C-14 Production in CTR Materials and Blankets

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Abstract. A consecutive study of the source terms, specific and total production of ^{14}C as the major contributor to the external costs of fusion was performed by neutron activation analysis of the low activation structural materials, coolants and breeders suggested for future power fusion reactors. It shows that the specific ^{14}C activity induced in the materials of interest is significantly dependent upon the assumption for nitrogen content. Gas-cooled, water-cooled and lithium self-cooled blanket concepts were considered from the ^{14}C production point of view. A comparison of the ^{14}C activity induced by CTR blankets and by natural and artificial sources as nuclear tests and power fission reactor is given in the report. It is recommended to minimise the nitrogen content in beryllium and in the low activated structural materials below 0.01 wt %. Then due to environmental and waste disposal reasons the ^{14}C generation in CTR will have negligible impact on the cost.

1. Introduction

The socio-economic aspects of fusion power are under comprehensive consideration in Europe, Japan and other countries ([1], [2]). It shows that fusion is found to be a new energy source with acceptable direct costs and very low external costs, while a long-lived carbon-14 ($T_{1/2}$ (^{14}C)=5730 yr) produced in cooling water, breeding blanket and structural materials of a commercial thermonuclear reactor (CTR) is probably the major contributor to the external costs of fusion.

For this reason a consecutive study of the source terms, specific and total production of ^{14}C in low activation structural materials, coolants and breeders suggested for future power fusion reactors was performed by neutron activation analysis.

Among other candidate materials silicon carbide, vanadium alloys, and ferritic steels, water, helium and liquid lithium coolants and solid breeders have been selected as they correspond to maintenance, recycling and waste disposal requirements, and for waste disposal acceptance after 50 and 100 years of cooling. At other times a role of the long-lived ^{14}C in the ingestion dose becomes significant.

A comparison of estimated ^{14}C activity induced by different perspective types of CTR blankets and by natural and artificial sources, as nuclear tests and power fission reactors, is given in the report.

In this respect the study seems to be in a rule of the IAEA programs on determination of possible ^{14}C production, releases and wastes from different facilities ([3]).

2. The Main Sources of ^{14}C in the Environment

The “natural” carbon-14 is produced continuously in upper layers of the atmosphere at a rate of ~1400 TBq/a by action of cosmic ray neutrons on the nitrogen of the atmosphere (as $^{14}N(n, p)^{14}C$). The total equilibrium inventory in the atmosphere is in 100 times higher (~1.4 x 10^5 TBq), while the main ^{14}C activity of about 1.0 x 10^7 TBq is located in the deep oceans [3].

It was shown as early as 1958 in [4] that nuclear explosions were the first substantial manmade source of the radioactive carbon-14. It was stressed in [4] also that because of its relatively long half-life and residence time in the environment, and because of its readily incorporation into biological systems, the artificial carbon-14 could have a significant radiological impact on the living matter.

It is considered now (See [3]) that the total ^{14}C inventory in the atmosphere was more than doubled by atmospheric nuclear weapons tests in the 1950's and 1960's (~2.2 x 10^5 TBq was added), that caused an increase of the natural radiation background level by ~1%.

For several more past decades it was noted that remarkable amounts of carbon-14 are produced in other artificial facilities – nuclear reactors. ^{14}C is generated there by neutron interaction mainly with ^{14}N, ^{17}O and ^{13}C, and may be present in all their elements and systems (as the nuclear fuel, moderators, primary coolant etc.).

Thus the rates of ^{14}C production, that depends essentially on the reactor type and its capacity (See below), is necessary to be controlled.

Now in the beginning of the fusion industry formation, both generation and emission of carbon-14 from the commercial thermonuclear reactors of the nearest future have to be carefully assessed that was performed partially by this consideration.

3. Effective Cross Sections and Expected Reaction Rates

Among others the major ^{14}C producing neutron activation reactions $^{14}N(n, p)^{14}C$, $^{17}O(n, \alpha)^{14}C$ and $^{13}C(n, \gamma)^{14}C$ possess of relatively high cross-sections for thermal neutrons of ~1.8, 0.22 and 0.0013 barn, respectively (Fig.1) that makes for increasing the ^{14}C generation in a softer neutron energy spectrum.

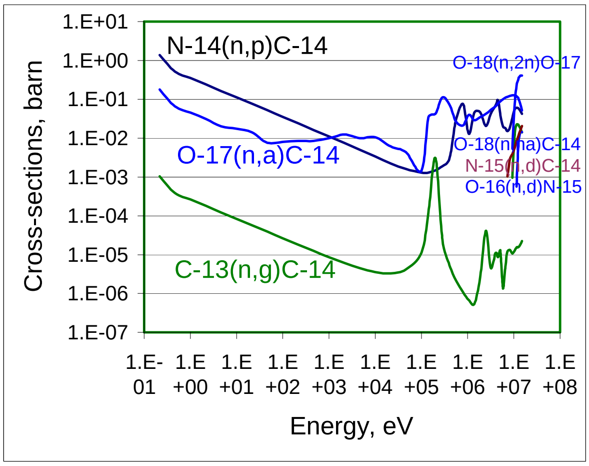

FIG. 1. ^{14}C-producing reaction cross sections

On the other hand, a growth of the cross section energy dependence in the fast neutron energy range, remarkable in Fig.1, tends to increase the ^{14}C production in a harder energy spectrum of fusion reactors in comparison with fission reactor conditions (Fig. 1.).

Besides a sensitivity of ^{14}C production rate caused by a spectrum variation throughout the fission reactor blankets might be expected.

The present assessment of the power system nuclear performances becomes aware of the fact that both the total and fast neutron fluxes in the first wall and blanket regions of a fusion reactor are about a factor of 2 and 2.6 higher, respectively, than that in a typical water cooled fission reactor core (as PWR or WWER) of equal electrical power. (See Fig.2.)

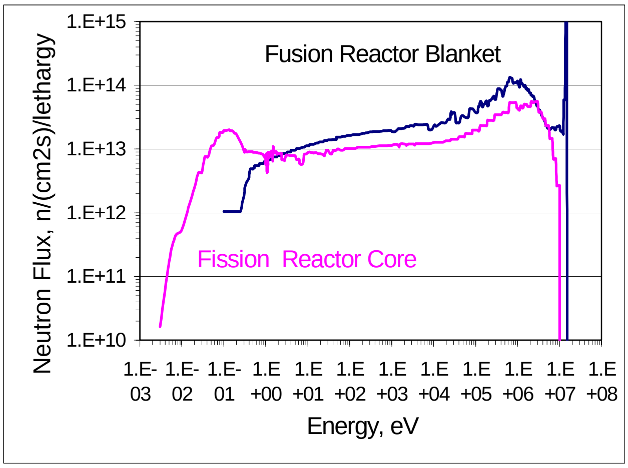

FIG. 2. Neutron spectra in 1-GW(electric) water cooled fission and fusion reactors

This fact is descended from a lower specific energy released per one neutron in fusion reactions than at nuclear fission.

The average total neutron fluxes, effective reaction cross sections and specific production rates calculated per one source nucleus of the most important contributors to the ^{14}C production are compared in Table 1 for a fission reactor core and for a water cooled blanket.

TABLE I: AVERAGE NEUTRON FLUXES ($F_{n\text{-}tot}$), EFFECTIVE CROSS SECTIONS (σ_{eff}) AND ^{14}C PRODUCTION RATES (RR) IN 1-GW(e) FISSION AND FUSION REACTORS

Dominant reactions		**PWR**	**CTR**	**units**
	$F_{n\text{-}tot}$	3.3×10^{14}	6.5×10^{14}	$cm^{-2}s^{-1}$
^{14}N, 99.6% (n, p) ^{14}C	σ_{eff} (^{14}N) RR (N_{nat})	~130 4.3×10^{-11}	~60 $3.9\ 10^{-11}$	mbarns $N_{nat}^{-1}s^{-1}$
^{17}O, 0.038% (n, α) ^{14}C	σ_{eff} (^{17}O) RR (O_{nat})	~40 5.0×10^{-15}	~40 9.6×10^{-15}	mbarns $O_{nat}^{-1}s^{-1}$
^{13}C, 1.1% (n, γ) ^{14}C	σ_{eff} (^{13}C) RR (C_{nat})	~0.18 6.5×10^{-16}	~0.13 9.3×10^{-16}	mbarns $C_{nat}^{-1}s^{-1}$

The maximum specific ^{14}N (n, p) ^{14}C-reaction rate of 4 x 10^{-11} atoms of carbon-14 per second per one initial nitrogen nucleus was estimated in the conditions of a fusion reactor as well as in the conditions of a fission reactor of the same electric power (Table 1). It exceeds the values for other two main reactions with the ^{17}O and ^{13}C by 3 and 4 orders of magnitude, respectively. Note, that the letter reactions are even more intensive than in a fission reactor neutron spectrum.

Reactions on oxygen should be essential in case of using the water coolant and ceramic breeders. Strongly speaking other oxygen isotopes ^{16}O and ^{18}O also contribute about 12 % and 17%, respectively, to the total ^{14}C activity from oxygen by giving the threshold reactions with neutrons, shown above in Fig.1.

Other reactions, including a negligible amount of carbon-14, as a ternary fission product in nuclear fuel, are unimportant in nuclear reactors and do not occur at all in fusion reactors.

Thus, as a primary impurity in fusion reactor materials, nitrogen seems to be the most important contributor to the ^{14}C production.

4. ^{14}C Production Rates in Low Activated Materials

The detailed chemical compositions of the low activated materials, including based alloying elements and anticipated specified impurities, and provided by material manufacturers, were considered with respect to the ^{14}C generation under irradiation in a first wall neutron spectrum.

The substrate nitrogen, oxygen and carbon, initially presented in all structural materials, breeders and beryllium multiplier in a form of unavoidable impurities or constituent elements and identified as the main source terms of ^{14}C, were the most carefully analysed.

The activation analysis was performed with the EASY inventory code system [5]. It shows clearly (Table 2) that a nitrogen content in CFC composites, V-alloys and in the beryllium predominates the ^{14}C generation so that 1 wt. ppm N in those materials irradiated to the first wall neutron fluence of 1 MWa/m^2 results in ^{14}C activity appearance of ~1.1 x 10^5 Bq/kg.

TABLE 2: CALCULATED ^{14}C PRODUCTION RATES IN THE LOW ACTIVATED MATERIALS OF CTRs, PER 1 MWa/m^2

Material	g/cm^3	O, wt.%	C, wt.%	N, wt.%	MBq / kg
C (pure)	2.1		100		3.2
CFC Dunlop	2.1		~100	0.001	4.3
Be	1.85	~1	~0.1	0.028	32
H_2O	0.82	89		0.0001	34
Li industr.	0.53	0.005	0.03	0.03	~30
Li purified	0.48	0.001	0.001	0.001	1.9
Li_2TiO_3	2.9	~47			18
Li_2ZrO_3	4.1	~31			~12
F82H	7.9	0.005	~0.1	0.002	2.1
V-5Cr-5Ti	6.0	0.04	0.015	0.015	17
Zircalloy 2	6.5	0.1		0.05	~60
SiC_f (CG)	2.3	5.8	28.2	0.11	130

For all realistic materials of interest the specific ^{14}C activity varies from ~4 to 130 MBq per 1 kg after irradiation in a typical first wall neutron spectrum to the unit first wall neutron fluence of 1 MWa/m^2.

All these activity levels exceed a natural background activity of 250 Bq of ^{14}C per kilogram of stable carbon by 4-5 orders of magnitude and ought be considered as pollution.

5. A Neutron Wall Loading to Electric Power Conversion Ratio

Amounts of ^{14}C produced by different types of reactors vary considerably, depending on relative masses of structure materials, neutron multiplier, breeder and coolants, and on the concentrations of nitrogen in these systems. Thus a detailed design geometry and neutron space-energy distributions are required in general to accurate prediction of the ^{14}C generation for each particular fusion power system.

Nevertheless a simplified approach may be implemented within the scope of model-building of the future fusion power industry, taking into account its economic and environmental aspects and concerns.

In particular, a transition from traditional neutron wall loading dependent values to electricity production may be performed because of a linear dependence of a power blanket nuclear performance on the DT-neutron first wall loading.

Based on our own and published system studies of a DEMO and power fusion reactors (e.g. [6], [7], [8]) an almost dimensionless conversion ratio of ~2.2 MW(DT-n)/m^2/MW(e) relating the neutron wall loading to the electric power has been drawn. It connects the DT-neutron first wall loading in terms of MW/m^2 with the electric power produced in a power blanket behind.

The factor constitutes a combination of an effective neutron energy multiplication factor of ~1.4-1.5 typical for different blanket designs, and the thermal efficiency of ~0.31-0.41. Also it takes into account ~10 - 15 % losses of thermal energy released in non-power parts of the reactor as a divertor and external heating systems.

The conversion factor obtained is conservative enough and applicable to different kinds of power blankets with a ~10% accuracy. Using this factor the specific ^{14}C production in a power fusion reactor can be calculated based only on a power blanket radial build-up.

6. Carbon-14 in Components of Power Blanket Designs

Three different basic models of power blankets as a fusion reactor core were considered from the ^{14}C production point of view, differing in used cooling mediums and typical blanket concepts suggested in [6], [7] and [8]:

- a pebble bed blanket cooled with water and superheated steam and ferritic steel as a structure, chosen similar to [7] as the most reliable and probable materials for the early generation of commercial thermonuclear reactors;
- a helium cooled blanket with solid breeder and a vanadium alloy as a structure [6]; and
- a liquid Li self-cooled blanket as proposed in [6] and [8].

All the reference blanket designs of ~56, 45 and 50 cm in thick, respectively, include beryllium as a neutron multiplier and have the same power level of 1000 MW(e).

Table 3 summarises material volume fractions for power blanket designs considered at present and specific quantities of ^{14}C generation in coolants, structure, multiplier and breeder materials per unit of electricity produced.

TABLE 3: MATERIAL COMPOSITIONS (in vol.%) AND ^{14}C PRODUCTION RATE DISTRIBUTIONS WITHIN THE EVALUATED BLANKET DESIGN OPTIONS (per 1 GW(e)·a)

Water cooled blanket						
F82H	Li_2ZrO_3	Be	H_2O	Zr	total	total
14	10	42	13	1	80 vol.%	
1.1	0.8	2.7	0.9	1.1	= 6.6 TBq	= 180 Ci
Gas cooled blanket						
V5Ti5Cr	Li_2TiO_3	Be			total	total
11 %	11 %	53 %			75 vol.%	
6.0	2.3	7.7 *)			= 16 TBq	= 430 Ci
Liquid lithium self-cooled blanket						
V5Ti5Cr	Liq. Li	Be			total	total
16 %	7 1%	13 %			100 vol.%	
2.1	0.3	0.8			= 3.2 TBq	= 90 Ci

*) It includes ~1.2 TBq appeared in a 1-cm Be coverage of the first wall proposed in this particular design option.

A variation of neutron flux energy-space distributions throughout the blankets was taken into account at this evaluation. The wall load to electricity conversion ratio mentioned above was used for normalisation.

The total ^{14}C activities of ~7, ~16 and ~3 TBq/a (or ~40, ~100 and ~20 g ^{14}C/a) are expected in those power blankets, respectively, at the end of one continuous operation year.

(An estimated impact of radiation shield components into the total ^{14}C generation of ~5 % was neglected in this consideration.)

The analysis shows that ^{14}C production in the first wall coverage, in the structural elements and in the Be-multiplier dominates the overall ^{14}C production. It is significantly dependent upon the assumption for nitrogen level in the materials, which may range from 20 ppm in ferritic steels to 280 ppm in the beryllium. (Even a very high value of 1100 ppm N is specified in Table 2 for SiC/SiC-composite, that may be found to be a concern for the advanced fusion projects as the American ARIES and Japanese DREAM.)

Higher values evaluated for the gas cooled option reflect an increase of ^{14}C production rate by ~30% in a softer neutron spectrum expected in "homogeneous" breeder/multiplier mixtures of a pebble bed blanket type in comparison with multilayered ("heterogeneous") water cooled blanket structures of the CREST type. An additional impact of the 1-cm Be-first wall protection layer proposed in the analysis is also remarkable for the gas-cooled blanket option (See Table 3).

The dissolved nitrogen content in cooling water was assumed to be very low (usually ~1-5 ppm), and so this source reaction may be ignored in comparison with ^{17}O.

Analysing power performances of water cooled fission and fusion reactors (as an advanced Russian WWER-1500 that is similar to PWR, smaller transport nuclear reactors, ITER and CREST) we have recognised that a water coolant flow rate required in all cases is near to ~5 kg/s per 1 MW(thermal). Thus the value of ~1 TBq/MW(e) indicated here may be probably expected in other cases of a water cooled blanket design.

As to the liquid lithium blanket option, the nitrogen content was proposed to be sustained below the 50 ppm level since nitrogen, preventing tritium recovery by the formation of nitrides on the tritium getter surface, should be permanently removed from the cooling system loops.

Thus an impact of the water coolant of ~0.9 TBq and oxygen free liquid lithium breeder/coolant of ~0.3 TBq into the total ^{14}C activity produced is about 13 %.

A similar impact of oxygen containing solid breeder materials into the overall ^{14}C production is expected due to the ^{17}O neutron reaction. (A nitrogen impurity in the breeders was not assumed in this analysis.)

The most part of ^{14}C is in solid blanket components. But a dynamic of its transfer through the systems and release should be further investigated. It may, in particular, accompany tritium in purge gas at the tritium recovery process.

7. Fusion Produced ^{14}C and Other Sources

A summary of the evaluated ^{14}C production rates in various types of the water cooled, gas cooled and self-cooled fusion reactors is given in Table 4 per unit of electricity generated in comparison with data for nuclear reactors given in Ref. [3].

TABLE 4: SPECIFIC ^{14}C PRODUCTION RATES FOR VARIOUS TYPES OF FISSION AND FUSION REACTORS, TBq / 1GW(e)a

Fission reactors		Fusion power blankets	
Light water	1 - 4	Water cooled	7
Gas cooled	6 - 18	Gas-cooled	16
Heavy water	10 - 12		
Fast breeder	0.5 - 1	Lithium self-cooled	3

It shows that the fusion related values are in the range of the annual ^{14}C production typical for modern power fission reactors such as light water reactors, fast breeders with liquid potassium cooling, for high temperature gas cooled reactors and heavy water reactors.

As distinct from fusion systems the maximum ^{14}C production rate in heavy water nuclear reactors is highly dependent on ^{17}O in big volumes of the D_2O moderator, while in high temperature gas cooled nuclear reactors it is predetermined by a nitrogen level in the graphite similar to the fusion system considered.

It may be shown also that ~140 kg tritium will be burnt in a CTR chamber to produce 1 GW·year of electricity, and 4.5-5x10^{28} DT-neutrons will be released at that. The neutron yield of such a level is 6.5-7 times higher than that from a nuclear reactor core of the equivalent electric power.

In spite of the similar specific values, the amount of ^{14}C containing waste from fusion reactors is a few times more than that from fission reactors because of ~10 times bigger volumes of materials irradiated in blankets and first walls than that in modern light water nuclear reactors.

All evaluated qualities of ^{14}C are almost by 5 orders of magnitude lower than the total additional activity of ^{14}C appeared in atmosphere after the nuclear tests in the former century. In this respect fusion neutrons providing the power production in CTRs are quite "peaceful" than neutrons released at those tests.

Nevertheless, in frame of an extensive fusion power industry that may be imagined at the moment (e.g., 1000 GW(e) x 1000 yr), the ^{14}C will be stockpiling only somewhere in the environment or depositories, and this difference might be got over.

8. Conclusion

The consequent analysis of ^{14}C production source terms in water cooled, gas cooled and self-cooled commercial thermonuclear reactors of the nearest future has been performed, assuming low activation materials and realistic designs recommended by the developers.

It shows that ^{14}C production rates are in the range of the values typical for modern nuclear power reactors. But they are not obviously lower as might be expected for the fusion neutron spectrum conditions. In CTRs with 1000 MW(e) full load capacity ^{14}C will be produced at rates of 3-16 TBq/GW(e)a (90-430 Ci/a) mainly according to the nitrogen impurities in the structural elements, neutron spectrum variation and design features of the reactor types. The water coolant impact into these values is < 1 TBq/GW(e)a while the assessed impact of the oxygen containing ceramic breeders is ~1-2 TBq/GW(e)a.

A ^{14}C activated operational release may occur, in particular, from plasma facing component drops and from the purge gas together with tritium and operation wastes from the cooling water or liquid lithium coolant. Also liquid nitrogen becomes a potential source of a ^{14}C release in case of its using at fusion facilities as a primary coolant for superconductors and the thermal shield for cryopumps. A radiation shield should be foreseen to attenuate the total neutron flux by four orders of magnitude and to restrict in that way the ^{14}C generation in those systems and loops.

But the main part of the ^{14}C build-up will be released at dismantling of CTRs, their component recycling and during decommissioning operations.

More accurate analysis and assessing the expected ^{14}C activity induced both in blankets and radiation shields and ^{14}C releases are still needed to define corresponding material and design requirements. In particular, as nitrogen has waste disposal management concern due to ^{14}C, it has to be included in a list of controlled elements at the low activation material development, along with other impurities as Nb, Mo, Ag, Co, Ni. In this respect it is highly desirable to decrease nitrogen content in these materials to the value of <0.01 wt.% N (<100 ppm N) and even lower if it's practically possible. This is especially imporant for beryllium presuming its multiple reuse. As a result several times lower ^{14}C production rates may be expected than upper values assessed in the report.

Then due to environmental and waste disposal reasons the ^{14}C generation in CTRs will be acceptable and have negligible impact on the cost of electricity produced.

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