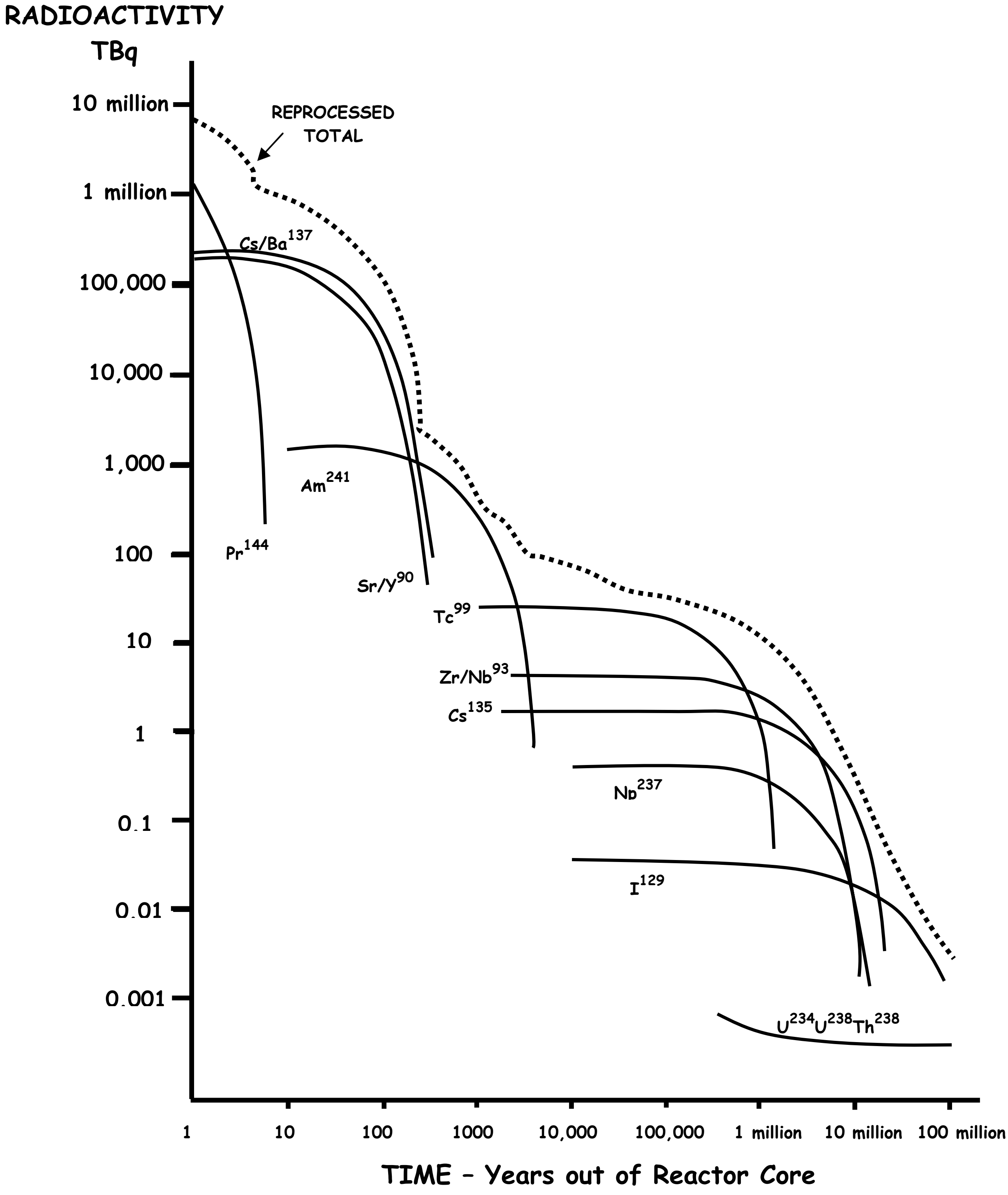
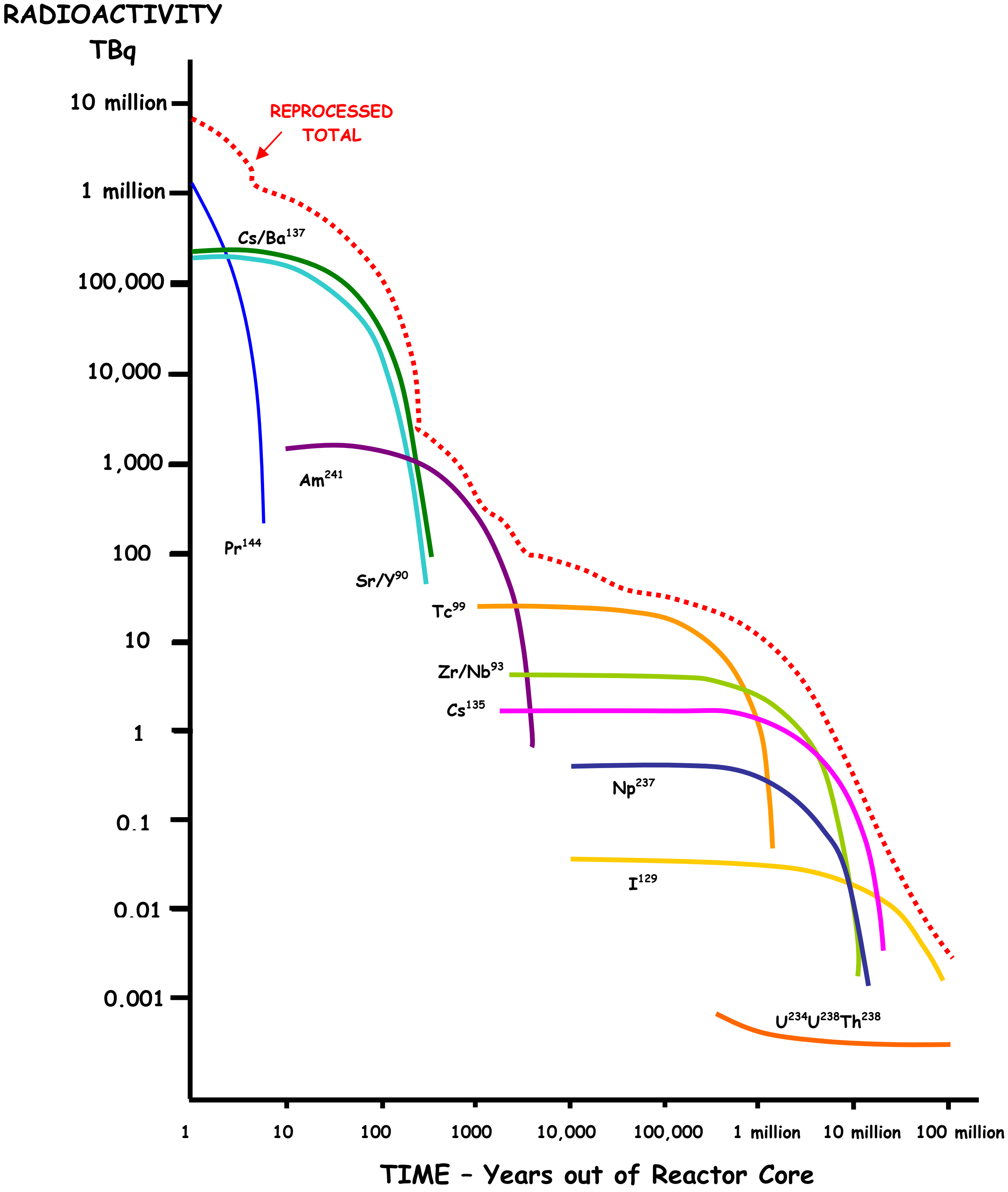
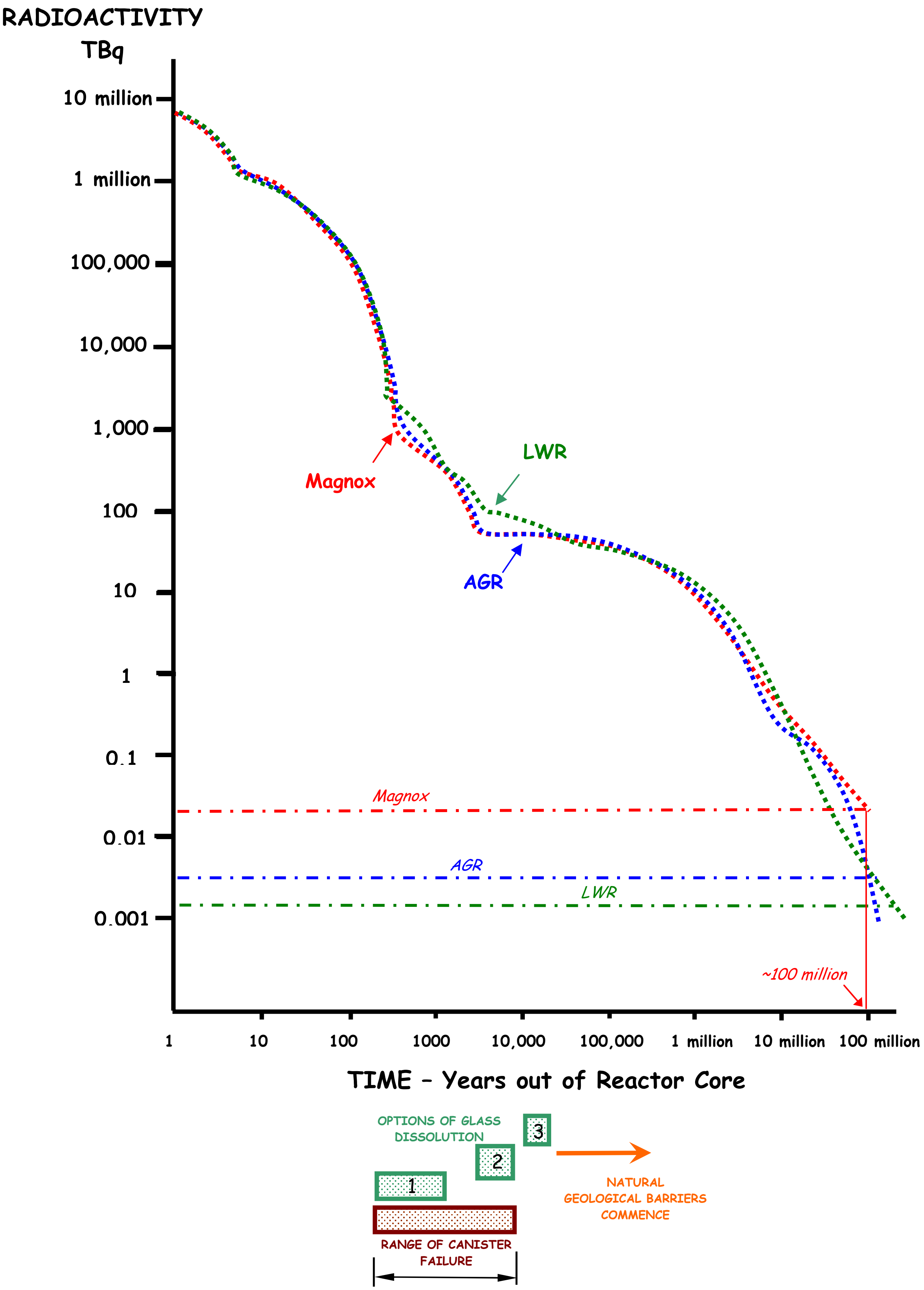


3) Total & Individual Fission Product Decay Characteristics – Light Water Reactor Fuel per 1 GWy(e) Generation – 33GWday/teU



3) Total & Individual Fission Product Decay Characteristics – Light Water Reactor Fuel per 1 GWy(e) Generation – 33GWday/teU



4) Total Fission Product Decay – Magnox/AGR/LWR Vitrified Waste per 1 GWy(e) Generation