

# **NULET: A simpler, more cost-effective and ultimate-safety reactor using natural uranium and a novel liquid moderator material**

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## **Introduction**

The moderator in graphite-moderated power reactors reaches high temperatures even in designs where the nuclear fuel is cooled in a separate circuit (vis. RBMK, max. graphite temp. 750°C; max. water coolant temp. 350°C). Temperatures as high as the graphite would make a great medium for heating steam for a Rankine cycle, comparable to high-efficiency fossil-fired plants (slightly lower efficiency would be possible with a nitrogen Brayton cycle at the same T-high).

Unfortunately the graphite can't be pumped around a heat exchange (HX) loop. It also has issues with degradation in the neutron flux of reactors, as well as radioactive waste disposal (or the cost of re-cycling and re-manufacturing radioactive moderator blocks).

The moderator in heavy-water (HW) cooled reactors can be pumped around an HX loop, but the boiling point is only 101.4°C at ambient pressure, so costly pressure vessels or pressure tubes are required just to get to ~300°C – a temperature at which the steam Rankine cycle yields at most about 35% conversion efficiency, and the turbo-generator equipment set is large and costly.

Moreover, high-purity HW is also costly, as are the systems required for tritium management and periodic HW upgrading.

No other moderator type has been found to be suitable for large-scale power plant application in combination with low-cost natural (unenriched) uranium to date.

In terms of cooling media, a wider selection of materials have been tried in commercial or experimental reactors. Both gases and liquids of various types have been tried in combination with solid fuel, as have a few types of liquid fuel.

A variety of gases with favorable neutronic properties and reasonable cost are available, but effective heat transfer requires high pressure, similar to water cooling, and the low density and large number of cooling channels makes for a reactor core that leaks a lot of neutrons.

The obvious attraction of coolants having a high boiling point is that high-pressure reactor vessels or cooling channels can be avoided, thus reducing at once cost, safety issues, as well as parasitic neutron absorption in thick-walled structural materials. The problem is that there are few high-temperature liquids with suitable neutronic properties and without some other issues.

In thermal spectrum reactors using NU fuel, the problem is especially acute, as liquid metals such as sodium, potassium, lead, lithium, tin or gallium are strong neutron absorbers (except lead-208, which requires costly isotopic purification). In addition, some of these have safety issues with flammability. The same is true of molten salts, with LiF being the archetypical example requiring isotopic separation of Li<sub>7</sub> (in addition to being a strong tritium producer, along with BeF<sub>2</sub> in the typical eutectic mix).

Organic coolants, in particular Terphenyl, have been tried as well. These were used successfully as heat transfer media, but are flammable if the molecules have hydrogen – which is also given off as hydrogen gas due to radiolysis, and must be managed to reduce risk of explosion.

The radiolysis of Terphenyls also produces small amounts of heavy organic sludge over time (polymerisation), although this has been found to be manageable with small on-line chemical treatment system (a distinct advantage compared to graphite in solid moderator blocks).

Fluorocarbon organics (including potentially fluorinated terphenyls, eliminate the hydrogen explosion risk, but create a new problem of extremely corrosive gas production (unlike molten salts, which are ionic compounds not prone to releasing fluorine atoms by radiolysis).

Ideally, one would like a liquid substitute for graphite that doesn't have either hydrogen or fluorine, to be used as both moderator and coolant. Liquid fuel is also preferable, as it can be used to facilitate both rapid core de-fuelling (for ultimate safety decay heat rejection) and some degree of on-line fission product processing, ranging from xenon and other gas removal, to high-performance full-scope processing (either on-line or in batch mode or a combination of the two).

Historically, some of the early nuclear "pile" experiments in Germany attempted using carbon dioxide (CO<sub>2</sub>) in condensed form. Condensed CO<sub>2</sub> is a good moderator, non-flammable, but only used in the "dry ice" form in laboratory experiments, because of the high pressure required for liquid CO<sub>2</sub>, even at ambient temperature – never mind several hundred degrees.

Like the hydrocarbon and fluorocarbon molecules (fluoro-)methane, (fluoro-)ethane, (fluoro-)propane, (fluoro-)butane, etc. carbon also forms "oxo-carbon" analogues, of which the monoxide (CO) and dioxide (CO<sub>2</sub>) are just the commonly known examples.

As in the case of the hydrocarbon and fluorocarbon molecules, the heavier oxo-carbon analogues tend to have higher boiling points, requiring less pressure to liquefy them at any given temperature (below supercritical).

Unfortunately, all of the more common oxo-carbons have boiling points far below 300°C at ambient pressure, and many of the heavier ones are also chemically unstable (decomposing soon after forming). With at least one known exception: Ethylenetetra-carboxylic dianhydride (ET for short) is a tightly bound, unsaturated organic ring compound with the composition C<sub>6</sub>O<sub>6</sub> – somewhat similar to Terphenyl, albeit only in the sense that both are covalently bonded organic ring compounds. Several ET synonyms are possible, using chemistry nomenclature, which complicates document searches (see comments in annex).

Interestingly, the elementary molecular stability issue is similar in hydrocarbon and oxo-carbon compounds, when comparing linear and aromatic (ring) covalent bond structures. In particular, linear chains of carbon atoms containing a series of unsaturated (double or triple) bonds tend to be less stable, be they hydrocarbon or oxo-carbon type, whereas aromatic molecules having the same or greater number of carbon atoms tend to be more stable.

For example, while triple-bonded acetylene (C<sub>2</sub>H<sub>2</sub>) is very stable, C<sub>4</sub>H<sub>2</sub> (combining two triple bonds with one single C-C bond) is unstable. Similarly for C<sub>6</sub>H<sub>2</sub> (three triple bonds and two single C-C bonds). Likewise for linear oxo-carbons heavier than C<sub>3</sub>O<sub>2</sub>: C<sub>4</sub>O<sub>2</sub>, etc. are all unstable (the C-C bonds are all double in this case, as are the oxygen bonds).

By contrast, aromatic carbon compounds like C<sub>6</sub>H<sub>6</sub> (benzene) and the numerous types of phenyls, are models of chemical stability (some of them are the basis or building blocks for commercial polymer plastics).

When some or all of the hydrogen atoms in phenyls are replaced by halogens, they result in very stable and non-flammable compounds like the infamous PCBs (Poly-chlorinated bi-phenyls) formerly used in

electrical power transformers (Destruction of PCBs is generally only possible with very high-temperature incineration).

Thus it comes as no surprise that the ring compound ET -  $C_6O_6$  - is also quite stable, where the long linear oxo-carbons are not.

Although ET is not widely used in industry at this time, a few basic pieces of information are publicly known: ET is a pale yellow oily liquid, used in certain types of polymer synthesis in the laboratory (proprietary, possibly patented), with a melting point slightly below  $300^\circ C$ .

Unfortunately, the all-important data on density-versus-temperature, required for detailed neutronic analysis, appear not to be publicly available at this time. For similar compounds, *ChemSpider* reports densities of about 2.0g/cc, which seems likely for ET – about twice that of liquid  $CO_2$  ( $-20^\circ C$ ), and about the same as graphite (largely due to the heavier oxygen atoms in ET).

The lack of detailed published information further leads to guesstimating that maximum operating temperature with ET – which should not be more than about  $150^\circ C$  above the melting point – maybe around  $450^\circ C$ .

It's possible that slight changes in the molecular structure may yield a more suitable, higher temperature compound than ET. For example, carbon suboxide ( $C_3O_2$ ) polymerises upon heating under pressure to form “oligomers” with 3 to 6 units – the smaller ones like  $C_9O_6$  being of interest here.

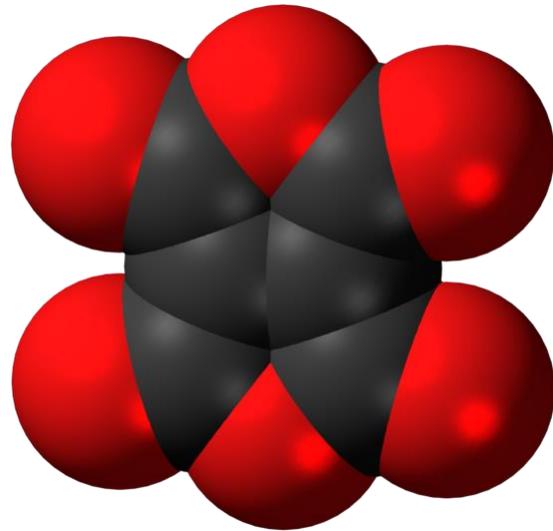
Another candidate, known to be reasonably stable is Mellitic anhydride,  $C_{12}O_9$ , where three anhydride groups are attached to a benzene ring (see illustrations in annex). Unfortunately it's not clear whether it has a suitable melting point or just sublimates. Predictions available in *ChemSpider* list a boiling point ranging from  $441^\circ C$  to  $757^\circ C$  – not inspiring much confidence.

In this age of “designer drugs” it may be that “designer oxo-carbons” are also possible – albeit probably less profitable. In particular, a  $C_7O_6$  molecule, “Allene difurandione”, with a structure similar to  $C_6O_6$  may be feasible, if the central ethylene in ET can be replaced with an allene. These heavier oxo-carbons would tend to have higher melting and boiling points, if they are actually stable compounds.

By basic chemistry reasoning, radiolytic decomposition of ET can only yield simpler oxo-carbons like CO and  $CO_2$ , along with small amounts of oligomers. These by-products do not present an explosion or fire risk, like hydrogen or terphenyl or some of the liquid metals, nor are they corrosive like fluorine compounds.

## NULET reactor concept fundamentals

NULET is in a class of nuclear reactor concepts generically called “bi-modal spectrum”, due to the presence of both thermal and fast neutron spectra, in discrete parts of the reactor core.



Ethylenetetracarboxylic dianhydride molecule ( $C_6O_6$ ) in electron space-filling model (black is carbon, red is oxygen). To a neutron the molecule would “look” very different, with only tiny nuclei “visible”, in roughly equal size for carbon-12 and oxygen-16. (Source: Wikipedia)

Unlike homogeneous (chemical or nanoparticle) mixtures of fuel and moderator or coolant, heterogeneous arrangements can be made sufficiently discrete – in both physical fuel and moderator separation as well as density differences – to yield a bi-modal neutron spectrum (Heterogeneous arrangements with little separation – such as thin fuel channels and moderator blocks, relative to neutron mean free path length – will also behave like homogeneous reactors).

Due to the very high fission x-sections of fuel nuclides at low neutron energies (< 0.1 eV), it is typically the thermal neutron spectrum in a bi-modal spectrum reactor that dominates the reactivity (i.e. controllability), while the fast spectrum acts to boost the fertile fuel conversion to fissile fuel, by way of both increased fission neutron yield per fission at high energy (> 1 MeV), as well as by direct fission of  $U^{238}$ , which does not occur at all below a threshold of about 1.2 MeV.

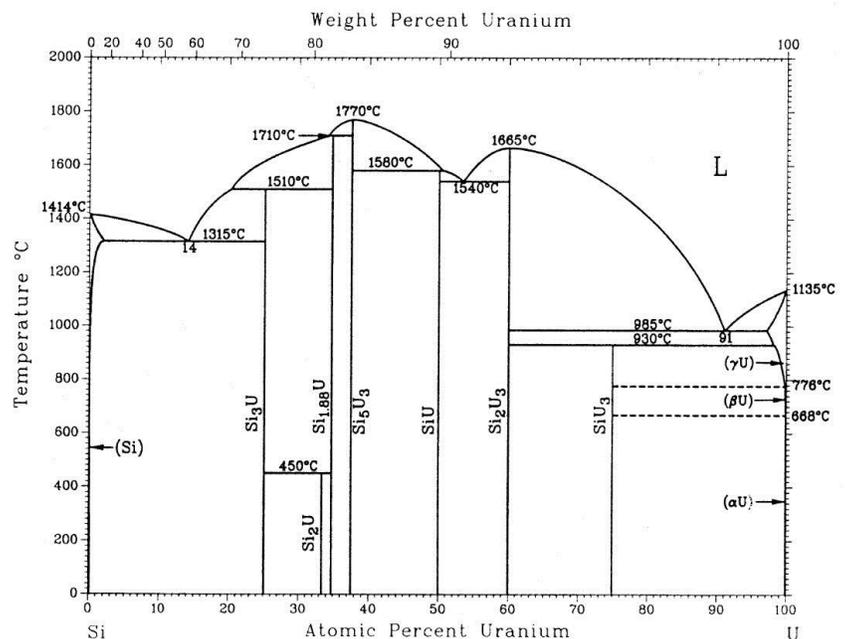
Dominance of the thermal neutron spectrum is important in terms of average prompt neutron lifetime (orders of magnitude longer than fast spectrum prompt neutron lifetime), while  $U^{238}$  fast fission contributes an exceptionally high delayed neutron fraction. Both of these factors add to the controllability and safety of a bi-modal type of nuclear reactor.

Only an in-depth neutronics computer analysis can provide answers about geometry optimisation of the reactor lattice, useful for mechanical design purposes. But some expected NULET concept traits or trends may be surmised from data on related materials and concepts.

As a bulk moderator-coolant material, ET would be an extremely low neutron absorber, slightly lower even than graphite (largely due to manufacturing impurities in the latter, but also due to the higher abundance of oxygen atoms). Its neutron moderating ability would likely be slightly inferior to nuclear-grade graphite, due to the lower content and density of carbon atoms, versus the slightly heavier oxygen atoms (12amu vs. 16amu), but better than fluorocarbons (19amu for fluorine atoms), and substantially inferior to water or hydrocarbons like terphenyl (1 or 2 amu for H and D respectively).

As such, a reactor moderated by ET and fuelled by liquid NU – or NULET – would tend to be large, like graphite moderated types, due to a relatively long neutron mean free path combined with small lethargy increment per elastic collision with moderator nuclei.

On the other hand, the combination of both liquid moderator/coolant and liquid fuel permit a more optimized core lattice, with wider lattice pitch and fatter fuel channels (Solid fuel can only dissipate heat by conduction and can sustain damage to its crystalline structure, leading to fracturing and even local melting, whereas liquids propagate heat more efficiently by convection and don't sustain damage other than radiolytic decomposition, in the case of covalently-bonded molecules).



Phase diagram of the U-Si system (ANL-37763)

The optimisation may be carried to a maximum in NULET with high-density liquid NU metal – actually a eutectic with a small percentage of Silicon (~9a% Si for 985°C m.p. eutectic versus 1,135°C m.p. for pure NU) – in combination with a wide lattice pitch (assumed here at ~350mm pending detailed analysis) and 150mm I.D. fuel channels. For comparison, this fuel channel diameter is about 10mm greater than the diameter of the entire EBR-I reactor core (excluding reflector), which included sodium cooling channels between its pencil-thin U-Zr alloy fuel elements.

With its very high density, NU metal is an excellent shielding material for thermal neutrons. Consequently, the great majority of fission reactions – and resulting heat release – will occur at or near the surface of the fuel channels, with the interior experiencing largely fast neutron fission, including  $U^{238}$  (neutrons only become “thermal” after extensive passage through the moderator, hence “originate” outside the fuel).

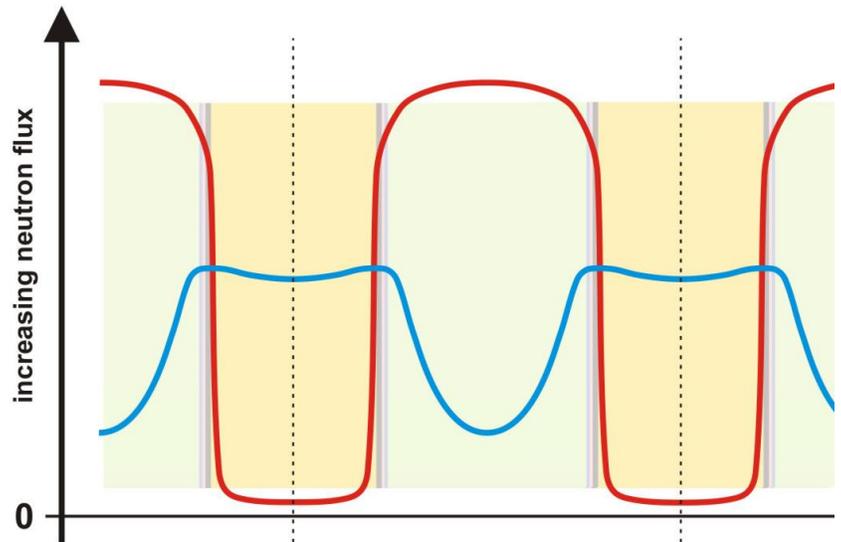
Fission heating at or near the fuel channel cylinder surface is mixed with the bulk fuel interior by convection, and then transferred to the moderator calandria tube by conduction through the annulus separating the fuel channel tubes from the calandria tubes (for safety and corrosion reasons). Some heat is also deposited directly in the bulk moderator by gammas and neutrons.

Unlike other calandria-style reactors like Candu and RBMK, where the coolant circulates through the fuel channels, in NULET the metal fuel would be stagnant, were it not for the internal convective circulation: Each fuel channel is in effect a long thin container with a closed bottom, which includes a small drain line with a freeze plug.

The normally expected poor heat transfer properties of “stagnant” fuel channels – in this respect similar to solid fuel – is to a high degree compensated by the far higher operating temperature of the fuel channel containing molten NU, as well as by an annulus that is filled with liquid metal instead of a gas (Thus, in contrast to Candu, where the gas annulus is optimised to *minimise* heat transfer to the moderator, in NULET the annulus is optimised to *maximise* heat transfer).

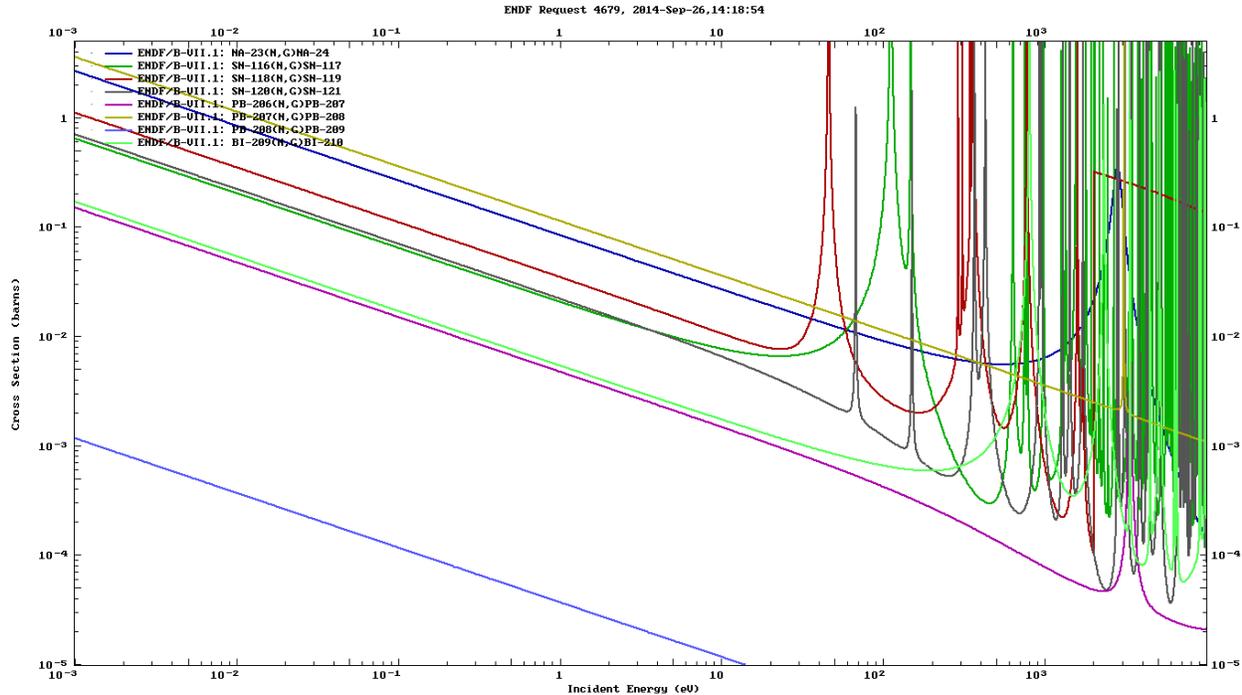
In NULET, the liquid metal in the fuel channel annulus would also have its own drain with freeze plug, such that overheating would cause it to drain, thus instantly cutting heat transfer to the moderator tank from the fuel channels, independent of what happens in the latter (in effect a dual safety drain system, from the ET moderator/coolant heat exposure perspective).

Since the annulus gap is relatively small (somewhat like the “sodium bond” for fuel pellets inside stainless steel clad fuel rods of fast neutron reactors), and the number of “fat” fuel channels relatively few, the choice of material is less restricted neutronically than it would otherwise be, if large quantities of



Notional illustration of neutron flux across two NULET fuel channels and the ET moderator/coolant between them. Thermal flux at 0.05eV is traced in red, fast flux at 2MeV in blue. Short sections of fuel channel (150mm I.D.) and row/column spacing (~350mm) are shown to scale, in pale colors, with dashed centerlines (diagonally, the spacing would be ~500mm). Note the very high *ratio* of fast-to-thermal neutrons inside the fuel channels, and the inverse in the moderator. The associated expected neutron spectrum in the reactor as a whole is sketched in the appendix, showing the “bi-modal” form, with two distinct peaks at thermal and fast ends.

material were required – or as it would be if one were faced with the difficult problem of finding a metal for a uranium eutectic combination: Uranium does not like to alloy with low-melting-point metals, only refractory things like zirconium, iron and molybdenum; it pays to have two separate liquid metals. The exception is Bismuth, but as BNL scientists discovered in the 1950s with their LMFR project for a liquid fuel graphite moderated reactor, Bismuth dissolves only a tiny percentage of Uranium, so they were



Comparison of neutron absorption x-section for Sodium (Na), Tin (Sn), Lead (Pb) and Bismuth (Bi) isotopes, in the thermal spectrum (from BNL NNDC)

forced to use highly enriched uranium (93% U235) – which is of course unacceptable in today’s political environment as regards non-proliferation regulation of civilian nuclear facilities.

An obvious choice for the annulus would be pure tin, since it is fairly inert in air, even at high temperature. Tin would also be suitable as a layer for oxidation protection of the liquid NU metal at its top surface, inside each fuel channel, as well as in fuel drain tanks (Tin will float on uranium without dissolving in it, particularly when both are stagnant in a container). A eutectic mixture of tin might be preferred for its lower melting point, but the combination must be equally resistant to oxidation at high temperature, which seems unlikely.

In the thermal neutron spectrum, tin absorbs neutrons almost as much as zirconium (calandria tubes), so the thickness of both must be kept to a minimum (At 0.01eV the combined value for tin isotopes is about 280mb, whereas of for zirconium isotopes the corresponding value is about 310mb; At 0.058eV – which corresponds to 400°C – the x-sections drop by equal proportions to about 125mb and 140mb respectively). A 1.5mm-thick annulus thermal bond seems feasible, over a 3m length, from a mechanical point of view.

NULET would be a relatively low power-density reactor – due to the fat, non-circulating fuel channels – relying instead on easy scalability due to low-cost materials, principally the ET moderator and the NU fuel which is nearly “dirt-cheap”.

Assuming an average heat transfer rate of 8 kW/100 sq.cm of fuel channel (and including direct ET heating by radiation), each ~3m-long fuel channel could provide about 1.2 MW of heat. Therefore, one

would need some 80 fuel channels for a 100 MWth reactor (producing perhaps 40 MW of electricity through a steam Rankine cycle). This rate of energy production would put NULET in the class of SMRs – Small Modular Reactors, less than 300 MWe.

### **A few engineering design issues**

Every type of nuclear reactor concept has a long list of design issues. It is not the intent here to address all the issues with a NULET-type concept, but merely to point out a few of the more important or interesting ones.

As for structural materials for the fuel channels, calandria tubes, calandria vessel, and dump tanks & plumbing for fuel and annulus metal, this is where a bit of ingenuity (and engineering) is required – and may perhaps be judged as the one instance where a difficult trade-off was made for the gains obtained by abandoning exotic and costly materials (“unobtainium”) requiring isotopic purification, or requirements for management of large amounts of tritium, or periodic disposal of large quantities of radioactive graphite moderator. The trade-off may only be judged fairly if some potential structural materials can be identified, that are compatible with all three liquids proposed for NULET.

For the calandria and its tube set, containing hot ET on the tank side and molten tin on the fuel channel annulus side, zircalloy is the obvious choice, as in Candu reactors. In Candu reactors the 6m-long calandria tube I.D. is 131mm with a 1.55mm wall. For NULET with ~3m-long vertical calandria tubes, 163mm I.D., a 1.5mm wall would be more than adequate.

The calandria vessel of Candu reactors is made of stainless steel, but the operating temperature is only about 80°C, so the interface seal with the zircalloy calandria tubes is perhaps less of an issue than in NULET, where the temperature is on the order of 400°C – hence making an all-zircalloy calandria probably preferable, in spite of the added cost.

Zircalloy is a highly corrosion resistant metal, to all but halide compounds like fluoride salts. Compatibility with hot tin must be verified, and if unsatisfactory a corrosion inhibiting coating may be applied. Silicon carbide (SiC) would be a neutronicly superior alternative to zircalloy by far, and perhaps even cheaper. But there remains the issue of reliable and durable leak-free bonding or interfacing between the calandria tubes and the top and bottom vessel heads (the tubes will experience a small amount of both thermal and radiation damage creep over time – perhaps less so with SiC tubes – which must be taken into account in detailed design).

The first choice of material for the fuel channels is carbon-carbon composite (CC), an extremely strong high temperature resistant material that allows use of thin walls for optimum heat transfer from the fuel to the annulus (CC comprises graphite fibers in a carbon matrix). Compatibility with tin is assured, but molten NU may require additional protection, such as surface impregnation with yttrium oxide – the inhibitor used successfully in AVLIS machines for isotopic separation of uranium in the form of metal vapor.

### **Start-up, operation and shutdown**

In contrast to the distinct *disadvantage* of reduced heat transfer potential with stagnant fuel, the start-up, operation and shutdown of NULET offers other advantages, similar to solid fuel reactors.

Obviously, no pumps are needed to circulate the fuel through heat exchangers. This is a non-trivial advantage at extreme fluid temperatures, even if solid-state electromagnetic pumping were possible. Fouling of heat exchangers with fission products is also avoided, as is the risk of freezing in an HX and loss of delayed neutrons outside the reactor (with operating safety implications).

Start-up involves simply loading fuel channels filled with cold, solid NU metal into the reactor core. For ease of loading into the fuel channels, the NU may be in the form of a series of thick discs stacked on top of each other. The freeze plug valves at the bottom of each channel must be closed prior to criticality and heat-up, to avoid the melted fuel draining out.

Similarly for the tin thermal bond in the fuel channel annulus – although loading would be easier in the liquid state in this case.

ET moderator loading in the calandria must of course be in the liquid form, with adequate pre-heating required.

Control rods would be used for reactor start-up and fine adjustment. These would pass through the ET moderator, similar to Candu reactors (more benign environment than the fuel channels). Neutron poison injection into ET moderator could also be used as a secondary emergency shutdown system, like Candu reactors, but moderator dump would be cheaper if acceptable (see below).

As with other fluid fuel reactors, the excess reactivity on start-up is negligible compared to solid fuel reactors, since fuel adjustments are made on a continuous basis, as opposed to annually (or longer), and the issue of xenon poison-out vanishes thanks to constant de-gassing. Consequently the “rod worth” of control rods would be relatively small, as would the associated neutron losses (which may therefore be applied beneficially to boosting conversion ratio).

Besides the material difference in the moderator of NULET versus Candu, the large difference in calandria heat transfer and flow rate (fluid velocity) in the two cases may have significant impact on detailed mechanical design (rigidity) of the reactivity control mechanisms.

Planned shutdown for inspection & maintenance would involve draining the fuel channels and annulus prior to lifting out the empty fuel channels (any pipe connections at the top would also need to be disconnected). That also applies to replacement of defective or distorted (aged) fuel channels – on a schedule of some 5 to 10 years (old fuel channels would be radwaste, but on a far smaller scale than bulk graphite moderator).

As illustrated in the two schematics included at the end, each fuel channel simply sits on a seal ring which separates the tin in the annulus from the NU fuel. If the seal ring were to start leaking, the immediate result would be of no consequence: NU doesn't mix with tin, and an overflow line near the top of the annulus would take care of changes in fluid level. The seal ring would be replaced during maintenance.

During normal operation, vents at the top of each fuel channel would be used to collect and store gaseous and volatile fission products like xenon, krypton, iodine and cesium (small quantities of tritium are produced by tertiary fission, but this would be expected to mostly diffuse out through fuel channel walls and into the reactor building).

If desired, a small heat-traced line could be used to take small amounts of fuel through the top lid of fuel channels, for more intensive fission product processing. This should be easier with metal fuel than any ceramic fuel pellets or even halide salts (it is noted as one of the advantages of metal fuel used in GE's IFR – the Integral Fast Reactor, where “integral” refers to on-site reprocessing of U-Pu-Zr alloy fuel elements).

Even with metal fuel, on-site reprocessing may only be cost-effective on multi-unit sites. If intensive reprocessing is not used to improve neutron economy and boost conversion ratio (iso-breeder), then tiny

amounts of LEU will have to be added periodically to maintain criticality over a decade or more of continuous operation.

These same lines could also be used for continuously homogenizing the fuel mixture between all fuel channels in the reactor, in order to balance-out the fuel burn-up (fissile depletion and/or breeding of Plutonium) – the preferred alternative to periodically shuffling whole fuel channel loads around the core, using the main drain line at the bottom of each fuel channel.

A vent (and safety pressure relief) line at the top of the calandria would be used to collect gaseous radiolytic dissociation products of the ET moderator. These could be recycled into new ET and sent back to the calandria as necessary.

Four large drain lines on the bottom head of the calandria can be used to rapidly dump all ET in the vessel and the cooling circuit into storage tanks, by opening normally closed valves. Although this will shut down the reactor, it is not credited as a third safety shutdown system in Candu reactors, because it is deemed to be too slow against the potentially available excess reactivity. However, in NULET the maximum excess reactivity is close to zero, so moderator dump might be accepted by regulatory authorities as a second emergency shutdown system – in place of a poison injection system, which adds a fair bit to overall cost.

Calandria draining in NULET is also used in case a leak develops in a calandria tube and tin starts leaking into the ET moderator/coolant (since tin has higher density and static pressure head than ET, any leakage would be into the ET rather than vice-versa).

No serious safety impacts would be expected from such a calandria tube leak, since the amount of tin in the annulus is small and no energetic reaction is expected between tin and ET. Tin in the calandria would be rapidly cooled to ~300°C and tend to spread along the bottom of the vessel, collecting in the drain line nozzles upstream of the drain valves. The reduced level of tin in the thermal bond annulus would cause NU in the fuel channel to overheat and drain through the melted plug in the drain line. Similarly for the remaining tin in the annulus.

A leak of NU from a fuel channel into the annulus could be detected by an increase in radioactivity in the tin or, more easily, in the air space above it (appearance of volatile fission products). No serious safety impacts would be expected from such a fuel channel leak, if the affected unit is drained soon after leak detection, to avoid damage to the zircalloy calandria tube: Uranium alloys with zirconium, so long-term contact with the calandria tube wall would lead to gradual wall thinning and eventual failure (Zircalloy has a melting point of 1855°C, some 650°C above the maximum normal fuel temperature and 870°C above the melting point of the fuel in the drain freeze plug valve).

The maximum pressure experienced by the calandria tube wall in case of such an incident is just the hydrostatic pressure head of the uranium at the bottom of the fuel channel, minus the smaller hydrostatic pressure head of the ET moderator on the other side of the wall – the exact value of which would depend on the relative elevation of the steam generators and the top of the calandria overflow and pressure relief line. By contrast, the pressure inside sealed zircalloy tubes of solid fuel rods, such as used in LWRs, gradually builds up to some thousand psi, as volatile fission products accumulate (the zircalloy tube wall is a fraction of a millimeter thick – around 0.38mm).

For long-term or emergency shutdown the fuel is dumped into storage tanks equipped with decay heat management systems relying on passive heat dissipation.

Depending on the irradiation time of the fuel and extent of non-volatile fission product extraction during operation, the amount of decay heat generated may or may not allow the fuel to solidify in storage. This affects detailed design of systems for recovering and reloading the fuel into the reactor (omitted here).

### **Comparison with other innovative concepts**

NULET most closely resembles what might be termed a thermal version of the recently proposed SMSR fast reactor (*Simple Molten Salt Reactor*, presented by Ian R. Scott M.A., Ph.D and John Durham, Moltex Energy LLP).

Both NULET and SMSR use liquid fuel in “stagnant” fuel channels.

Beyond that there is little similarity: The chloride fuel salt in SMSR is intended to operate at similar temperature as the metal in NULET, but the inferior heat transfer capability of salt versus metal requires a slightly greater number of thinner fuel channels to achieve the desired reactor power output. Besides that, the much more costly fuel (12% Pu<sup>239</sup>) drives the SMSR design to a more compact, heat transfer optimised core design – leading to the proposed 2,293 fuel tubes, each 45mm in diameter (1,500 MW thermal power reactor).

On a unit-power basis, this represents about 1.5 times the number of fuel tubes compared to NULET, but the heat transfer area per tube is about 3.5 times larger in NULET – indicative of much less severe heat transfer conditions (SMSR doesn't have calandria tubes or thermal bond annuli around the fuel tubes, impeding heat transfer).

It would be impractical to include a system of freeze plug drain valves and connecting plumbing with such a large number of fuel channels, so the SMSR doesn't have any.

The SMSR presentation suggests that adequate safety can be achieved without the rapid fuel draining option, relying instead on the thermal inertia of a large pool of secondary salt.

Moltex do not discuss publicly potential issues such as the failure of some of the 2,293 fuel tubes and resulting leakage of the fuel salt into the secondary salt – and how the contamination is rectified (an important issue in a reactor using costly fuel salt with isotopically pure chlorine-37)

Also, whereas SMSR avoids fuel salt pumping, like NULET, it does use forced circulation in the secondary (fluoride) salt pool circuit. In contrast, the ET organic oil in NULET's secondary (moderator/coolant) circuit is much more benign for pump and steam generator equipment.

*(Note on terminology: Although the molten fuel is nominally “stagnant” in both NULET and SMSR fuel tubes, the natural convective circulation inside them seems to justify the term “primary circuit” for the fuel and “secondary circuit” for the coolant or moderator in the reactor vessel – particularly in the SMSR case, where both use molten salts: fuel salt is “primary” salt and coolant salt is “secondary” salt; Other interpretations may be used, but should also be consistent).*

In terms of reactivity control and safety, SMSR and NULET are conceptually similar in relying on thermal expansion for negative reactivity feedback. However, the far shorter prompt neutron lifetime in a fast reactor like SMSR makes this issue one of potentially more grave consequences, than in a thermal spectrum reactor, or a bi-modal reactor in which the thermal portion controls reactivity (For comparison, the prompt neutron lifetime in a Candu-type reactor is 0.9 milliseconds; in a PWR it is 0.05 milliseconds; in fast reactors it is on the order of 0.001 milliseconds; effective delayed neutron lifetime is of course far longer, making mechanical reactor control feasible).

Moltex say that “loss of cooling (accident or design) heats fuel to sub-criticality” as a result of decreased density and resulting increase in neutron leakage. This is the same mechanism successfully demonstrated in the EBR-II fast reactor, but in that case the fuel was a metal alloy (zirconium-uranium), with a very fast density change response to increased temperature. Moreover, leakage of neutrons from the dense metal EBR-II fuel to the less-dense sodium coolant means that eventually neutrons leak right out of the reactor – whereas in SMSR the two salts (fuel and coolant) are of a more similar density, so leakage from the fuel may not necessarily also mean a large increase in leakage out of the reactor. It remains to be seen whether SMSR’s combination of chloride salt fuel and fluoride salt coolant would show similarly quick response and negative reactivity insertion as EBR-II.

In NULET the uranium metal fuel will have quick density response to power and temperature changes like EBR-II fuel, but the reactivity consequences of that are not obvious in a bi-modal spectrum reactor and need to be analysed in detail.

A somewhat better, or at least instructive analogue in this case might be the TRIGA research reactor, a thermal spectrum pool-type research reactor that uses uranium-zirconium hydride fuel and, crucially, a combination of light water and the hydrogen in the fuel as moderators. Reduced fuel density due to increased power and temperature reduces neutron moderation in TRIGA’s hydride fuel, which causes an instant drop in reactivity and a corresponding drop in power.

A decrease in NULET’s fuel density will not affect neutron moderation, only leakage out of the fuel channels. Leakage from the top and bottom ends of the fuel channels (i.e. out of the reactor as a whole) will definitely have a negative reactivity effect. In the radial direction though, a momentary increase in *influx* of thermal neutrons from the moderator into the fuel may be more significant than an increase in leakage of fast neutrons out of the fuel. There may be a very brief power spike before a subsequent dominant negative reactivity trend reduces power.

In any event, the desired reactivity response or its magnitude is not required to the same degree in a thermal spectrum dominated reactor as in a fast one, since the associated power spikes are orders of magnitude smaller.

In a slower response to increased power, the large volume of ET moderator in NULET’s calandria will also expand with increased temperature, thus increasing neutron leakage. In the extreme, boiling of ET will further increase neutron leakage, at perhaps 450°C or 500°C (exact b.p. currently unknown; ET may also decompose to CO & CO<sub>2</sub> before boiling). An overflow collection system is required.

SMSR’s secondary salt (coolant) doesn’t start boiling until ~1,150°C (exact b.p. is composition dependent), and the reactivity and power response, like thermal expansion w/o boiling, is unlikely to be quick enough for a fast reactor to rely upon for safety.

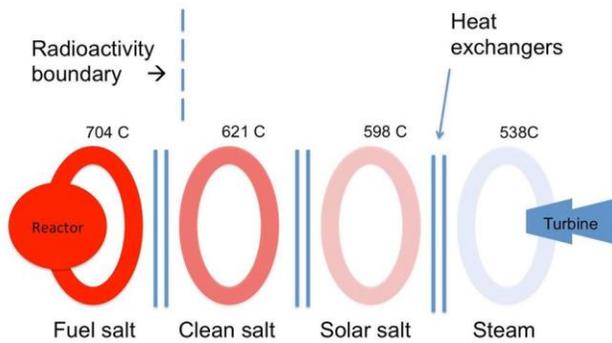
More general comparisons can be made with various molten salt reactor (MSR) concepts such as those proposed recently by Terrestrial Energy, Thorcon and TransAtomic.

Terrestrial Energy’s IMSR concept seeks to avoid tritium issues by using non-lithium fluoride salts in a graphite-moderated reactor. The idea is to reduce hardware and operating costs whilst sacrificing fissile conversion performance and paying for LEU fuel with higher enrichment. The graphite waste issue remains (moderator limits core life to 7 years), as do other things like the requirement for pumps, HXs and valves resistant to fluoride salt corrosion at high temperature. Terrestrial Energy’s publicity graphics contrast the small size of the IMSR relative to mPower’s and NuScale’s LWR SMRs, but fail to note that the latter two’s pressure vessels include the steam generators, whereas the IMSR does not (only primary and shutdown cooling HX units are included inside the IMSR reactor vessel). Moreover, the IMSR

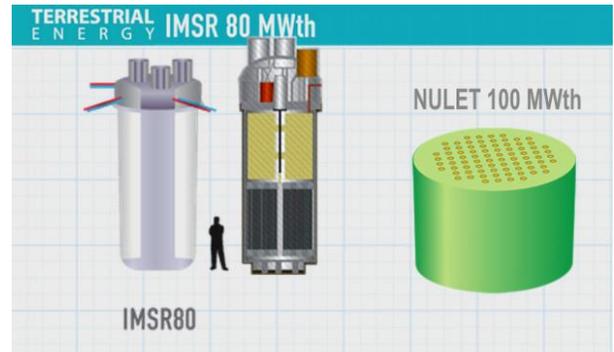
requires at least one extra HX loop to separate the fuel salt from a “clean” salt, which may then be used for non-nuclear industrial applications such as a steam Rankine cycle electricity generation set.

In contrast to the IMSR, other MSR concepts relying on traditional carrier fluoride salts with 99.995% pure Lithium-7 require not only this “unobtainium” salt, but also a series of barriers for containing the large quantities of tritium byproduct. This typically consists of a series of HX loops – up to four, as in ThorCon’s concept. The problem stems from the high temperature of the reactor and secondary loops, since tritium easily diffuses through metal walls at high temperature (as opposed to low temperature, such as the Candu moderator calandria, where tritium is readily contained for many years).

**ThorCon converts energy via four heat transfer loops.**



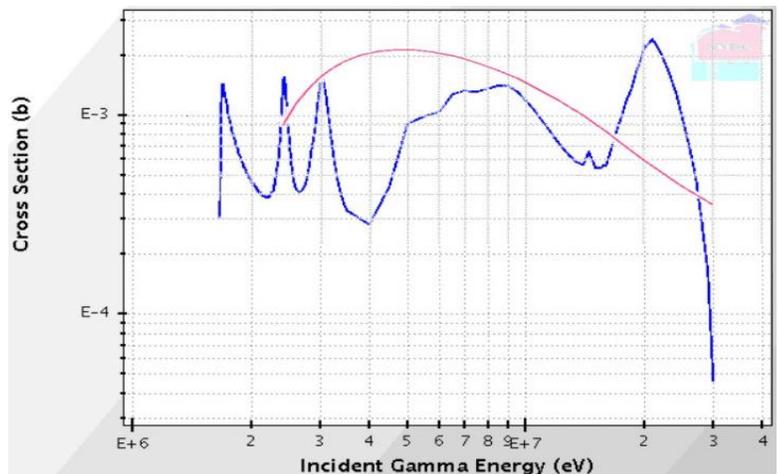
ThorCon’s concept for an MSR relies on the traditional Lithium-7 carrier fluoride salts. A series of four HX loops are proposed for effective tritium containment.



Size comparison of the NULET concept, ~100MWth, with Terrestrial Energy’s 80MWth IMSR. The latter’s vessel includes primary HX loop and shutdown decay heat HX equipment – making up about half the vessel – but not secondary salt HX loops or steam generators.

Lastly, TransAtomics’ TAP concept (formerly WAMSR), while also proposing liquid fuel, replaces graphite moderator with zirconium hydride, also a solid ( $ZrH_{1.6}$ ). Since hydrogen is by far most effective at slowing neutrons, this allows a very compact high-power density reactor (cited as  $86\text{MWth/m}^3$ , in contrast to only  $4\text{MWth/m}^3$  for MSRE). Indeed, TAP’s 1,200MWth (520MWe) places it well beyond the 300MW limit of SMRs. And while the concept succeeds at combining compact size with a large source of power – not unexpected for reactors with circulating rather than stagnant fuel – it comes up short in other respects.

First off, TAP relies on pure Li7-fluoride carrier salt, like most other MSRs, albeit without any  $\text{BeF}_2$ . As such it faces the same “unobtainium” procurement and corrosion issues, and assumes the standard Hastelloy structural material for both reactor vessel and moderator tubes. TAP also loses delayed neutrons in the HX circuit, like other MSRs with circulating fuel, which impacts controllability and safety to some extent. Moreover, by eliminating beryllium from the carrier salt,



Neutron-producing photo-nuclear reaction x-sections for Beryllium and Deuterium:  $\text{Be9}(g,n2a)$  in blue,  $\text{D}(g,np)$  in red (from BNL NNDC).

it also loses delayed neutrons from the  $\text{Be}9(\text{g},\text{n}2\text{a})$  photo-nuclear reaction – the equivalent of which in Candu reactors,  $\text{D}(\text{g},\text{np})$ , is valued for the large effective delayed neutron lifetime, much longer than delayed neutrons from fission products. NULET doesn't get any delayed neutrons from beryllium or deuterium, but it doesn't lose any delayed neutrons from fission products in an external HX either, since the fuel doesn't circulate outside the reactor (depending on the amount of fuel circulating outside the core, MSR's typically lose between one third to one half of their delayed neutrons, while imparting radioactivity in secondary-side components).

In terms of tritium production and containment, TAP will also need multiple serial HX loops, like ThorCon's concept (perhaps only three instead of four if the "solar salt" is avoided). The corrosion issue could get much worse for TAP than other MSR's in case a leak develops in one or more moderator tubes during operation, because reaction between the fluoride fuel salts and ZrH would produce hydrofluoric acid (HF) – far worse than any fluoride salt.

In some ways, TransAtomics' TAP concept is the reverse of Moltex' SMSR: instead of having fuel salt in closed tubes, they put the ZrH moderator in tubes around which the fuel salt circulates. But the issue is similar in that heat transfer out of closed tubes is very limited, hence requiring a great number of thin tubes, to avoid excessive ZrH temperatures. Thousands of tubes in fact, in both TAP and SMSR. If nothing else, one must consider the impact on maintaining all those tubes leak-free over many years of operation, as well as providing the means for detecting leaks and managing retrieval/replacement of failed tubes.

In terms of core lattice design, this inhibits optimisation of the reactor lattice, where fatter ZrH tubes would have been preferable, were it not for the inefficient heat transfer and limited temperature tolerance of ZrH. Secondly, the consequences are much worse for TAP than for SMSR, since a large amount of Hastelloy in a thermal spectrum (TAP) is a far worse drain on neutrons than in a fast spectrum (SMSR). Thirdly, as already noted earlier, zirconium itself is a fairly strong neutron absorber – with the large bulk of ZrH moderator in the TAP reactor adding significantly to the Hastelloy problem (Hydrogen is also a strong neutron absorber, but Zr91 is about 3.6 times more so, and Zr92 only slightly less; Some of the chromium, nickel and molybdenum isotopes in Hastelloy have absorption x-sections about 10 times larger than Zr91).

Nevertheless, TransAtomic claim that their analysis indicates that TAP could operate with fuel enriched only to 1.8%  $\text{U}^{235}$  (i.e. 2.5 times NU) and a fissile conversion ratio (CR) not far below unity (i.e. close to an "iso-breeder"). That is a remarkable result, if true, and suggests that a more CR-optimised reactor like NULET could easily reach or exceed a CR of 1.0 – albeit in a relatively low power density SMR configuration.

– End –

**ET and related reference links (in progress):**

1. Wikipedia: [http://en.wikipedia.org/wiki/Ethylenetetrahydroxydicarboxylic\\_dianhydride](http://en.wikipedia.org/wiki/Ethylenetetrahydroxydicarboxylic_dianhydride)

2. Product Chemical Properties

[http://www.chemicalbook.com/ProductChemicalPropertiesCB6718246\\_EN.htm](http://www.chemicalbook.com/ProductChemicalPropertiesCB6718246_EN.htm)

Lists melting point as greater than 300°C. The material is listed as a "fine chemical" used for synthesizing other organic molecules or as a reagent for high-performance polymer research.

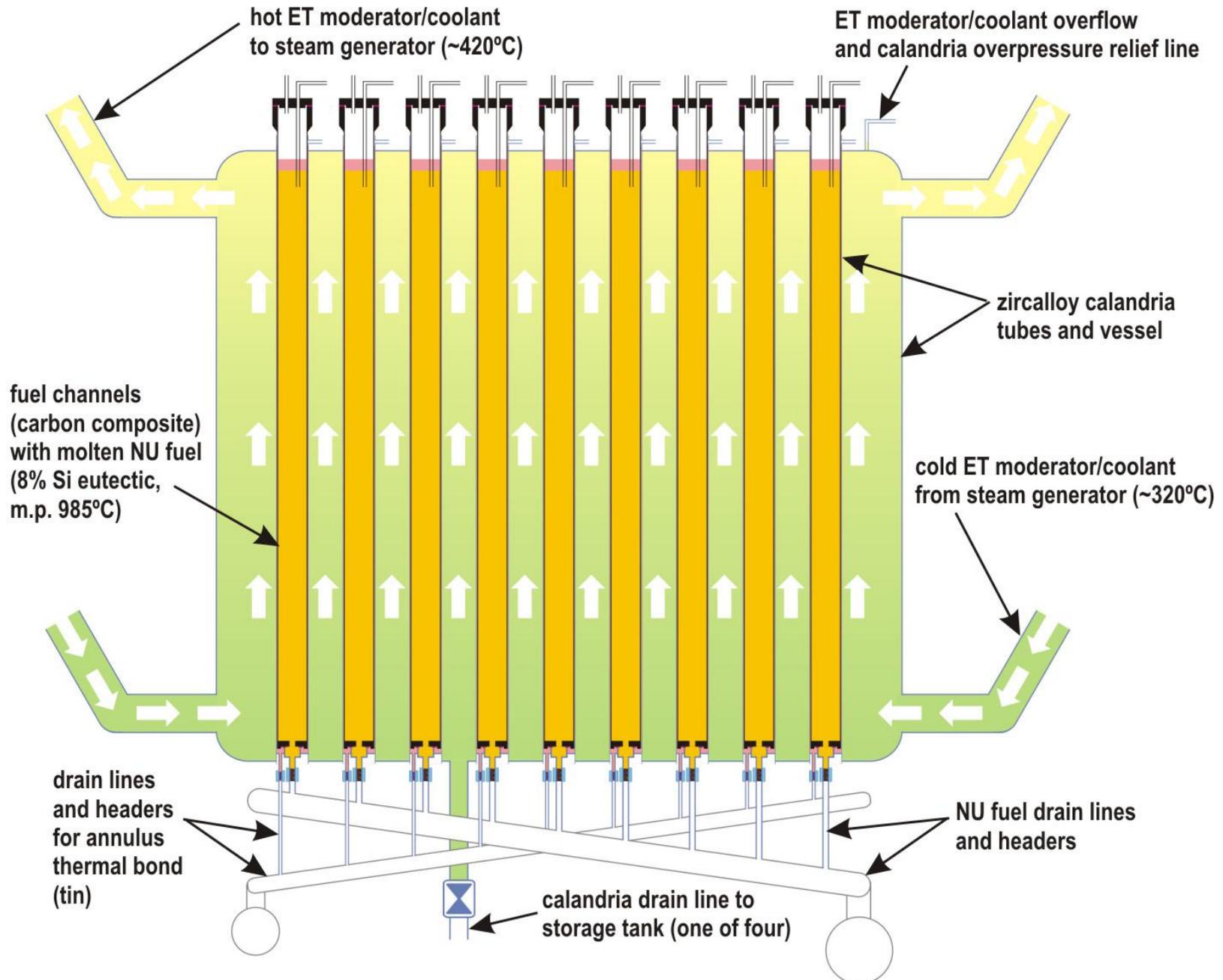
It's called an "Organic Building Block" or "specialty chemical" or a "pharmaceutical intermediate".

3. Chemical Abstracts Service Registry Number. CAS RN for ET is **15016-12-3**

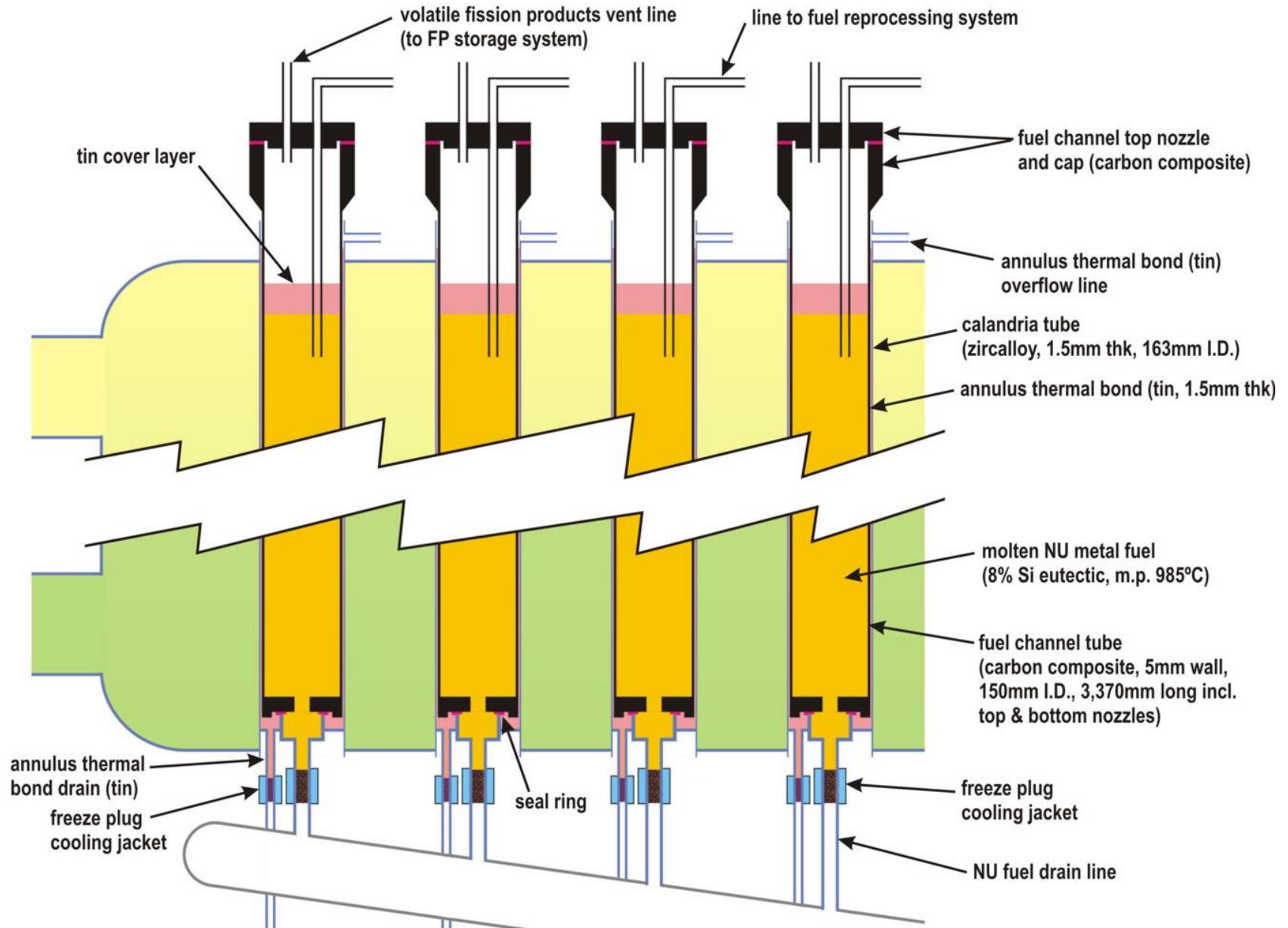
4. "New oxides" and "Polymeric oxides" at <http://en.academic.ru/dic.nsf/enwiki/10699167>

5. *ChemSpider* <http://www.chemspider.com/>

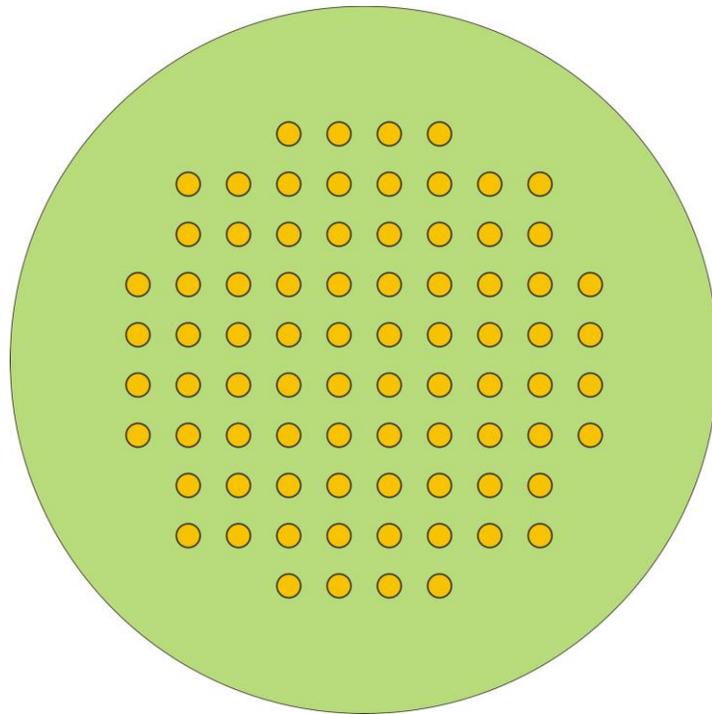
6.



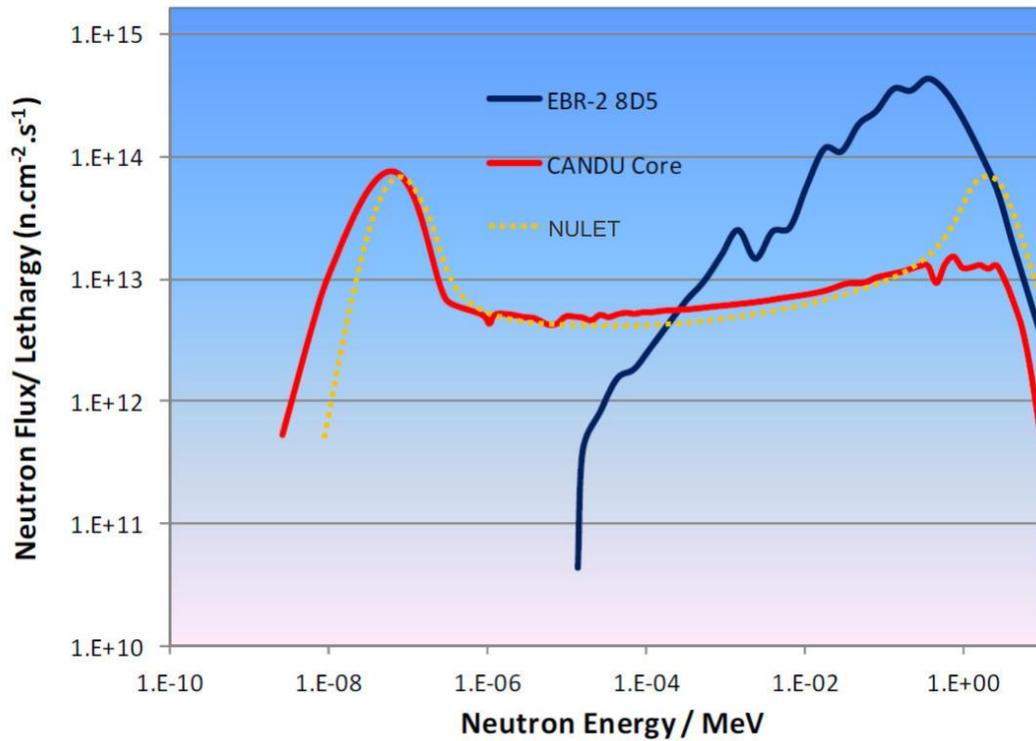
Schematic diagram of NULET reactor concept, with Ethylenetetracarboxylic dianhydride (ET) moderator/coolant and natural uranium (NU) molten metal fuel, in a calandria-style arrangement with large lattice pitch (on the order of 350mm; fuel channel I.D. 150mm). Temperature parameters are merely representative, pending detailed data on ET material (or similar oxo-carbon substitute).



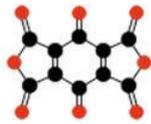
A few details of the schematic diagram of NULET reactor concept, showing the molten tin thermal bond annulus between the calandria tubes and fuel channels, and the drain lines for both annulus and fuel channels. Dimensions are only representative.



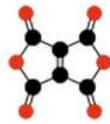
Representative NULET reactor lattice layout: 80 fuel channels (150mm I.D.) at 355mm pitch, in 5m-diameter calandria, for 100 MWth.



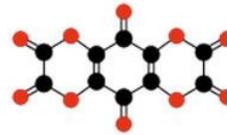
Expected NULET reactor core spectrum compared to Candu and EBR-II. The fast neutron peak at right, although smaller than EBR-II fast reactor, may be slightly shifted to the right, due to absence of sodium inside the liquid NU fuel channels. The thermal neutron peak is likewise expected to be shifted to the right, relative to Candu, due to the hotter moderator/coolant – 370°C average (0.055eV) vs. 80°C (0.03eV).



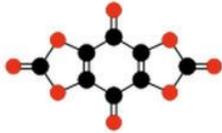
$C_{10}O_8$   
Benzoquinone-tetracarboxylic dianhydride



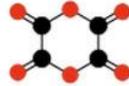
$C_6O_6$   
Ethylene-tetracarboxylic dianhydride



$C_{10}O_{10}$   
Tetrahydroxy-1,4-benzoquinone bisoxalate



$C_8O_8$   
Tetrahydroxy-1,4-benzoquinone biscarbonate



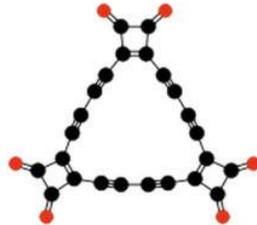
$C_4O_6$   
Dioxane tetraketone



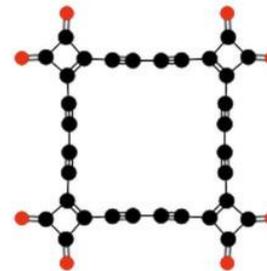
$C_{12}O_{12}$   
Hexahydroxybenzene trisoxalate



$C_9O_9$   
Hexahydroxybenzene triscarbonate



$C_{24}O_6$   
Tris(3,4-dialkynyl-3-cyclobutene-1,2-dione)

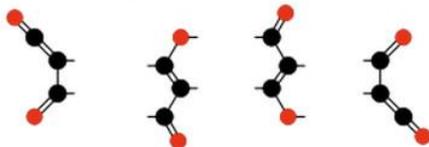


$C_{32}O_8$   
Tetrakis(3,4-dialkynyl-3-cyclobutene-1,2-dione)

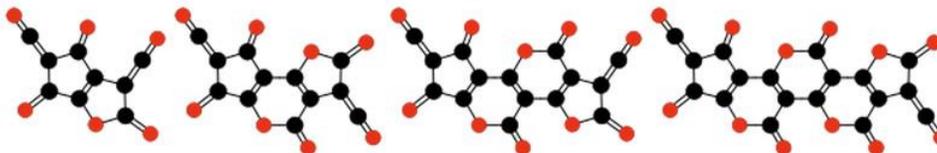


$C_{12}O_6$

<http://en.academic.ru/dic.nsf/enwiki/10699167>

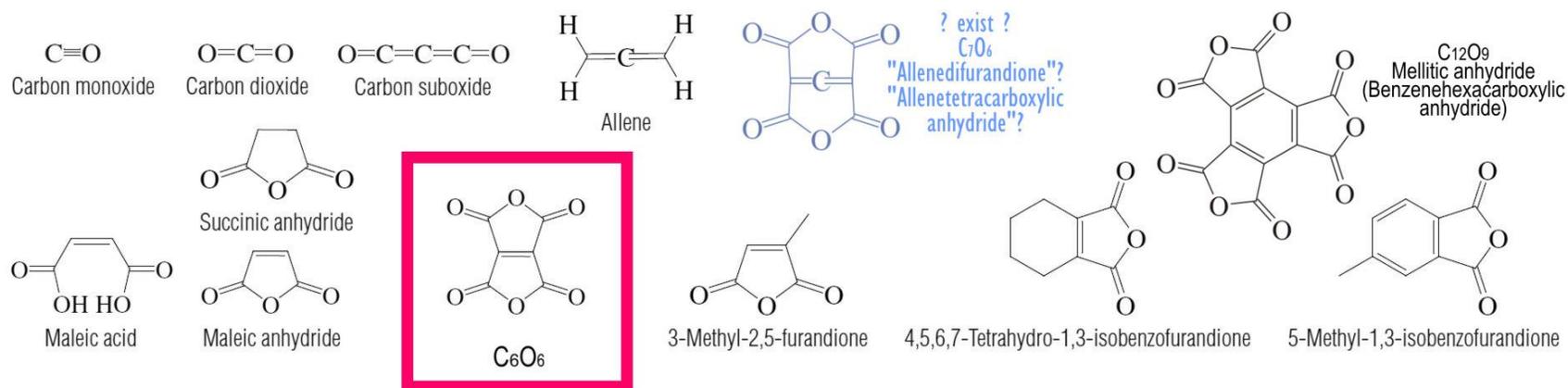


Terminating and repeating units of polymeric  $C_3O_2$ .<sup>[4]</sup>



Oligomers of  $C_3O_2$  with 3 to 6 units.<sup>[4]</sup>

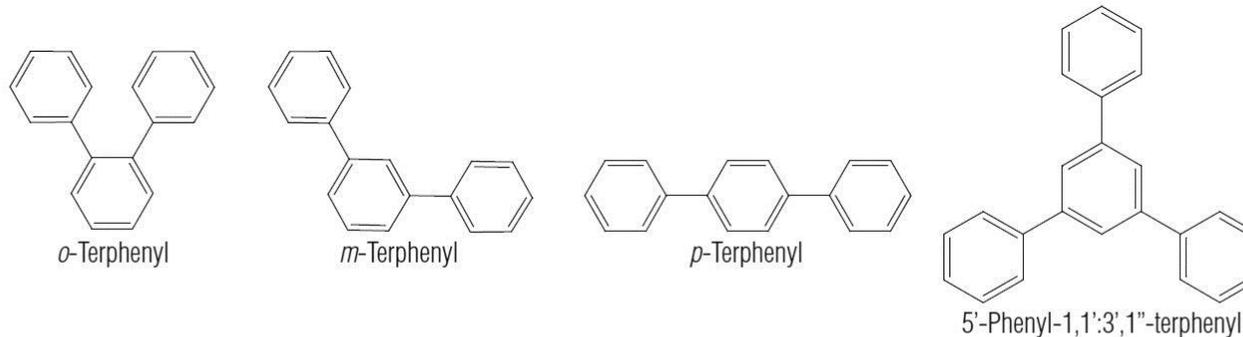
An overview of some of the many types of oxo-carbons. The larger ones, as well as those containing square carbon rings are generally highly unstable.



A few more examples of oxo-carbons and related molecular structures and their nomenclature. Neither  $\text{C}_6\text{O}_6$  nor Ethylenetetracarboxylic dianhydride appear in the *CRC Handbook of Chemistry and Physics* (at least not in the 89th Edition, Internet Version 2009). A few similar compounds are included however (table of properties on next page). The diversity of names applied to similar structures indicates that  $\text{C}_6\text{O}_6$  may take on a variety of synonyms as well. For example, "Tetracarboxylic anhydride" may be used without the "ethylene" and "di" prefixes, even though *the ethylene at the center is what holds together the two sets of anhydride groups*; "Maleic dianhydride" might be used, in line with the acid used in its preparation; "Difurandione" may also be legitimate, going by the last three examples. Shown in pale blue is a hypothetical aromatic oxo-carbon with an Allene backbone. Mellitic anhydride,  $\text{C}_{12}\text{O}_9$ , is synthesized from Mellitic acid,  $\text{C}_{12}\text{H}_6\text{O}_{12}$ , with the removal of three water molecules (similar to converting Maleic acid to Maleic anhydride).

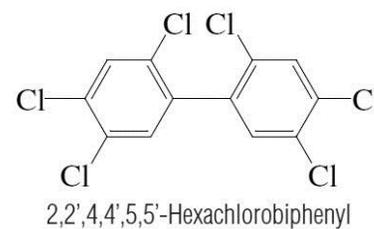
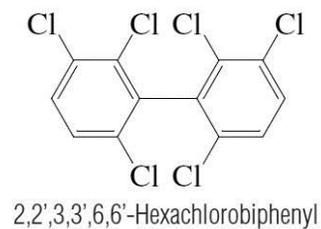
No.	Name	Synonym	Mol. Form.	CAS RN	Mol. Wt.	Physical Form	mp/°C	bp/°C	den/ g cm <sup>-3</sup>	n <sub>D</sub>	Solubility
1741	Carbon dioxide	Carbonic anhydride	CO <sub>2</sub>	124-38-9	44.010	col gas	-56.56 tp	-78.5 sp	0.720 <sup>25</sup> (p>1 atm)		sl H <sub>2</sub> O
1746	Carbon monoxide	Carbon oxide	CO	630-08-0	28.010	col gas	-205.02	-191.5	0.7909 <sup>-19</sup>		sl H <sub>2</sub> O; s bz, HOAc
1754	Carbon suboxide	1,2-Propadiene-1,3-dione	C <sub>3</sub> O <sub>2</sub>	504-64-3	68.031	col gas	-107	6.8	1.114 <sup>0</sup>	1.4538 <sup>0</sup>	s eth, bz, CS <sub>2</sub>
6660	Maleic anhydride		C <sub>4</sub> H <sub>2</sub> O <sub>3</sub>	108-31-6	98.057	nd (chl, eth)	52.56	202	1.314 <sup>60</sup>		s H <sub>2</sub> O; s eth, ace, chl; sl lig
7394	5-Methyl-1,3-isobenzofurandione		C <sub>9</sub> H <sub>6</sub> O <sub>3</sub>	19438-61-0	162.142		93.0	295			
9625	Succinic anhydride		C <sub>4</sub> H <sub>4</sub> O <sub>3</sub>	108-30-5	100.073	nd (al), orth pym (chl)	119	261	1.2 <sup>20</sup>		i H <sub>2</sub> O; s EtOH, chl; sl eth
9864	4,5,6,7-Tetrahydro-1,3-isobenzofurandione	1-Cyclohexene-1,2-dicarboxylic acid, anhydride	C <sub>8</sub> H <sub>8</sub> O <sub>3</sub>	2426-02-0	152.148	pl (EtOH)	74		1.2 <sup>105</sup>		s EtOH, ace, chl; vs eth

Chemical data on a few examples of oxo-carbons and related compounds, from the *CRC Handbook of Chemistry and Physics* (89th Edition, Internet Version 2009).



No.	Name	Synonym	Mol. Form.	CAS RN	Mol. Wt.	Physical Form	mp/ $^{\circ}$ C	bp/ $^{\circ}$ C	den/ g cm $^{-3}$	$n_D$	Solubility
9696	$o$ -Terphenyl		C $_{18}$ H $_{14}$	84-15-1	230.304	mcl pr (MeOH)	56.20	332			i H $_2$ O; s ace, bz, chl, MeOH
9697	$m$ -Terphenyl		C $_{18}$ H $_{14}$	92-06-8	230.304	ye nd (al)	87	363	1.199 $^{20}$		i H $_2$ O; s EtOH, eth, bz, HOAc; sl chl
9698	$p$ -Terphenyl		C $_{18}$ H $_{14}$	92-94-4	230.304		213.9	376			i H $_2$ O; sl EtOH; s eth, bz, CS $_2$
8999	5'-Phenyl-1,1':3',1''-terphenyl		C $_{24}$ H $_{18}$	612-71-5	306.400	orth nd (al or HOAc)	176	462	1.199 $^{30}$		i H $_2$ O; s EtOH, eth, HOAc; vs bz; sl chl

Terphenyl structures and properties, from the *CRC Handbook of Chemistry and Physics* (89th Edition, Internet Version 2009). See preceding text for context. Clearly,  $m$ -Terphenyl is the most attractive of the four shown, for reactor coolant fluid, although it is flammable and produces hydrogen gas by radiolytic decomposition – in contrast to hydrogen-free oxo-carbon compounds.



Examples of PCBs – Polychlorinated biphenyls