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POLARIZATION OF SOLID DEUTERIUM-TRITIUM FUEL FOR NUCLEAR FUSION

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ABSTRACT

The advantages of using polarized DT-fuel are discussed. An outline is given of the standard methods employed to produce polarized solid hydrogen targets for nuclear physics. The applicability of these methods for obtaining polarized DT is analysed. Large polarizations seem to be realisable in spite of the tritium decay heat, which hinders the attainment of low temperatures.

POLARISIERUNG VON FESTEM DEUTERIUM-TRITIUM BRENNSTOFF FÜR DIE KERNFUSION

ZUSAMMENFASSUNG

Es werden die Vorteile der Verwendung polarisierter Kerne bei der DT-Fusion besprochen. Die Standardmethoden der Kernphysik, um polarisierte feste Wasserstofftargets herzustellen, werden beschrieben und deren Anwendbarkeit zur Polarisation eines DT Gemisches analysiert. Hohe Polarisationswerte scheinen realisierbar zu sein trotz der Zerfallswärme von Tritium, welche das Erreichen tiefer Temperaturen erheblich erschwert.

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1. INTRODUCTION

Some years ago discussions have started about the usefulness and the feasibility of employing nuclear polarized fuel for nuclear fusion reactors [1]. For light particles with energies in the keV region the fusion cross section is determined by resonances: particle-unstable states in the compound nucleus. If one resonance dominates, as in the D-T case, a clear increase in fusion cross section can be achieved by the employment of properly aligned polarized particles.

The low-energy D-T cross section is almost completely determined by the $I^\pi = 3/2^+$ resonance at 107 keV, where I and π denote the nuclear spin and parity. At low energies the orbital angular momentum $\ell = 0$. Since the deuteron spin is 1 and the tritium spin is 1/2, the reaction only occurs for particles with parallel spins. For unpolarized particles the statistical weight of this situation is 4, that of the anti-parallel situation is 2. Hence in the ideal case a 50% increase of the cross section can be achieved with polarized particles. A small fraction of the cross section is ascribed to a state 3 MeV above the $\frac{3}{2}^+$ state. Estimates range from less than a percent to a few percent[2,3]. Its small influence is neglected in this paper.

deuteron polarization	triton polarization	cross section gain
100%	100%	50%
90%	95%	43%
70%	95%	33%
50%	95%	24%

Table 1.: Gain in fusion cross section for various deuteron and triton polarizations.

For partially polarized tritons and deuterons the maximal gain of 50% appears to be reduced proportionally to the size of both polarizations. A few examples are shown in Table 1. For deuterons lower polarization values were taken than for tritons, because deuterons are more difficult to polarize. The table shows that incompletely polarized particles still may yield a considerable gain.

A second aspect of polarized fuel is the anisotropy in the angular distribution of the reaction particles generated by the polarization. The redistribution of neutron flux may appear advantageous for reactor design, because of the option to position some sensitive instruments at locations with less neutron flux.

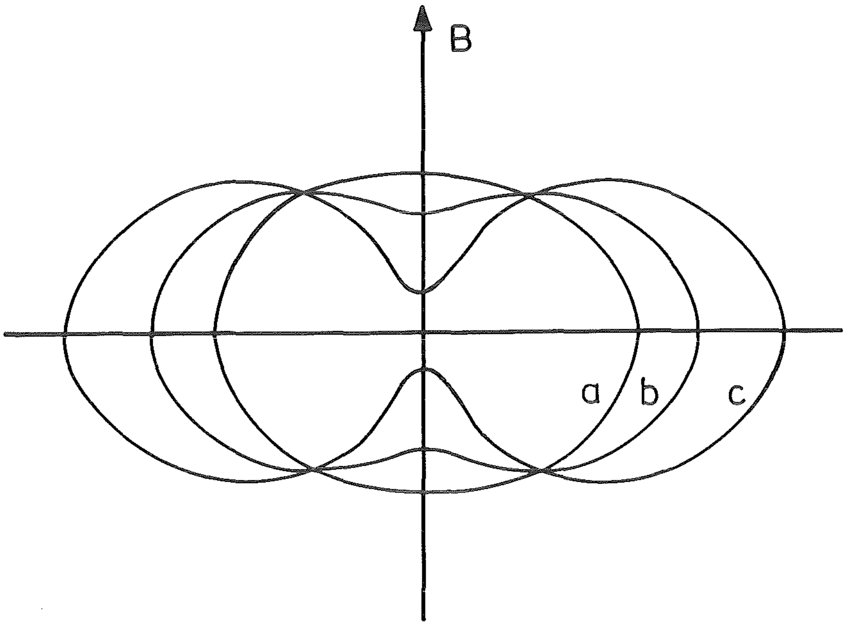

Fig.1: Anisotropies of neutron emission for DT fuel, polarized along the magnetic field B. The curves have been calculated for a triton polarization of 95% and deuteron polarizations of 50% (a), 70% (b) and 90% (c).

It is possible to suppress the emission in the direction of the field (by polarizations parallel to $\vec{B}$) or to enhance the emission in the direction of $\vec{B}$ (by polarized deuterons perpendicular to $\vec{B}$). In the latter case, however, the anisotropy is only small and there is no gain in cross section. Some examples for polarizations parallel to $\vec{B}$ are shown in fig.1., using expressions from ref.1. It appears that the flux ratio $\sigma(0^\circ)/\sigma(90^\circ)$ can become very small.

The employment of polarized fuel would be of no practical use if the depolarization rate in the reactor would be faster than the reaction rate. Up to now theoretical investigations indicate that the reaction rate is faster both for magnetically confined plasmas 1) and for inertially confined plasmas 4).

In magnetic confinement reactors the polarized fuel may be considered to be injected in the form of neutral beams or in the form of solid beads. In recent years solid bead injection has shown to be advantageous, because in this way higher plasma densities and longer confinement times can be achieved5,6).

The inertial confinement scheme employs pellets with DT-fuel inside an enclosure of some other material. The first laser pellets were tiny glass balloons containing gaseous fuel. Newer designs, especially in connection with ion beams, conceive multilayered pellets with an inner layer of solid DT-mixture and a relatively large empty space in the centre.

In this paper we investigate the possibilities to polarize solid DT-fuel both for magnetic confinement and for inertial confinement reactors. Ideas have come up to produce polarized solid fuel starting from polarized atomic beams. Such schemes will not be pursued here. Here we consider cold beads or pellets containing solid fuel, which is to be polarized after solidification with methods that are well established in nuclear and particle physics to produce polarized targets.

In section 2 some general features of polarizing nuclei in solids are discussed. Section 3 describes the methods that are successfully applied in nuclear physics to produce polarized hydrogen targets in the form of solid hydrogen compounds. Some properties of pure solid hydrogen are discussed in section 4. They are of relevance since the nuclear polarization results in pure hydrogen have been limited to date. Nuclear polarization in solid H_2, D_2 and HD is discussed in section 5. The experimental results are extrapolated into prospects for polarizing DT in section 6. The problem of self heating due to tritium β-decay is treated extensively. Section 7 deals with the polarization decay during transfer to the reactor. The results are summarized in section 8.

2. NUCLEAR POLARIZATION IN SOLIDS

Magnetic moments can be oriented in space by putting them in a magnetic field. This also applies to nuclei with a magnetic moment, i.e. nuclei with spin $I \neq 0$. A sample of nuclei is said to be oriented when the populations a_m of the magnetic substates with quantum number m are not all equal. The degree of orientation of an ensemble of nuclei is most generally described by the (2I+1) x (2I+1)-dimensional density matrix. From it orientation parameters of increasing order can be deduced. The first one is called the polarization P. It can be expressed in the populations a_m as follows:

$$P = \frac{1}{I} \sum m a_m . \tag{1}$$

In thermal equilibrium of the nuclei with their surroundings at the temperature T the polarization is given by:

$$P = \frac{2I+1}{2I} \coth \left(\frac{2I+1}{2I} \frac{\mu B}{kT}\right) - \frac{1}{2I} \coth \left(\frac{\mu B}{2IkT}\right), \tag{2}$$

where μ is the magnetic moment of the nuclei and B the magnetic field. In order to obtain sizeable polarizations the thermal energy kT should be lowered until it becomes of the magnitude of the magnetic energy μB. This is shown in fig.2. for the three hydrogen isotopes at a magnetic field of 15 T. This is about the

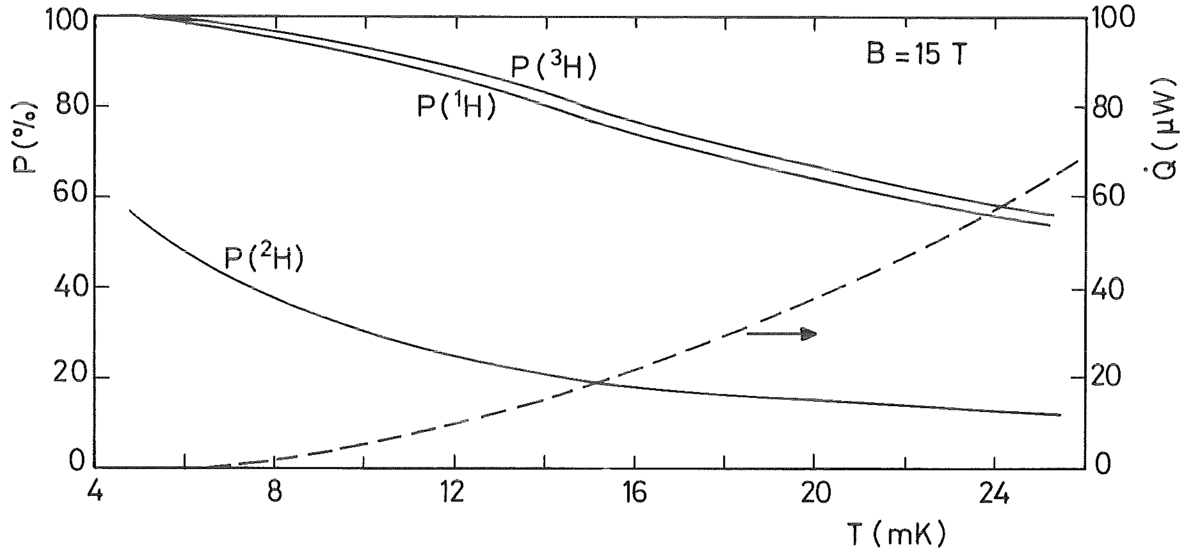

Fig.2: Brute-force polarizations of the hydrogen isotopes in a 15 T magnetic field. The dashed line represents the cooling power of the most powerful dilution refrigerators to date.

limit of what is obtainable nowadays for small to medium-size superconducting solenoids. One sees that below 25 mK the proton and tritium polarizations rise above 50%. At this temperature the deuterium polarization is still very low, however. One has to go below 5 mK in order to find a deuterium polarization above 50%. The spins and magnetic moments of the hydrogen isotopes are listed in Table 2.

Isotope	I	μ (nucl.magn.)
$^1H=H$	1/2	+ 2.793
$^2H=D$	1	+ 0.857
$^3H=T$	1/2	+ 2.979

Table 2.: Spins and magnetic moments of the hydrogen isotopes.

The term brute-force polarization is used when the nuclei are in thermal equilibrium with the lattice and when the magnetic field at the nucleus is due only to an external magnetic field. In many cases the external field is enhanced, weakened or otherwise disturbed at the position of the nucleus by the magnetic properties of the solid. These phenomena do not play a role in the discussions in this paper.

Alternative to the thermal equilibrium method another polarization method has been developed with relaxed requirements with respect to magnetic field and temperature. In this method the sample is doped with a small amount of paramagnetic electrons. Due to their much larger magnetic moment these already become polarized in a magnetic field of a few Tesla at a temperature around 1 K. Subsequently the electron polarization is transferred to the nuclei by microwave irradiation. This method is discussed in more detail in the next section.

An important quantity for polarizing nuclei in solids is the nuclear spin-lattice relaxation time. It is a measure for the strength, with which the nuclear spin is coupled to the lattice. In the case of brute-force polarization one needs a fast spin-lattice relaxation in order to reach the equilibrium polarization as fast as possible. For non-equilibrium methods, on the other hand, one needs a slow spin-lattice relaxation in order to retain the non-equilibrium nuclear polarization. The relaxation time has the tendency to increase towards lower temperatuares, but it varies enormously from subtance to substance. At 0.01 K, e.g., some metals have a spin-lattice relaxation time of the order of a second, whereas pure dielectrics can have relaxation times up to years.

3. POLARIZED PROTONS AND DEUTERONS IN HYDROGEN COMPOUNDS

3.1 Brute-force polarization

It is shown in fig.2. that temperatures down in the mK region are necessary to polarize the hydrogen isotopes. The only device, which reaches such low temperatures in a continuous mode is the ^{3}He - ^{4}He dilution refrigerator. The cooling power of the most powerful dilution refrigerators existing today is also shown in fig.2. It is about 60 µW at 25 mK, 6 µW at 10 mK and drops below 1 µW at 5 mK. Thus only small heat loads can be tolerated at these temperatures. The two sources of heat, that are important for the hydrogen isotopes are the ortho-para conversion heat and the heat produced in the β-decay of tritium. Both will be dealt with in later sections of this paper.

In this section we consider the brute-force polarization of hydrogen in chemical compounds. The clear advantage of a compound is, that in it no ortho-para conversion heat is released. For a successfull nuclear polarization the material has to meet two requirements: the heat conductivity must be high enough and the spin-lattice relaxation must be fast enough. This is the reason why generally dielectrics such as CH_4 can not be considered, but metalhydrides are favoured, because of their metallic properties.

The only compound in which brute-force polarization of protons has been demonstrated up to now is TiH_2. This material has been produced and polarized in our laboratory$^{7)}$, where it served as a polarized proton target for fast polarized neutron scattering. Polarizations up to 70% have been achieved in a magnetic field of 9 T and at a temperature of 10 mK. Brute-force polarization of deuterons has not been demonstrated yet.

3.2 Dynamic polarization

This method started to be developed in the early sixties to produce polarized proton targets for nuclear physics scattering experiments. The basic principle will be outlined here. The first step is to introduce some paramagnetic electrons into the sample. Because of their high magnetic moment μ_B compared to nuclear magnetic moments μ_n such electrons can be polarized quite easily (Fig.3a). A thermal equilibrium polarization of 93% is attained at B = 2.5 T and T = 1.0 K. The next step is to irradiate the sample with microwaves with frequency ν obeying $h\nu = 2\ (\mu_B\ B-\mu_n\ B)$, see Fig.3b. A quantum of this radiation can be absorbed by an electron-nucleus pair, causing a spin exchange between the electron and the nucleus (Fig.3c). The electron flips back quickly (Fig.3d) due to its large magnetic moment and its strong coupling to the lattice. The nucleus depolarizes only slowly due to its weak coupling to the lattice. The nuclei can transfer their polarization due to mutual spin flips. In this way a small number of paramagnetic electrons can pump the nuclear spins of the whole sample.

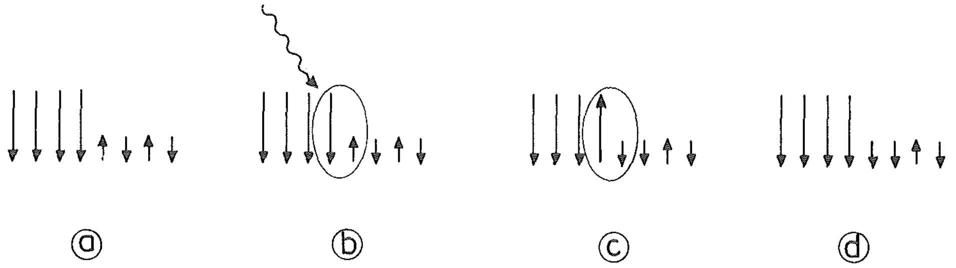

Fig.3: Principle of dynamic polarization: see text for details.

In practice two slightly different mechanisms occur. The first is called "solid effect" and proceeds as described above. The second mechanism is known as "DNP" (dynamic nuclear polarization). Here the electron and nucleus do not flip simultaneously, but they are coupled by an additional energy reservoir. For more details the reader is referred to the literature, e.g. ref.8. Proton polarizations over 90% are achieved at B = 2.5 T and T ≈ 0.4 K in alcoholes and in ammonia[9]. At these conditions the deuteron polarization is limited to 40 - 45%. For pushing it up one has to lower the temperature and/or to increase the magnetic field. Lowering the temperature of the coolant appears to be not very effective, but one can employ larger magnetic fields than the 2.5 T, which are usually employed for polarized proton targets. A deuteron polarization of 49% in ND_3 has been reached in Bonn9) at 3.5 T; polarizations of 64% and 71% in ^{6}LiD have been achieved in Saclay 10) at 4.8 T and 6.5 T, respectively.

A major issue is how to have stable paramagnetic electrons in the sample, because usually they are very reactive and pair off to spin 0 electron pairs. In the early days of polarized proton targets the 4f-electrons of the lanthanides were employed for this purpose, e.g. in lanthanum-magnesium-nitrate8). Later one has found special stable complexes containing a paramagnetic electron, which were employed to polarize protons in alcoholes11). The most recent method, applied e.g. in ammonia12) and lithiumhydride, is to create them by irradiation with electrons, protons or gamma's. This is done at low temperature to prevent the recombination of the paramagnetic electrons. In ammonia the paramagnetic electrons appear to be situated at the radical $\overset{\bullet}{N}H_2$.

4. PROPERTIES OF SOLID HYDROGEN

The properties of solid hydrogen are crucial in studying the feasibility of the production of polarized fuel. The properties that will be discussed here are: the ortho-para transition, the heat conductivity and the nuclear spin-lattice relaxation. Quite recently Silvera13) published a comprehensive review on solid hydrogen. Much of the material presented in this chapter has been taken from this paper.

In the solid state hydrogen appears in the form of H_2-molecules, as in the gas. The wave-functions that describe the rotation of an isolated molecule are almost undistorted by the interactions with neighbours in the solid due to the large intermolecular distance. Thus the solid can be visualized as an assembly of molecules localized at lattice sites, but freely rotating even down to 0 K.

Because the protons in an H_2-molecule are identical fermions the total wavefunction of the molecule has to be anti-symmetric. This brings about a correlation between the rotational states and the wavefunction of the proton spins.

For symmetric rotational states (rotational quantum number J is even) the nuclear spin wavefunction has to be anti-symmetric. This means the total molecular nuclear spin $I_{tot} = 0$ and the protons are anti-parallel. For anti-symmetric rotational states (J is odd) the nuclear spin state is symmetric with $I_{tot} = 1$. The situation is the same for tritium. Deuterium is different because deuterons are bosons with spin 1, therefore the total molecular wavefunction has to be symmetric. The rotational states involving the largest nuclear spin-degeneracy are designated ortho, the others para. The allowed combinations for the hydrogen molecules are listed in Table 3.

molecule	I_{mol}	J	$\sum(2I_{mol}+1)$	designation
$^1H_2$, $^3H_2$	0	even	1	para
	1	odd	3	ortho
$^2H_2$	1	odd	3	para
	0,2	even	6	ortho

Table 3: Allowed combinations of nuclear spin states and rotational molecular states for the mono-isotopic hydrogen molecules, I_{mol} is the total molecular nuclear spin, J the rotational quantum number. The species with the largest spin degeneracy are designated ortho.

In hydrogen the energy spacing between the J = 0 and J = 1 rotational states corresponds to a temperature of 170 K, in deuterium it is half this value. At thermal equilibrium at T = 1 K or below the molecules are almost purely in the J = 0 state, therefore. Such low temperatures are needed for nuclear polarization. Inspection of table 3 shows, however, that it is not possible to have polarized protons in J = 0 H_2-molecules, because the protons are anti-parallel. The same holds for tritium, but polarization of deuterons in D_2-molecules is possible. The mixed molecules such as HD do not have the ortho-para distinction, because the two nuclei are not identical. Thus no such limitation exists for polarizing the nuclei in mixed hydrogen molecules.

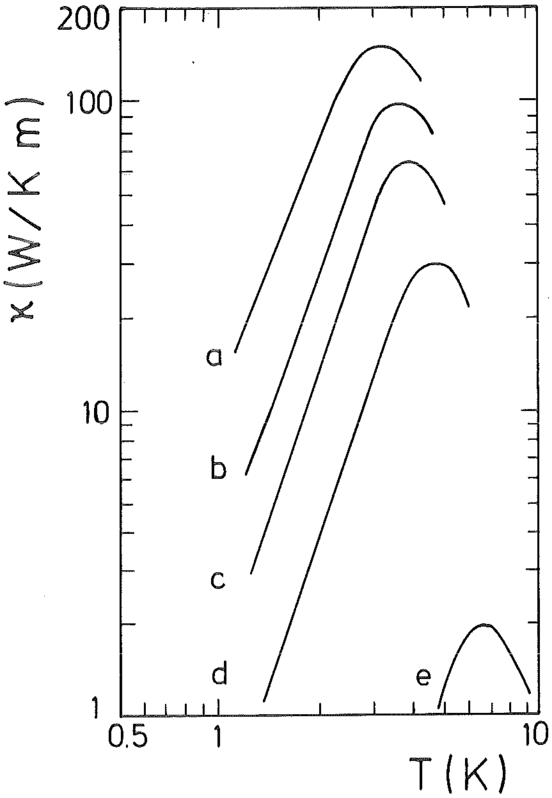

Fig.4: Thermal conductivity of solid hydrogen for various ortho-H_2 concentrations: a (0.3%), b (1%), c (2.5%), d (5%), e (30%), data from refs. 14 and 15.

The transition from the J = 1 to the J = 0 rotational state is called conversion. For isolated molecules conversion is forbidden. If an H_2 molecule is converted due to the magnetic dipole moment on another H_2 molecule, the conversion is called intrinsic. A J = 0, I_{mol} = 0 para molecule has no magnetic moment and cannot cause conversion. As a consequence the intrinsic conversion rate is quadratic in the concentration C_o of the odd species:

$$\frac{dC_o}{dt} = -KC_o^2. \tag{3}$$

For hydrogen in the solid state the conversion constant has the value K = 1.9%/h. The conversion equation of solid deuterium contains a linear term in C_o also. At C_o = 1 its conversion rate amounts to 0.06%/h.

The ground-state even rotational state is spherical, whereas the odd states are anisotropic in charge distribution. By varying the concentration of the latter, one can vary the amount of anisotropic interaction present in the sample. This has large impact on properties such as heat conductivity and spin-lattice relaxation. For a sample, which is left by itself, these features gradually change due to the odd-even conversion.

Figs.4-6 show heat-conductivities and spin-lattice relaxation times of solid H_2 and of solid HD around T = 1 - 4 K for various concentrations of ortho-H_2.

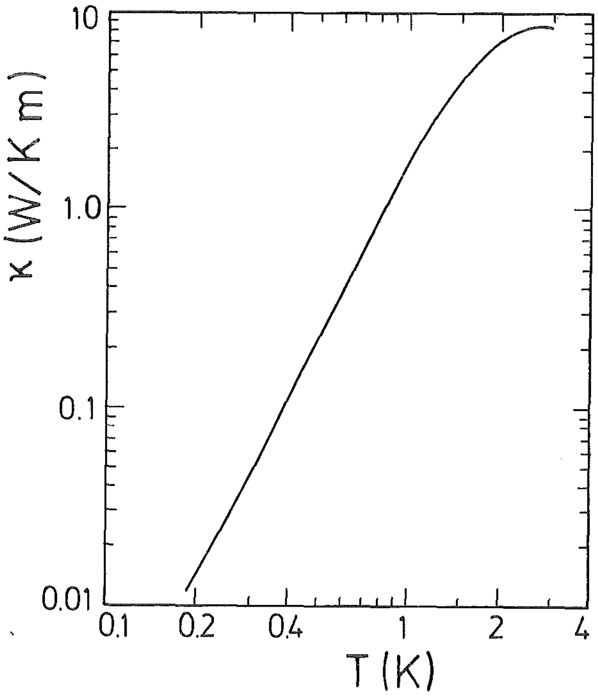

Fig.5: Thermal conductivity of solid HD[16].

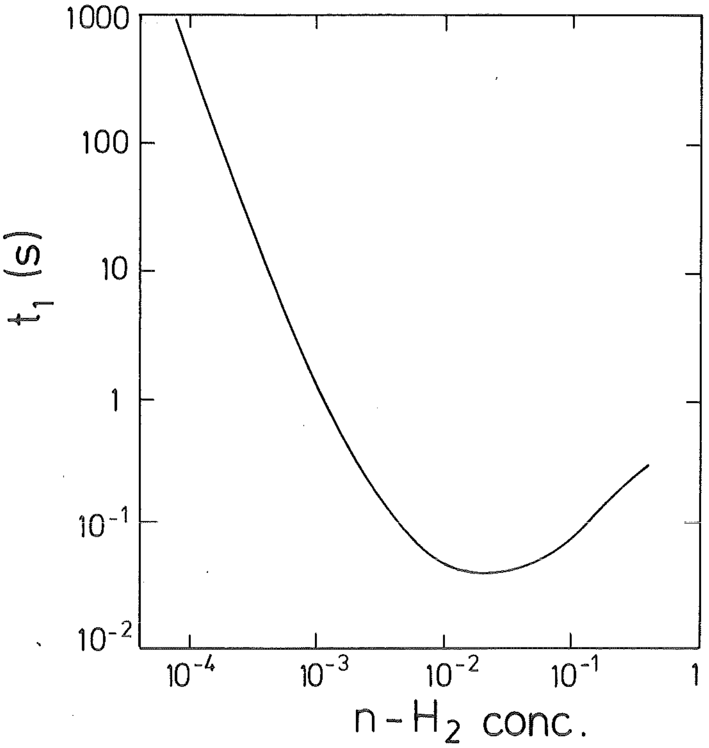

Fig.6: Proton relaxation time of solid HD for various ortho-H_2 concentrations[17], n-H_2 contains 75% o-H_2.

5. POLARIZED PROTONS AND DEUTERONS

5.1 Brute-force polarization

5.1.1 Solid H_2

In thermal equilibrium at low temperatures solid hydrogen occurs almost purely in the form of para-H_2 molecules in which the two proton spins are oriented anti-parallel to each other. In this state the protons can not be polarized. However, the ortho-para conversion is rather slow. This means that after quickly solidifying hydrogen gas from room temperature we will have a solid sample with a high temperature ortho-para distribution, i.e. 75% ortho and 25% para. It is even possible to have higher ortho-concentrations than 75% by preferential adsorption of para-hydrogen onto specific surfaces[13)]. One might imagine to polarize such a sample, employ it for a short time and produce a new sample.

The problematic factor here is the ortho-para conversion heat. In Table 4 the heat produced in 0.1 mol of solid H_2 (2.3 cm^3) has been calculated for various ortho-concentrations employing eq. (3). The ortho concentration C_o simultaneously represents the maximum polarization that theoretically can be achieved for the sample. At C_o = 25% the heat load still amounts to 47 µW. Fig.2 shows that at this heat load the dilution refrigerator reaches 22 mK, yielding a proton polarization at 15 T of 60%. The overall polarization in the sample would be 25% x 60% = 15%. At higher C_o-values the overall polarization is comparable. Hence, the conversion heat is too large for the refrigerator apart from very small samples.

C_o = ortho-concentration	conversion heat
75%	419 µW
50%	186 µW
25%	47 µW

Table 4: Heat due to ortho-para conversion produced in 1 mol H_2 for various ortho-concentrations.

Very small samples still would suffer, however, from the low heat conductivity of solid hydrogen and from the high thermal boundary resistance (Kapitza resistance) between the bead surface and the cooling liquid. Suppose we have solid hydrogen with C_o = 50% in the form of beads with a diameter of 1 mm. The heat load due to conversion released in one such bead is 0.34 µW. The difference between the temperature of the surface T_s and of the liquid T_ℓ is given by:

$$T_s^4 - T_\ell^4 = \frac{\dot{Q}}{4\pi R^2 \alpha} , \qquad (4)$$

where $\dot{Q}$ is the heat load generated in the bead and R is the bead radius. The quantity α is the surface conductivity coefficient. For the solid H_2 to liquid He interface no values for α are available in the literature at present. We adopt the value $\alpha \approx 2.5 \times 10^{-3}$ W/cm^2 K^4 which is adequate for light dielectric solids. Assuming T_ℓ = 10 mK we find T_s = 182 mK. Decreasing the bead size by a factor of 10 to a radius R = 0.1 mm does not bring a significant improvement. In this case we find T_s = 102 mK. Apart from this temperature jump there is the radial gradient over the bead itself due to the heat flow to the surface. For a homogeneously distributed heat load $\dot{Q}$ it can be found from the following expression:

$$T_c^{\beta+1} = T_s^{\beta+1} + \frac{(\beta+1)\dot{Q}}{8\pi R\lambda} . \qquad (5)$$

Here T_c and T_s are the temperatures at the centre and at the surface, respectively, for a bead of radius R. The heat conductivity $\kappa(T)$ is supposed to have a temperature dependence of the form:

$$\kappa(T) = \lambda T^\beta , \qquad (6)$$

which appears to hold for many substances below 4 K.

From the heat conductivity data[15)] for solid hydrogen we obtain $\beta = 3$ and $\lambda = 4 \times 10^{-3}$ W/K^{4}m for an ortho-concentration of 50%. Inserting these values in eq. 5 for the bead with R = 1 mm and T_s=182 mK, we find for its central temperature T_c = 348 mK. For the 0.1 mm bead with T_s = 102 mK the result is T_c = 125 mK.

These results sufficiently show that ortho-para conversion prevents brute-force polarization of protons in solid H_2.

5.1.2 Solid D_2

Table 3 shows that the lowest molecular state of D_2 is ortho-D_2 in which state the deuterons are polarizable. Hence after solidifying and further cooling of the D_2 in a magnetic field one will have a slowly rising polarization. The conversion time can be decreased to an acceptable value by introducing a paramagnetic impurity such as O_2. The very high magnetic field and the very low temperature, that are required (see fig.2) have prevented such samples from being produced up to now.

5.1.3 Solid HD

The para-ortho distinction does not apply for HD-molecules, because proton and deuteron are distinguishable particles. Thus protons and deuterons can be polarized in HD. A proton polarization of 40% has been achieved by Bozler et al.[18)] at T = 23 mK and B = 10 T. A small amount of ortho-H_2 had been added to the HD in order to decrease the proton spin-lattice relaxation time, which otherwise would have been on the order of many days.

By the subsequent conversion of the ortho-H_2 the spin-lattice relaxation time will increase steadily in such a polarized sample, because the proton-lattice coupling in para-H_2 is very weak. This decoupling of the polarized protons from the lattice opens up the possibility to operate the sample at moderate fields and higher temperatures, e.g. 4.2 K, where relaxation times exceeding a day are expected if one has waited long enough[19)]. At such temperatures the thermo-dynamic properties of HD (such as heat capacity and heat conductivity) are much less troublesome than at 10 mK. E.g., the heat conductivity of HD at 4.2 K is in between that of stainless steel and copper, whereas at 10 mK it is far below that of stainless steel.

5.2 Dynamic polarization

5.2.1 Solid H_2

The process of dynamic polarization requires the presence of paramagnetic electrons. If these are introduced in ortho-H_2 (the polarizable, metastable specimen) they speed up the conversion rate enormously[20]. Because of this reason dynamic polarization of protons in solid H_2 can not be realized.

5.2.2 Solid D_2

In D_2 the situation is just opposite to H_2 (see Table 3). Here the lowest molecular state is the one in which the nuclei can be polarized. Hence the introduction of para-magnetic electrons has a favourable side-effect in decreasing the conversion time to the desired state. Over 20 years ago about 0.35% polarization has been achieved[21] at $B = 0.85$ T and $T = 4.2$ K. Since then this has not been developed any further. Extrapolating naively these early results, one may expect a polarization over 60% for $B = 15$ T and $T = 0.4$ K. Another increase might come from optimizing the concentration of the paramagnetic centers and the microwave irradiation.

5.2.3 Solid HD

In the HD-molecule the nuclei are different, hence there is no symmetry requirement limiting the proton and deuteron polarization. Also the problems of conversion heat and conversion time are absent. This makes HD to an attractive substance.

The dynamic polarization of the nuclei in HD has been studied by Solem[20]. The paramagnetic electrons were created by exposing the solid samples to a thick target bremsstrahlung beam having a maximum energy of 60 MeV. The radiation breaks up a fraction of the HD-molecules, yielding H- and D-atoms. The D-atoms are not stably trapped and combine to D_2-molecules. A part of the H-atoms is stably trapped in the solid before recombination and can be used for the polarization process. Their EPR (electron paramagnetic resonance)-spectrum has two lines, some 500 G apart. In pure HD the electrons relax too slowly for effective dynamic polarization. This problem can be solved by mixing in a fast-relaxing, paramagnetic impurity such as O_2. A concentration of some parts in 10^4 appears to be sufficient.

The total radiation dose was about $1.5x10^7$ R. From the size of the electron resonances the density of the paramagnetic centers was estimated to be 10^{-4} H-atoms per HD-molecule. After irradiation the proton relaxation time dropped to about 20 s and then increased to about 70 s (at 4.2 K) at a rate characterized by ortho-para conversion, because some of the HD dissociated during irradiation had recombined to H_2 and D_2.

Polarization determinations were carried out at two values of the magnetic field: 0.83 T and 1.24 T, and at two temperatures: 4.2 K and 1.2 K. A maximum proton polarization of 3.75% was obtained at 1.24 T and 1.2 K. Only one of the EPR lines was driven. A doubling might be achieved by using also the other line. If in addition the field were raised to 15 T and the temperature lowered to 0.4 K, a proton polarization close to 100% may be expected. A maximum deuteron polarization between 0.3 and 0.4% was reached in this experiment. In a previous experiment[22)], in which the H_2 and D_2 impurities were allowed to convert for a longer time, a polarization around 1% was obtained. Starting from this value a polarization of 2x12x3 = 72% may be expected by driving both EPR lines, by increasing the field to 15 T and by lowering the temperature to 0.4 K.

6. PROSPECTS FOR POLARIZING DT

6.1 Tritium decay heat load

In the previous chapters we have seen that polarization of protons in H_2 can not be realized, neither by the brute-force method nor by dynamic methods. The same holds for tritium. Therefore, the only useful substance appears to be DT if both polarized deuterons and polarized tritons are required to be present in one sample.

The difficulty that has to be overcome is the heat generated in the tritium decay. Tritium is a β-emitter with a half-life of 12.3 y and an end-point energy of 18.6 keV. The average β-energy is 5.7 keV. The range of these low-energy β's is only very small, of the order of 0.01 mm in solid hydrogen. Thus for samples with larger dimensions all the β-energy will be dissipated within the sample itself.

The activity contained in 1 mol of DT is $A = 1.07\text{x}10^{15}$ Bq (= 29 kCi). This corresponds to a heat load of $\dot{Q} = 1.07\text{x}10^{15}\text{x}5.7\text{x}10^{3}\text{x}1.6\text{x}10^{-19} = 0.98$ W. In the next sections we calculate the thermal gradient that is accompanied by this heat load in some targets due to the Kapitza resistance and to the heat conductivity within the targets. First, we consider massive beads consisting of pure DT. Such beads would be useful for injection into magnetically confined plasma reactors. For ICF-reactors usually multilayered hollow beads with an inner shell of DT are considered. The thermodynamic properties of such a bead can be quite different from those of a pure DT bead. The multilayered beads are discussed in section 6.4.

6.2 Brute-force polarization of DT

Let us consider a DT-bead with a radius of 1 mm. It contains $0.21\text{x}10^{-3}$ mol DT, assuming a molar volume of 20 cm^3 in the solid state. The heat released in the bead is $0.21\text{x}10^{-3}\text{x}0.98 = 0.21$ mW. This is obviously too much for the brute-force polarization method, see fig.2. If we reduce the size to 0.1 mm radius, the heat load becomes 0.21 μW, which is of the desired order of magnitude. However, as in the example of section 5.1.1., the Kapitza resistance causes an enormous temperature jump between the cooling liquid and the bead. Assuming a liquid temperature T_ℓ = 5 mK and inserting again α = 2.5×10^{-3} W/cm^2 K^4, we find for the temperature of the bead surface T_s = 509 mK from eq.4. This implies that even much smaller beads should be used, or very thin sheets of DT. This makes the concept of brute-force polarization practically unfeasable for application in nuclear fusion.

6.3 Dynamic polarization of DT

6.3.1 Radical creation

As for dynamic polarization the β-radiation also has a positive aspect: it causes the dissociation of a fraction of the DT-molecules, creating in this way the paramagnetic electrons that are necessary for the polarization process. The question arising in this respect is, whether the rate of dissociation is adequate, i.e. whether the desired amount of free atoms is attained and whether this occurs in a reasonable time after solidification. Preliminary conclusions can be drawn from two experiments.

In the first experiment[23)] a D_2 sample had been doped with 1% T_2. The EPR-spectra of the D- and T-atoms were studied. It was found that the concentration of free atoms of both species reached saturation after about 5 days. For the free atoms concentration a lower limit was deduced of 3×10^{-5} per D_2-molecule. In first approximation the saturation time would scale down to 2.4 hours for a DT-sample. A higher saturation concentration of free atoms may be expected for DT, because the recombination rate will be the same at the above concentration of free atoms, whereas the dissociation rate is 50 times higher. No polarization measurements were reported in this work.

The second experiment[20)] has been discussed already in the previous chapter. An HD-sample was irradiated by 7×10^5 R/h thick target "bremsstrahlung" with a maximum energy of 60 MeV. The exposition time was 21 h, resulting in a total dose of 1.5×10^7 R. The free H-atom density was estimated to be $\sim10^{-4}$ per HD-molecule. An equivalent dose will be reached in DT in 13 min. after solidification.

The results of both experiments indicate that sufficient free atoms will have been generated within a few hours after solidification. The polarization build-up time is shorter for higher free atom concentrations. Therefore, a high concentration is favourable. However, with increasing amounts of free D- and T-atoms, the concentrations of ortho-H_2 and para-D_2 will also increase. These molecules give rise to a significant reduction of the nuclear relaxation time. This time may, of course, not become shorter than the time necessary to transport the sample from the polarizing cryostat to the reactor vessel. If this problem shows up, the microwave power should be increased, which decreases the polarization build-up time.

6.3.2 Thermal aspects

Let us consider again a 1 mm radius DT bead and assume a liquid temperature of T_ℓ = 200 mK. The 0.21 mW/bead are no problem for large dilution refrigerators at 200 mK. The largest ones reach cooling powers up to 10 mW at this temperature[24)]. Hence several of these beads can be cooled simultaneously.

We apply again eq. (4) to find the surface temperature T_s of the bead, using $\alpha = 2.5\times10^{-3}$ W/cm^2 K^4. The result is T_s = 904 mK. This value is mostly determined by the surface conductivity coefficient α. Increasing the liquid temperature T_ℓ from 200 to 400 mK, e.g., yields T_s = 913 mK. Increasing the conductivity with a factor 2 gives T_s = 760 mK, decreasing it with a factor 2 yields T_s = 1075 mK. Hence, although α is not well known, one can expect a bead temperature around 1 K or somewhat lower. The thermal gradient over the bead, see eq. (5), appears to contribute only little due to the relatively high conductivity of solid DT.

It may appear that 1 K is not low enough to achieve high deuteron polarizations. In this case one might decrease the size of the bead again. For a 0.1 mm radius bead the result is T_s = 511 mK. Straight forward extrapolation to a magnetic field of B = 15 T of the results on HD discussed in section 5.2.3 yields at 1 K a tritium polarization $P(^3H)$ = 100% and a deuteron polarization $P(^2H) \approx 30\%$. At 0.5 K one would expect $P(^3H)$ = 100% and $P(^2H) \approx$ 60%. Experiments on DT itself will have to show, whether these expectations are justified.

6.4 Polarizing DT in ICF pellets

In the concept of Inertial Confinement Fusion the nuclear fuel is in the centre of a small pellet, surrounded by one or more layers of different material. Early laser fusion pellets, e.g., usually consisted of a thin glas balloon filled with high pressure deuterium - tritium gas. A recent design for a heavy ion fusion pellet, the HIBALL-2 pellet[25)] is shown in fig.7. The innermost shell is a thin layer of frozen DT. The interior of the pellet is empty. This construction allows for a very high density after compression. It is also favourable from a thermodynamic point of view, because the temperature jump, due to the Kapitza resistance to liquid helium, is reduced by the significant increase in surface area compared to a full solid bead. This

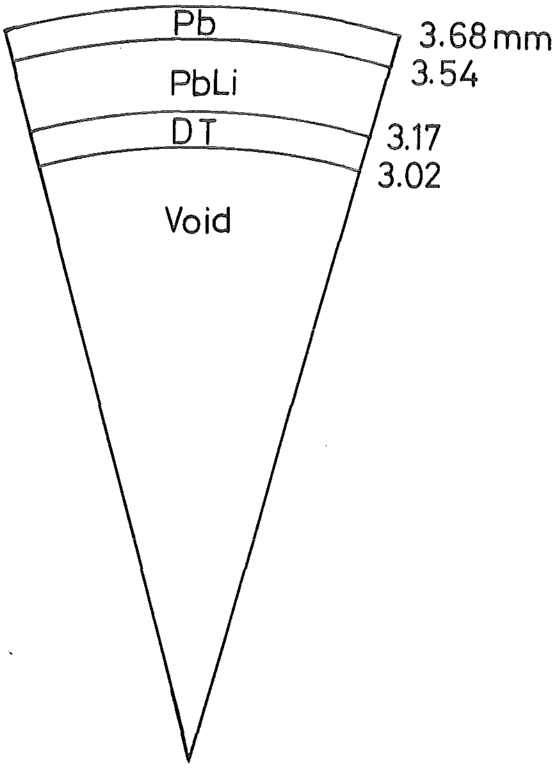

Fig.7: Design of the HIBALL-2 ICF pellet.

advantage, however, is reduced by the occurrence of extra thermal resistancies. Quantitative estimates are given below.

The outer shells of the pellet are metalllic. This is intolerable in view of the dynamic polarization mechanism. The required microwave radiation of about 400 GHz can not penetrate metallic layers due to the skin effect. At this frequency the skin depth of lead at low temperature is of the order of 0.1 µm, three orders of magnitude below the thickness of the lead layer. Hence non-metallic tamper and pusher layers have to be employed. In view of the fact that pellet design is still an open question and many types are discussed, this limitation should not be regarded as serious at this stage.

We will consider now a pellet with the dimensions of the HIBALL-pellet, but having non-conducting tamper and pusher shells. It contains 4 mg DT, corresponding to $0.8\text{x}10^{-3}$ mol. This generates a β-decay heat load of $0.8\text{x}10^{-3}\text{x}0.98$ W = 0.78 mW. The pellet is cooled with liquid helium of T_ℓ = 200 mK. In fig.8 a band of possible temperature profiles in the pellet is shown, which is obtained by allowing large variations in the thermal properties of tamper and pusher. The largest jump is expected to occur at the outer surface of the pellet due to the surface boundary (Kapitza) resistance to liquid helium. The surface

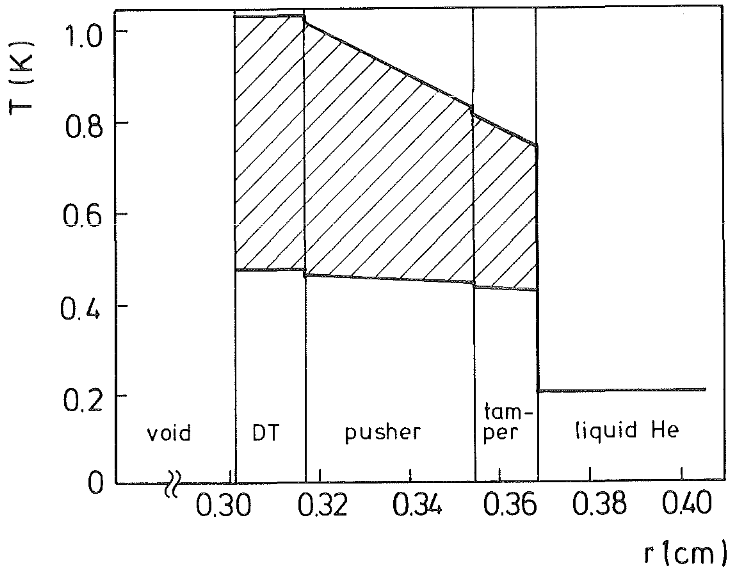

Fig.8: Calculated thermal gradients over the HIBALL-2 pellet for a large variation in tamper and pusher thermal properties

conductivity coefficient α was varied from 1.5×10^{-3} (upper curve) to 1.5×10^{-2} $W/cm^2\ K^4$ (lower curve). At the interior boundaries the surface conductivity was assumed to be 10 times higher. The bulk thermal conductivity κ of tamper and pusher was varied between 0.01 and 0.1 W/Km. These values range in the lower and middle region of conductivities[26).

The resulting average temperature of the DT-layer varies between 0.5 and 1.0 K. Hence, a variation of only a factor of 2, while the thermal properties of the pellet were varied within a factor of 10. This insensitivity increases towards higher temperatures, because of the steep increase of the surface and bulk conductivities.

These results show that it is quite probable that the DT-layer can be cooled to at least 1 K. In favourable cases temperatures down to 0.5 K or lower might be attainable. This would mean tritium polarizations close to 100% and deuteron polarizations from 30-60%, following the arguments of section 5.2.3. These polarization values are, however, extrapolations of old experiments at low polarizations. Deviations in both directions are possible.

7. POLARIZATION DECAY IN TRANSFER TO REACTOR

After finishing the polarizing procedure, the nuclear polarizations will start to decay. The relaxation rate is dependent on temperature and magnetic field and on the presence of magnetic impurities. Without magnetic impurities the only relaxation mechanism for $I = 1/2$ nuclei would be the interaction between the nuclear moment and lattice vibrations, which gives theoretical relaxation times of many years[27). In pure HD proton relaxation times up to a day have been observed[19). The deuteron may relax additionally by quadrupole interaction with the crystal fields, but this interaction is also very weak. In practice, the dominant relaxation mechanisms are interactions with electron paramagnetic impurities and cross relaxation with the $J = 1$ homonuclear impurities ortho-T_2 and para-D_2[28). In DT such impurities are created continuously by the radiation damage of the β's from tritium.

Relaxation times in HD have been measured after irradiations with γ-rays[17,20]), electrons and protons[19]). The results vary strongly as a function of the radiation dose. The lowest value of 20 s was measured for the highest dose: 1.5×10^7 bremsstrahlung, accumulated in 21 h at a sample temperature somewhat above 4.2 K in zero magnetic field. The relaxation time in DT decreases with time because of the continuous accumulation of radiation damage. The above dose of 1.5×10^7 will be reached in 13 min after solidification. At that instance a relaxation time can be probably expected of the order of the above 20 s, though deviations will occur due to different ionization density, irradiation time, sample temperature and magnetic field. The exact value of the relaxation time immediately after having polarized the sample is of crucial importance. If it is around 1 s it seems to be almost impossible to inject a polarized sample, if it is around 1 min or more then there seems to be a good chance. This implies that the polarizing procedure should be made as short as possible.

The transfer channel between the polarization refrigerator and the reactor vessel possibly will be divided into two parts. In the first part the pellet is transported through liquid helium of 1.2 K. This will limit the temperature of the pellet to below 2 K. In the second part the pellet is shot through vacuum into the reactor. Here the β-radiation heat will go almost exclusively into the DT-lattice. Fig.9 shows the development of the DT lattice temperature with time after the pellet has been taken from the liquid. The lattice specific heat has been calculated from a Debye temperature $\Theta_D = 110$ K[13]). The contributions to the specific heat of the electronic and nuclear spin systems appear to be negligible. The calculation assumes that all heat remains in the DT-lattice. In a layered pellet part of the heat will flow into the other layers, of course. Fig.9 shows a steep increase in the first second, followed by a much slower rise afterwards. The relaxation time decreases at increasing temperatures. Extrapolation of the results obtained in ref.17 suggests a relaxation time of some seconds at T = 10 K. This means that the vacuum transfer should occur very quickly, which is inherent to this method.

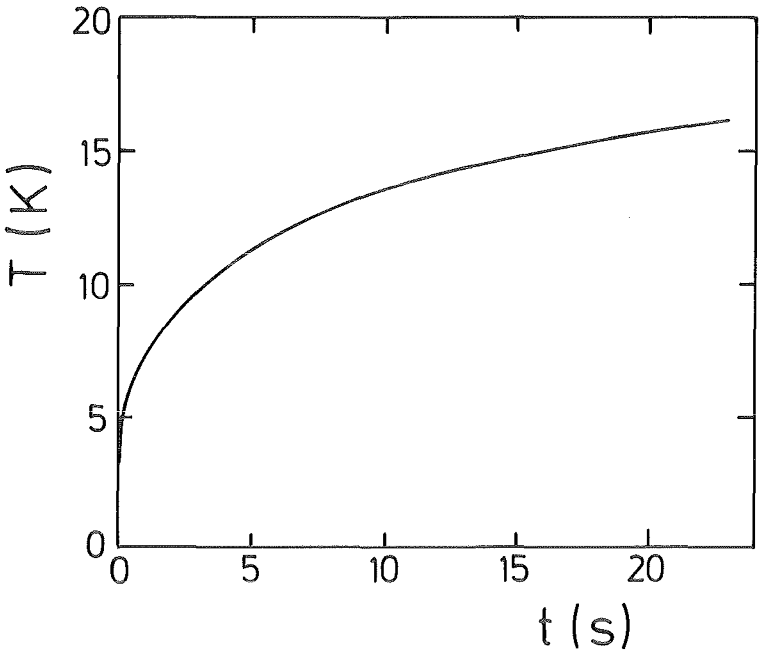

Fig.9: Adiabatic warming-up curve of solid DT due to the tritium β-decay heat.

In a plasma reactor with magnetic confinement the pellet will start to interact with the plasma as soon as it enters the cavity. In an inertial confinement reactor it has to travel to the center before the burning is initiated. Upon entering the reactor cavity the pellet will warm up additionally by the blackbody radiation of the cavity surface. This has been discussed in ref.29 for the HIBALL reactor design, assuming a surface temperature of 500°C. It was shown that a pellet velocity of 200 m/s would suffice to keep the DT temperature below 14 K.

The final step - polarization decay during the burn phase - will not be discussed here. The available studies[1,4)] point in the direction that the nuclear polarization will be largely preserved during burn.

8. SUMMARY

The employment of polarized fuel is an interesting topic for the development of nuclear fusion reactors. Completely polarized DT fuel would provide a 50% increase in fusion cross section, meaning relaxed confinement conditions and earlier break-even. The accompanying anisotropic emission of the neutrons may also be advantageous for reactor design. The price to be paid for polarization is an extra amount of technical equipment, mainly cryogenics if it is considered to polarize solid fuel as was done in this paper. However, cryogenic techniques are considered anyway for fuel injection, apart from polarization desires. It has been found for tokamaks that higher densities and longer confinement times can be obtained when the fuel is injected in the form of frozen solid beads. The most recent designs for ICF pellets conceive a multilayered hollow structure with an inner layer of frozen DT. Hence the concept of polarizing solid fuel fits well within the present ideas of fuel injection.

Polarized proton and deuteron targets are well established in the nuclear and particle physics community. As target material, however, always hydrogen compounds have been taken up to now. The reason being that $^1H_2$ (2 protons) can not be polarized, because at low temperature the molecules are in the para-state with proton spins aligned opposite to each other. The same is the case for tritium molecules. Polarization is possible for D_2 and the bi-isotopic molecules. Hence, polarization of a D-T mixture can only be achieved if DT-molecules are employed. No polarization data on DT are known, but some experience has been made with HD, in which polarizations of a few % have been achieved over a century ago. It may be assumed that the thermodynamic properties of HD and DT are very similar. Extrapolation of the HD results to what is technically achievable nowadays leads to the conclusion that high polarizations in DT are possible in spite of the self heating by the β-decay of tritium. The largest problem may be the nuclear relaxation time, which may be as short as a few seconds. This would require to transport the sample after polarization to the reactor in about a second.

Polarization experiments on DT itself will have to reveal, whether high polarizations can be achieved and retained. It seems worthwhile to carry out experiments on this, because they only require the application of well established techniques. In case they are successfull, they would bring the peaceful employment of fusion energy a step closer to its realisation.

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