#### Nuclear Fuel Waste Consumed and Eliminated: Environmentally Responsible, Economically Sound, Energetically Enormous CCTC 2013 Paper Number 1569693995

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#### Abstract

Fast-neutron reactors (FNRs) refuelled with the world's stored used nuclear fuel waste and depleted uranium would create \$ 2,300 trillion non-carbon electricity (\$ 320,000 for every person on earth). Compared to using coal to produce electricity this approach would avoid the release of 19 trillion tonnes of  $CO_2$ , 6.4 times the current  $CO_2$  content of the atmosphere. This course also eliminates the million-year radiotoxicity of used reactor fuel. The technology exists today with appropriate commercial FNRs now available. The approach would have a major global mitigating impact on the  $CO_2$  component of GHG emissions into our atmosphere.

Keywords: GHG reduction, energy security, nuclear waste elimination, fast-neutron reactor

#### Résumé

Réacteurs à neutrons rapides (RNR) ravitaillent avec le stock des déchets de combustible nucléaire et l'uranium appauvri du monde créerait 2,300 billions de l'électricité non-carbone (320.000 \$ pour chaque personne sur terre). Par rapport à l'utilisation du charbon pour produire de l'électricité cette approche permettrait d'éviter la libération de 19 milliards de tonnes de CO<sub>2</sub>, soit 6,4 fois la teneur actuelle de CO<sub>2</sub> dans l'atmosphère. Ce cours serait également éliminer complètement la radiotoxicité de millions d'années du combustible utilisé du réacteur. La technologie existe aujourd'hui avec appropriées RNR commerciaux actuellement disponibles. L'approche aurait un impact majeur sur l'atténuation de la composante CO<sub>2</sub> des émissions de GES dans l'atmosphère.

**Mots clés** : Réduction des GES, la sécurité énergétique, l'élimination des déchets nucléaires, réacteurs à neutrons rapides

#### 1. Introduction

Three of the major concerns facing the world are the disposal of highly radioactive nuclear fuel waste, a potential energy crisis as fossil fuels resources become more scarce and costly in a decade or three, and global warming caused in part at least by the introduction of greenhouse gases (GHGs) into our atmosphere from the production of energy from such fossil fuels. Somewhat curiously, it is a solution to the first concern, the productive elimination of highly radioactive nuclear fuel waste using fast-neutron reactors, that can simultaneously have a major impact on preventing a future energy crisis and on reducing GHGs for energy creation.

The world of nuclear nations over the last 60 to 70 years has accumulated about 370,000 tonnes of used reactor fuel, 45,000 tonnes of which are stored at reactor sites in Canada, and 68,000 tonnes at various sites in the U.S.A. [1-4]. Since this used fuel is highly radioactive, and

will remain so for close to one million years [5, p. 341], much thought has been given to its safe management, generally directed at its disposal in deep geological repositories. In addition, almost 2 million tonnes of depleted uranium have been stored, that were created as part of the production of fuel enriched in the fissile (splittable) isotope uranium-235 (U235), that is required in light-water cooled and moderated reactors, the most prevalent nuclear power reactors [6].

It has been known since the beginnings of nuclear power in the 1950s and 60s that all of these heavy atoms in stored used fuel and in depleted uranium can act as fuel in fast-neutron reactors [7, p.3; 8,p.1; 9, p.2]. This not only yields gargantuan amounts of non-carbon energy but also eliminates the long-term radiotoxicity of the used nuclear fuel completely, reducing the 400,000-year lifespan of the hazardous material to tractable safe storage of no more than 300 years.

This report outlines an alternative to the disposal of used nuclear fuel waste in a deep geological repository. The approach, using fast-neutron reactors (FNRs), would eliminate the long-term radiotoxicity of the waste. By consuming all of the heavy atoms in the waste as well as the stored depleted uranium, FNRs would extract close to 200 times the massive amounts of nuclear energy that uranium has already yielded in all reactors since the beginning of civilian nuclear power. Moreover, since all of this fuel is currently stored and available, its energy yield would produce no greenhouse gases. The equivalent energy derived from coal would create 19.6 trillion tonnes of carbon dioxide, more than 6 times the current total carbon dioxide content in the atmosphere.

Fuel replenishment of FNRs can be done with all heavy-atom actinides. For that reason the separation of any specific atom such as plutonium or its isotopes from used nuclear fuel waste is completely unnecessary, indeed unwanted, making the cycling of FNR fuel very much proliferation resistant. Moreover, a fuel cycling strategy is outlined that gives priority to the use of the long-lived radioactive transuranic actinides (TRUs) in the used fuel, substantially accelerating the elimination of this hazardous component.

The type of FNR envisioned for use is similar to the EBR-II [9], a reactor that has been tested and shown to have a degree of safety that would have avoided the Fukushima, Three-Mile-Island and Chernobyl accidents. Such an FNR based on the EBR-II design is now available commercially as the PRISM from GE-Hitachi in the USA.

## 2. Nuclear waste 101

Water-cooled reactors, the current major nuclear power sources, use thermal, or slow, neutrons to extract energy from the fission, or splitting, of heavy atoms. Such reactors rely on a minor component of uranium, the isotope U235, to deliver and maintain their power. Only 0.72% of natural uranium is U235; the remaining 99.28% is virtually all U238, an isotope that yields its energy about 50 million times less often in such reactors. When the U235 is split by a neutron in the reactor, it yields energy and breaks into two smaller atoms, fission products, most of which are stable atoms while about 30% are radioactive. A small proportion of the U238 in the reactor is converted to heavier radioactive atoms, isotopes of neptunium, plutonium, americium, curium, etc., the long-lived TRUs. A few of these isotopes can also be fissioned in a thermal reactor to yield energy. However, when most of the U235 is used up, the reactor has to be refuelled. The spent fuel is considered nuclear fuel waste.

For heavy-water cooled and moderated CANDU reactors, which are about 50% more efficient in the use of neutrons than light-water cooled reactors, refuelling occurs with unenriched natural uranium when 0.74% of the heavy atoms are used up [5, p. 341]. (For light water reactors this percentage drops to about 0.5% of mined uranium [3,6].) At that point the used CANDU fuel contains 0.74 wt% fission product atoms with atomic numbers from 35 to 65, a range that includes platinum group metals rhodium and palladium and also the increasing scarce and expensive rare earths. The remaining 99.26 wt% of the fuel is still primarily uranium, plus a small percentage of TRUs that nevertheless determine the long-lived radiotoxicity of all used nuclear fuel.

# 3. Reduction of long-term radiotoxicity

Spent fuel from CANDU reactors can be arbitrarily divided into three components: uranium, the fission products (FPs), and the transuranic actinides (TRUs), the latter two being products with quite different characteristics created inside the reactor.

The uranium in spent fuel is chemically and radiologically very similar to natural uranium. It is still largely U238 at 98.6%, with U235 depleted to 0.23% and 0.07% U236 created from neutron absorption of U235 [5, p. 341]. Between 70% and 80% of the FPs are non-radioactive [3,6]. For the other 20% to 30% FPs the radioactivity consists primarily of beta rays (emission of electrons) and gamma rays (X rays emitted from the nucleus). The TRUs and uranium predominantly emit alpha rays (high-energy particles that are helium nuclei) as well as gamma rays. Of these emissions, alpha particles are about 20 times more injurious than either electrons or gamma rays of the same energy [10]. Adjusted for both energy and biological effect, Fig. 1 shows the change in time of the radiotoxicity of the fission product and TRU components in used CANDU fuel relative to that of natural uranium. The fission product





Elimination of the transuranics (TRUs) in fast-neutron reactors (FNRs) and extraction of uranium (dashed horizontal line) would result in a huge reduction in radiotoxicity of used fuel waste, about 42,000X per unit time at 1000 years, and also shorten the time of decay to background levels of natural uranium from 400,000 years to 265 years. After 265 years the radiotoxicity of the fission products would be lower than that of the mined natural uranium from which they are created in the reactor. Note the log scales of both axes.

radiotoxicity decays to levels below that for natural uranium in less than 300 years, whereas the TRU radiotoxicity starts about 1000 times higher than that of uranium and remains above uranium levels for about 400,000 years. This long-term radiotoxic hazard of the used nuclear fuel is a major worldwide concern.

The heavy atoms in the spent fuel, including the hazardous TRUs, can all be fissioned in fastneutron reactors (FNRs) and converted to fission products. Therefore FNRs provide a means to eliminate the long-term radiotoxic hazard of the TRUs, reducing the radiotoxic lifespan of the resulting waste from 400,000 years to the 300 years of the fission products (Fig. 1).

The FPs, with their 300-year radiotoxic lifespan after they have left the fast-neutron reactor, still demand protective storage. Their storage for such a time-span is tractable. Moreover, such storage should permit easy retrieval for the FPs. After 300 years they are effectively non-radioactive and would constitute a valuable source of mineral atoms, such as rhodium, palladium and rare earths, altogether worth about \$2.5 million per tonne and extractable then by ordinary means [11].

# 4. Non-carbon energy from nuclear fuel waste: Canada and the world

## 4.1 Value of electricity

The major benefit of consuming spent fuel in fast-neutron reactors is clearly the more than onethousand-fold reduction in long-term radiotoxic lifespan of the fuel waste with the elimination of the TRUs. The future value of the FPs is a bonus.

There is, however, a third benefit to such a use of FNRs: consuming the stored spent CANDU fuel brings with it an over 130-fold increase in the yield of non-carbon energy over and above the huge energy that has already been extracted in CANDU reactors.

As reference, in 2008 CANDU reactors used 1390 tonnes of uranium to create \$ 11.4 billion electricity calculated at the current Ontario mid-peak time-of-use consumer price of 9.9 ¢/kWh [12,13]. Only 0.74% of the heavy atoms in the 1390 tonnes were fissioned to achieve this. In an FNR the remaining 99.26% heavy atoms could also be fissioned for an additional energy yield that would be 134 times bigger. Moreover, if the currently stored 45,000 tonnes of CANDU spent fuel were consumed in FNRs the yield would be increased a further 32 times over the 1390 tonne total. Therefore compared to the \$ 11.4 billion in 2008 the energy yield would be increased 4340 times to a total yield of \$ 49.4 trillion electricity, plus cogenerated heat.

This number is staggeringly large. Canada's stored spent CANDU fuel consumed in fastneutron reactors has the capacity to yield \$ 1.4 million non-carbon electricity for every one of Canada's current 35,002,447 inhabitants [14].

Worldwide the total stored spent nuclear fuel plus depleted uranium amounts to 2.07 million tonnes [2,3,6]. Used and cycled in FNRs this would produce an almost unfathomable \$ 2.27 quadrillion worth of non-carbon electricity. With the world population in February of this year being 7.065 billion [15], this stored fuel would produce \$ 322,000 non-carbon electricity for every person on earth.

#### 4.2 Carbon dioxide avoidance

Each MW-hour of electricity produced by nuclear power in Canada avoids 0.85 tonnes of carbon dioxide compared to the same amount of electricity produced by coal (0.5 tonnes if compared to natural gas) [16]. Since 6.25 fuel bundles, with a total of 120 kg of uranium oxide fuel (105 kg uranium), are used in Canada's CANDU reactors to create 1 MW-year or 8760 MW-hours of electricity [5, p. 351], those 120 kg of fuel would have avoided creating 7446 tonnes carbon dioxide from coal.

Those same considerations indicate that the 45,000 tonnes of uranium in currently stored used CANDU fuel have avoided releasing a potential 3.2 billion tonnes of carbon dioxide into the atmosphere. Nevertheless, this pales in comparison to the 134-fold greater potential that still exists if these 45,000 tonnes are used as fuel in fast-neutron reactors. The total carbon dioxide avoided then would become 424 billion tonnes of  $CO_2$ , equivalent to 13.7% of the current carbon dioxide content of the atmosphere [17,18].

Table 1 summarizes these results and also indicates that the combined 2.07 million tonnes of currently stored spent nuclear fuel in the world and the stored depleted uranium, if used as fuel in fast-neutron reactors would produce enough non-carbon electricity to avoid creating 19.7 trillion tonnes of carbon dioxide compared to the use of coal. This amount is 6.4 times the weight of 3.1 trillion tonnes carbon dioxide currently in the atmosphere [17,18].

Table 1           Potential Value of Electrical Energy Created and Tonnage of Carbon Dioxide Avoided Using					
Stored Nuclear Fuel Waste and Stored Depleted Uranium in Fast-Neutron Reactors					
<u>Canada</u>	current	potential FNR fuel	<u>Electricity</u> created value*		<u>CO</u> 2 avoided (toppes)
	<u>ruor uougo</u>	dougo	oroatoa	<u>value</u>	((011100)
			MW-h	\$ trillion	trillion
used uranium fuel 45,000 tonnes	0.74%	100%	499x10 <sup>9</sup>	49.4	0.424**
<u>World</u> used uranium fuel 370,000 tonnes depleted uranium 1,700,000 tonnes total 2,070,000 tonnes	0.5 %	100%	2.32x10 <sup>12</sup>	2293	19.7
<ul> <li>* Based on Ontario mid-peak time-of-use consumer price of 9.9 ¢/kWh [11]</li> <li>** Carbon dioxide content of atmosphere = 3.09 trillion tonnes [16,17]</li> </ul>					

## 5. Fast-neutron reactors and fuel cycling

Research FNRs have been built since the 1950s in England, France, Germany, Russia, India, Japan and the US. Russia also has commercial FNRs. Past and present FNRs predominantly used uranium/plutonium oxide fuels, achieving a fuel utilization of 10 to 11 per cent [19-21]. Used fuel was normally not recycled, since fresh uranium fuel was and remains relatively inexpensive. Moreover, used fuel waste volumes were so small at the time that management and disposal were not a major concern then. This has changed, with current world estimates of used fuel waste being 370,000 tonnes [1-3,6].

However, recycling of all of the used fuel is possible in FNRs, and has been proven safe. One FNR, the sodium-cooled metal-fuelled EBR-II in the U.S.A. recycled about 34,000 used fuel pins, equivalent to five complete reactor refuelings during its 30 years of safe and uneventful operation ending in 1994 [22]. With its fuel canister design, the EBR-II regularly achieved 20% fuel utilization. A modification of that design achieved a 25% burn-up in the French Phenix reactor [23].

A reactor modeled after the successful U.S. EBR-II is now commercially available, the GE-Hitachi PRISM reactor [24,25]. It was recently offered to the UK to dispose of that country's



Figure 2. Consumption of Uranium and Other Actinides in a Fast-Neutron Reactor Refueled with Used CANDU Fuel.

Two refueling cycles are shown, with fission products extracted at the end of each cycle and replaced with the same weight percent used fuel. At the end of each cycles all actinides are at the same level as at the end of the previous cycle, indicating that any actinides added in the topup, whether uranium or transuranics (TRUs), are completely consumed. Please note the change of scale between panels A and B. excess plutonium as fuel. Our calculations [26-28]. based on the PRISM design, indicated that a 35% burn-up is theoretically possible before fuel replenishment is required (Figure 2).

Fuel cycling facilities (FCFs) that would normally be associated with an FNR to be able to consume all actinides completely have been operating at the Argonne National Laboratories in the USA since 1996 to process the core fuel of the now-decommissioned EBR-II [9, p. 181].

Thus all the necessary technologies are in place, requiring virtually no further research and development to begin to utilize and "detoxify" this massive but massively misunderstood source of non-carbon energy, used nuclear fuel waste. But is a fast-neutron reactor safe?

#### 6. Safety: the EBR-II experience

The safety characteristics of a sodium-cooled metal-fueled FNR, the US EBR-II, are described in detail by Till and Chang [9, Ch.7] and by Koch [29, p.24/25]. Only three characteristics will be mentioned here, most pertinent to the stressors of the kind that befell the reactors at Fukushima, Three-Mile-Island, and Chernobyl.

Two crucial experiments were performed in 1986 to test the passive safety characteristics of EBR-II reactor under full power that likely would not be done with thermal reactors: 1) deliberately cutting off cooling to the core of the reactor after having inactivated the control rods, then letting the event run its course without automated or human intervention (Fig. 3); and 2) similarly shutting off cooling of the heat exchanger inside the reactor tank. In both cases the reactor shut down and reached a stable temperature in 300 to 500 seconds [9, p.148/150].



#### Figure 3. Core Temperature Response of EBR-II on Loss of Cooling Test

Cooling to the core of the EBR-II FNR under full power was shut off at time = 0 seconds, after control rods had been deliberately inactivated. After an initial rise the temperature returned to normal and below within 400 seconds due to a very strong negative temperature coefficient (from [30], with permission).

The third characteristic was the built-in passive cooling by two convective liquid metal heat exchangers in the reactor tank, called shut-down coolers, which carried fission product heat to two convective atmospheric heat exchangers outside the reactor building at every normal shut-down of the reactor [29, p. 24].

While many redundant safety systems in current reactors work very well indeed, these characteristics as exemplified in the EBR-II would have provided a safe passive response to the events leading to the happenings at Chernobyl, Three-Mile-Island and Fukushima Daiichi.

# 7. Fuel cycling with proliferation resistance

To extract energy from every heavy atom in used nuclear fuel, it must be recycled, with the fission products formed separated from the fuel exiting an FNR after 15%, or theoretically 35%, of the fuel has been consumed (Fig. 2). Ideally only fission products (FPs) need be separated. However, the chemical or electrochemical procedures are not ideal for separating only the FP component, since they have traditionally been geared to the separation of fissile isotopes that are still left in the used fuel: U235, Pu239 and Pu241. Such separations were costly, and are not needed for cycling FNR fuel. Indeed such efficient separations are not wanted, since they engender the danger of the misuse of such purification for creating nuclear explosives. Instead, methods are required that do not separate the heavy elements. Two of these are touched on here, chosen because they also have the potential of accelerating the elimination of the long-term radiation hazard effected by the transuranic actinide content of stored nuclear fuel waste.

# 7.1 Pyroprocessing

The method envisioned by Till and Chang [9] to be most appropriate for cycling FNR fuel is pyroprocessing. In brief, this is a non-aqueous electrolytic process carried out in molten salts of sodium, potassium and lithium at around 500°C.

Separation occurs in two steps. In the first step most of the uranium in the used fuel is plated out on an iron electrode until the concentration of uranium in the molten salt is reduced to about one quarter of the concentration of the transuranic actinides (TRUs) that include plutonium, americium, curium, etc. This uranium is virtually devoid of U235, and so definitely not of use for nefarious purposes.

The second separation step occurs on an electrode that is a small vat of molten cadmium submerged in the molten salt. Here the remainder of the uranium plates out, along with all of the TRUs. The crucial characteristic of this electrode is its lowering of the free energy difference between uranium and the TRUs so that both types of actinides can plate out. However, since a difference in free energy still exists, the relative uranium concentration in the molten salt has to be lowered to between one-third and one-quarter of the TRU concentration in the first step so that the TRUs will plate out as well. The fission products (FPs) are the only constituents that stay in the molten salt, from which they are extracted for time-limited storage.

Pure plutonium, the only potentially sensitive material, is never obtained, plutonium remaining mixed with the other TRUs and with uranium.

## 7.2 Uranium nitrate crystallization

While used CANDU fuel could be used directly to replenish the heavy atoms fissioned in an FNR, separating out part of the relatively benign uranium leaves behind a higher concentration of the TRUs to replenish the FNR fuel. Pyroprocessing could be used here as well, but the low concentrations of TRUs in used CANDU fuel suggest an alternative, low-temperature method: uranium crystallization.

In this process pure uranium is extracted in the form of uranyl nitrate crystals from nitric-aciddissolved used CANDU fuel. Extraction can be at levels from 75% to 99% at temperatures ranging from 20°C to -30°C respectively [31,32]. At 0°C the level of uranium extraction is 90%.

The remaining 10% dissolved material is a mixture of uranium, all of the TRUs including plutonium, and all of the FPs in the starting used fuel. This mixture is converted to metal form to serve as FNR fuel replenishment, while the pure uranium nitrate crystals are changed to uranium oxide form for easy storage. The FPs in the metal fuel form enter the FNR fuel cycle and are extracted by normal pyroprocessing of used FNR fuel at the end of the ensuing fuel cycle.

Again, there is no separation of purified plutonium.

## 8. Accelerated use of hazardous TRUs – reduction from 4300 years

If used FNR fuel had been consumed to 15% before cyclic refueling, a level achievable in the commercial PRISM design [24], the corresponding 15 wt% FPs extracted by pyroprocessing during FNR fuel cycling could simply be replaced by an equivalent weight of used CANDU fuel. This approach would provide enough fuel from currently stored CANDU fuel waste to create 4300 years of non-carbon nuclear electricity using FNRs operating at present levels of nuclear power in Canada. Such a long time-span before the stored CANDU fuel waste is consumed would imply that a long-term safe storage would still be required. However, different fuel cycling strategies can greatly shorten the time needed for safe storage.

## 8.1 Strategy 1 --- Elimination of TRUs in 430 years

The used FNR fuel, after extraction of the 15% FPs by pyroprocessing, can also be replenished with the fraction of used CANDU fuel left after uranium extraction by crystallization as outlined in Section 7.2 above. Assuming that the crystallization occurs at 0°C, then 90% of the uranium would have been extracted, leaving a 10% fraction in which the TRUs, including Pu, as well as the FPs in the used CANDU fuel would be concentrated 10-fold. Since the TRUs are merely heavy atom actinides, they serve equally well to replenish the FNR fuel as uranium does.

This approach of preferentially using the more concentrated TRU fraction would result in consuming all of the TRUs in the stored used CANDU fuel in close to 430 years. The extracted pure uranium would require no special storage facilities beyond what is currently used in the manufacture of natural uranium fuel. After the TRUs are consumed first, this extracted pure uranium fraction would then serve as FNR fuel replenishment until it would be used up 4300 years hence.

## 8.2 Strategy 2 --- Elimination of TRUs in 80 years

The strategy outlined in Section 8.1 can be enhanced by taking advantage of the uranium extraction that occurs as part of pyroprocessing. Pyroprocessing not only separates out the fission products from used FNR fuel, but extracts most of the uranium in the fuel on the iron cathode of the electrolytic cell as a first step in the process. If the FNR normally operates with and maintains a fissile content of 15% Pu and other TRUs, then the second cathode of molten cadmium in the pyroprocess would co-extract about 4% of the uranium with the 15% TRUs.

Since a further 15 wt% of the used FNR fuel would be FPs (the result of a 15% fuel burn-up), the total uranium extracted by the iron cathode would be 66% of the total fuel.

This offers the opportunity of using the concentrated used CANDU fraction to replace not only the 15% FPs in cycling FNR fuel but also the 66% uranium extracted in the pyroprocess, for a total replacement of 81%. This approach would accelerate the elimination of TRUs in used CANDU fuel by a factor of 54, corresponding to a time frame of 80 years at current rates of nuclear power production if supplied by FNRs. Again, the stored pure uranium would serve as fuel past those 80 years until used up in 4300 years.

The above two fuel cycling strategies are practically achievable. If it were to become chemically and economically possible to extract only the TRUs from the used CANDU fuel, then hypothetically all the long-lived radioactive TRUs could be consumed in as few as 16 years with current nuclear power levels. Moreover, should one increase the fleet of FNRs in going towards a non-carbon economy, then the elimination of long-lived TRUs would accelerate in proportion.

# 9. Conclusion

It is possible today to turn the world's stored spent nuclear fuel waste into gargantuan amounts of non-carbon energy using fast-neutron reactors (FNRs) which, with cycling of the used fuel, can use up all of the uranium, plutonium, americium and other transuranic elements (TRUs). The elimination of the TRUs also eliminates their long-term, million-year radiotoxicity. The technology exists, with an appropriate FNR now being commercially available, while proliferation-resistant safe cycling and fission product extraction of the spent fuel has been carried out for over 15 years. Since the fast-neutron approach is so efficient in fuel use, Canada's spent CANDU fuel alone would supply Canada's need in nuclear power at current levels for over 4300 years. A fuel cycling strategy is therefore proposed that results in the preferential use initially of the long-lived transuranic elements as fuel, effecting an achievable elimination of their million-year hazard in as few a nine decades.

The use of FNRs compared to the burning of coal for the creation of energy would avoid the release of over 400 billion tonnes of  $CO_2$  from Canada's stored nuclear fuel waste alone. If the stored nuclear waste of the world is used as fuel in fast-neutron reactors, as well as the accumulated depleted uranium stores, then the amount of carbon dioxide avoided for energy production would be 19 trillion tonnes, equal to 6.4 times the  $CO_2$  content of the atmosphere.

The approach would have a major mitigating impact on the carbon dioxide component of greenhouse gas emissions into our atmosphere.

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# 11. Biography

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