Double Chooz and the search for short range anti-neutrino oscillations

Francis X Hartmann

Division of Particle and Astroparticle Physics, Max-Planck-Institut fuer Kernphysik, Postfach 10 39 80, D-69029 Heidelberg, Germany

f.hartmann@mpi-hd.mpg.de

Abstract. The Double Chooz Experiment seeks to search for short range antineutrino oscillations from the nuclear reactors at the Chooz Nuclear Power Station operated by Electricite de France in Northeastern France. The measurements are of interest to constraining the value for θ_{13} in current neutrino oscillation models. New scintillator types based on beta-diketone and pH stabilized carboxylic acid chemistry are described. New results from the study of these scintillators in the context of the detector design are reported.

1. Physics and Concept of the Experiment

1.1. The physics

The nature of the neutrino is not well understood. The neutrino state arising from electron capture or nuclear beta decay is currently viewed [1] as a superposition of three neutrino mass eigenstates (*i*=1,2,3). The "electron neutrino" is the neutrino state of the three mass eigenstate particles produced in association with the charged lepton of the electron "flavor" (α). As such, the states of the neutrino types are defined in relationship to the charged current. The electron's loss of electric charge through the weak interactions leads not to an elementary particle, in a sense to which we are accustomed to defining such particles, but a mixture of three entities.

In the current model of three neutrino mass eigenstates, the state of the neutrino associated with the electron can be found from the matrix equation $\langle \nu_e \,|\, j \rangle$ whose three factors are:

$$\begin{pmatrix} \nu_e \\ \nu_\mu \\ \nu_\tau \end{pmatrix} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\theta_{23} & \sin\theta_{23} \\ 0 & -\sin\theta_{23} & \cos\theta_{23} \end{pmatrix} \begin{pmatrix} \cos\theta_{13} & 0 & \sin\theta_{13} e^{i\delta} \\ 0 & 1 & 0 \\ -\sin\theta_{13} e^{-i\delta} & 0 & \cos\theta_{13} \end{pmatrix} \begin{pmatrix} \cos\theta_{12} & \sin\theta_{12} & 0 \\ -\sin\theta_{12} & \cos\theta_{12} & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \nu_1 \\ \nu_2 \\ \nu_3 \end{pmatrix} \quad (1)$$

The angle θ_{23} is known from atmospheric neutrino studies [1] to be greater than 37 degrees (to a maximum of 45 degrees) and the angle θ_{12} is known from solar neutrino and reactor experiments [1] to be 30 to 35 degrees. The neutrino state is given by:

$$\nu_e = \left(\tfrac{\sqrt{3}}{2} \cos\theta_{13},\ \tfrac{1}{2} \cos\theta_{13},\ \sin\theta_{13} \right) \bullet \mathbf{B} \quad (2)$$

where **B** is the basis vector of the three massive neutrinos ν_j (one mass can be zero). The numbers are simplified by using $\theta_{23} = 45$ degree, $\theta_{12} = 30$ degree and setting $\delta = 0$ (when CPT holds, a non-zero value indicates CP violation). The propagation of the neutrino state through space and time involves many interesting physics issues which have some complicating aspects [2]. In the simple situation here, we can summarize that the electron neutrino state will propagate in distance L, such that the probability of detecting an electron neutrino state is given by:

$$\begin{aligned} P_{\nu_e \to \nu_e} &= \left| \sum_{j=1}^{3} \langle \nu_e | \langle j | \nu_e \rangle \langle j | \exp[-i(E_j t - p_j L)] | j \rangle \langle \nu_e | j \rangle | \nu_e \rangle \right|^2 \\ &= \tfrac{5}{8} + \tfrac{3}{8}[\sin^4\theta_{13} + (1 - \sin^4\theta_{13})\cos\Phi_{12}] - \tfrac{1}{2}\sin^2 2\theta_{13}[(\tfrac{5}{8} + \tfrac{3}{8}\cos\Phi_{12}) - \tfrac{3}{4}\cos\Phi_{13} - \tfrac{1}{4}\cos\Phi_{23}] \end{aligned} \quad (3)$$

where the phase factor differences $\Phi_{ij} = (E_i - E_j)t - (p_i - p_j)L \to (\Delta m_{ij}^2 L / 2E)$ per [1,2].

Consider the case of a 2 MeV neutrino. Close to the source, the $\cos\Phi_{12}$ terms approach unity since the experimental value [1] $\Delta m_{12}^2 = 0.00008$ eV implies an oscillation length of 62 km. Also $\Phi_{13} \approx \Phi_{23}$ since $\Delta m_{23}^2 = 0.00250$ and $\Delta m_{13}^2 = 0.00258$ eV [1] imply nearly identical oscillation lengths of 1.98 km and 1.92 km respectively. In equation 3 the first two terms sum to *ca*. 1.0; the second term oscillates such that at the minimum (where the $\cos\Phi_{13}, \cos\Phi_{23} \cong -1$) the neutrino deficit is directly equal to $\sin^2 2\theta_{13}$. The minimum occurs close to 1.0 km.

1.2. Concept of the experiment

Motivated by the foregoing discussion, a suitable experiment [3] would be where one can search for anti-neutrinos from a nuclear reactor at a distance of about 1.0 km from the core. A suitable situation exists in the town of Chooz in the northeastern Departement of Ardennes in France. Two cores, for a total thermal power P of 8.4 GW are operated by Electricite de France. The number of neutrinos per second is $N_\nu \approx 2 \times 10^{11} P(watts)$. The site of the first Chooz experiment [4] lies 1.05 km from the midpoint of the two reactor cores. A first "Far Detector" is sited here. A second "Near Detector" is planned 400 meters from the same point where the oscillation probability is much less (about 15 % of the maximum) so to minimize uncertainties having to do with the reactors themselves. The observable deficits are summarized in figure 1.

For the specific case of $\theta_{13} = 5$ degrees, figure 1, there is a 3% reduction of the neutrino flux at 1.0 km. The second detector permits lowering the systematic uncertainties by monitoring the actual neutrino spectrum in the near absence of oscillations. To see such a signal is still challenging. The first Chooz experiment achieved a limit of $\theta_{13} \leq 11.4$ deg for $\Delta m_{23}^2 = 0.0025$. To improve upon the situation the detector will incorporate a number of new features as described next.

2. Description of the detector

The Far Detector scintillators are described here. The Near Detector is planned to be identical, except it will have different background considerations. The Far Detector is located in a tunnel with an overburden of 300 meters water equivalent. The central region consists of a blue fluorescent scintillator containing Gd.

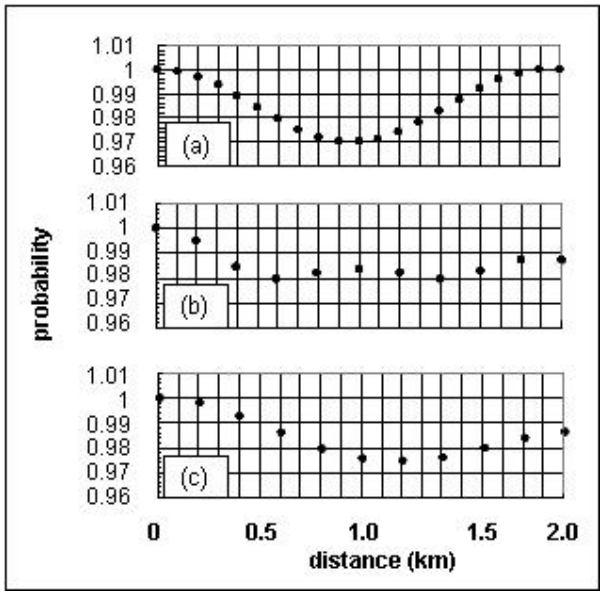


Figure 1. Neutrino Oscillations. The figure at the left (a) shows the oscillation minimum at about 1 km for a monoenergetic 2 MeV neutrino. The deficit is about 3%. A continuum of neutrinos is emitted by the U-235 fuel [5] which, in a simple histogram calculation (b), shows the reappearance of the lower energy neutrinos below 2 MeV leading to some reduction in the magnitude of the deficit. The neutrino detection method imposes a threshold (modeled as 2 MeV), so in (c), the deficit grows while the minimum broadens due to the presense of higher energy neutrinos.

The target scintillator, figure 2, gives a prompt signal following deposition of the positron energy and its annihilation; microseconds later, a delayed signal due to neutron capture on Gd follows. The follow-on gammas emitted from the Gd are detected in the concentric gamma catcher region. The target and gamma catcher regions involve new scintillator formulations. A non-scintillating buffer region shields the central targets from background gammas. The phototubes are mounted on the wall of the stainless steel buffer tank. These regions are contained in a larger tank which serves as an inner veto shield. Above these tanks is an outer plastic veto scintillator to "tag" cosmic ray events.

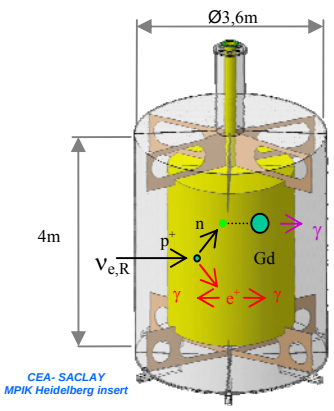


Figure 2. Depiction of a Neutrino Intereraction in the Gamma Catcher and the Neutrino Target. The anti-neutrino interaction with the proton of the Neutrino Target (10 m^3, 20% ortho-phenylxylylethane, o-PXE, and 80% normal-dodecane, n-C12) yields a prompt signal due to a positron summed with its own annihilation energy, thus recording the reactor neutrino energy. The microsecond scale delayed capture of the neutron on Gd leads to higher energy multi-MeV gammas that are captured in the Gamma Catcher (22 m^3, o-PXE, n-C12 and a C12-C14 mineral oil, MO). The Gamma Catcher is surrounded by an optically transparant liquid buffer region (102 m^3, MO and tetra-methyldecane, TMD). An inner veto (muon) detector (87 m^3, TMD and a mixture of linear alkyl benzenes, LAB) encases the main detector.

The neutrino rate is about 70 nu/day. Correlated background signals due to stopping and reacting muons or fast neutrons are expected to be about 2 /day. Accidental backgrounds originating from gammas from the rock or a fast neutron are expected to be < 5 /sec in the energy window of interest (E > 0.7 MeV). In addition errors are reduced not only due to background reduction but improved target weighing (no. of protons) and the use of two reactors to factor out reactor specific quantities. Reductions of the systematic uncertainty from 2.7% (Chooz) to 0.6% (Double Chooz) and better statistics is expected from a number of improvements: larger detector volume, longer exposure, two

detectors, lower e+ threshold, improved e+ and n detection efficiency, background shielding, radiopurity and overburden. One key improvement is in the scintillator which we describe next.

3. New Gd scintillators

3.1 Neutrino target chemistry

A carboxylic acid (CBX) is shown in figure 3 where R represents an arbitrary hydrocarbon of choice. A base will remove the proton of the –OH leaving a negative anion of "triangular" shape. The inclusion of an additional two carbons allows one to achieve a "hexagonal" shape seen in the center of figure 3 which is better configured to grab the Gd^{3+} ion. The choice of a double bond in the extension leads to a sp^2 planar hybridization of the complex such that all atoms of the hexagon lie in the same plane. The resulting enol is a tautomer and rapidly rearranges by movement of a proton to the third structure of figure 3. Here, the C=O is a ketone, the carbon alpha to it holds the moveable proton and the carbon beta to it is a second ketone – thus a "beta-diketone" (BDK). Removal of the proton leads to a negative anion of hexagonal shape that bonds to the Gd with the added feature of resonance stabilization via the out-of-plane p orbitals. Such complexes are much more stable than carboxylic acids, in fact they are stable to temperatures just above 200 deg C. They have considerable vapour pressures that can be used in their purification.

The original scintillator of Reines [6] was based on the use of propionic acid (R having three carbons). The cation was Cd++. The first complexes of Gd utilized ethylhexanoic acid [7]; Palo Verde made significant work with ethyl-2-hexanoic acid [8], and describe well their observed problems. The first Chooz experiment used gadolinium nitrate in hexanol dissolved in the organic scintillator. In the Double Chooz project, the desire is to have a very stable scintillator that lasts out to at least five years. As a consequence we developed, at MPIK, metal based scintillator systems based on the specific acid TMHA. These are produced in a pH stabilized manner [9]. The approach keys on providing a defense against hydrolysis reactions.

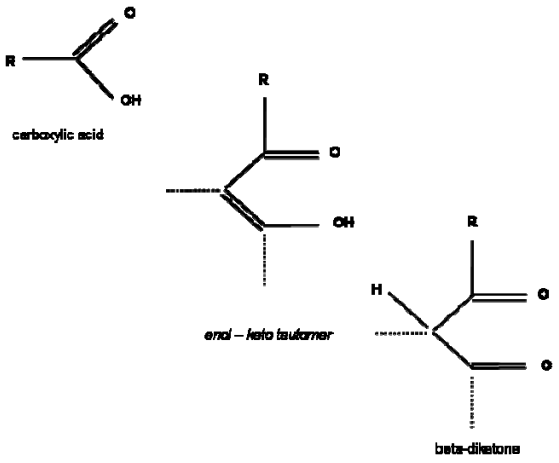


Figure 3. Carboxylic acid (CBX *e.g.* TMHA, section 3.3) and beta-diketone (BDK *e.g.* THD, section 3.2) ligands used in the Gd molecular investigations.

The first Gd-TMHA scintillators were made at MPIK in 2003 and first presented in detail in [10]. Investigations over the course of four years are reported in Section 3.3. The BDK scintillators [11] are described next in 3.2.

3.2. BDK scintillators

Beta-diketone systems are interesting from the viewpoint of "neutrino physics chemistry", that is "putting the right nucleus in the right place at the right time". It is useful to summarize some background information. The first use of beta-diketones in this regard was to produce gaseous forms of Ho-163 in measurements [12] that currently set the best limit on the neutrino mass (electron based) [1]. These experiments followed some ideas communicated to us by Davis who had tried to put Be in the gas phase as part of a Li solar neutrino experiment [13] using pentane-2,4-dione (acetylacetonate). In the LENS project [14] two scintillator systems were tested in prototype cells for an Indium solar neutrino experiment at Gran Sasso, one of which was the BDK form of Indium and the other a form of indium hydroxide suspended with the help of added carboxylic acid. From such experience, we investigated and developed the beta-diketone versions of Gd for a number of ligand species. The final system is Gd(III)-tris-(2,2,6,6-tetramethyl-hepane-3,5-dionate) or $Gd(THD)_3$. The structure takes the

basic backbone of the acetylacetonate and adds to it two tert-butyl groups whose steric effects accomplish improved properties in outer and inner sphere coordination chemistry and in vapor pressure. Stability of such Gd complexes was observed at Princeton when radioactively labelled complexes of $Gd(THD)_3$ were first studied by us [15] at elevated temperatures during testing of the Ho-163 neutrino mass experiment.

The six main areas that concern the development of a scintillator of this type are: molecular stability, light yield, optical absorbance, solubility, overall system stability and radopurity.

3.2.1. Molecular stability and light yield. The structure of the Gd complex is shown in the inset to figure 4 [16]. The central region is strongly bonded via resonance stabilized coordination. The resonance structures have some absorbance in the ultra-violet. We develop the scintillator so as to construct energy transfer pathways. The pathways are studied using a model based on [17] that has now been extended to this more complicated situation [18]. As a consequence we attain a light yield for the project of about 45 % relative to anthracene (recorded versus BC505).

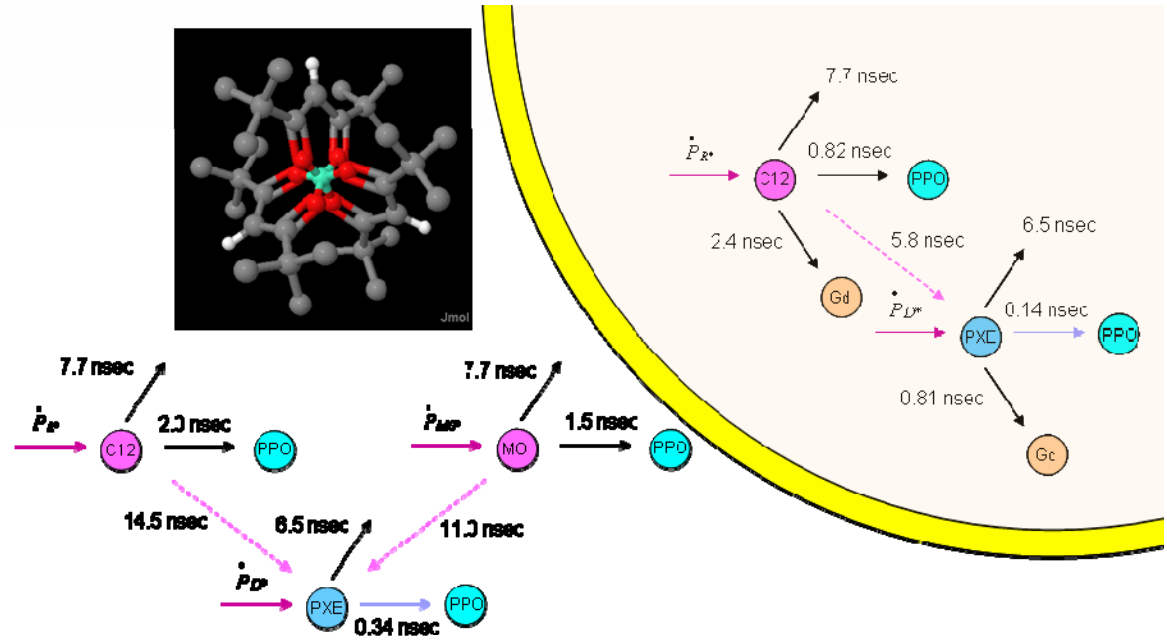


Figure 4. Molecular structure and light yield. The target in the upper right creates light via dodecane C12 and PXE channels, with one cross-transfer from C12 to PXE. The light yield in the gamma catcher is designed to match the target light yield and match as well in density. The degrees of freedom allow for one free parameter, the ratio of PPO to PXE in the gamma catcher. This freedom allows us to tune the pulse shapes so as to achieve a neutrino target – gamma catcher difference. At issue is adjusting the energy transfer pathway of the C12→PPO versus C12→PXE by PPO adjustment above and below a Forster-Dexter effective critical concentration as found for C12. Time constants shown are determined from the model after experimental determination of pairwise transfer coefficients. Pulse shape measurements are also made.

3.2.2. Absorbance, solubility and overall scintillator stability. Figure 5 summarizes the absorbance characteristics of the neutrino target. The issue of solubility over five years is a complicated subject that requires an understanding of the fundamental aspects of the inner and outer coordination sphere chemistry of Gd which is itself influenced by the nature of the coordinated ligands and their bonding.

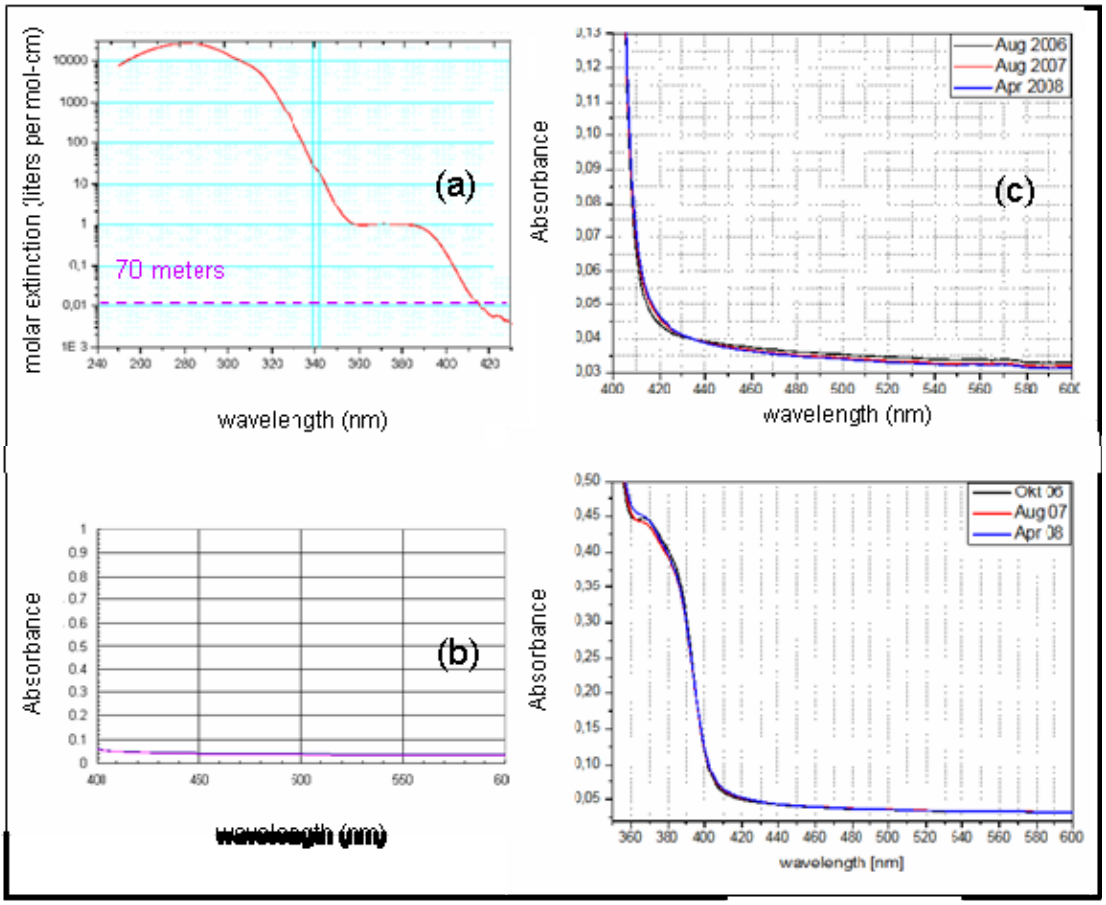


Figure 5. Absorbance, Solubility and Overall Stability. In the upper left (a), we report a precise measurement of the molar extinction coefficient of the Gd material [18]. In the visible region >410 nm, the Gd component, at 1 g/l, has an absorbance length well in excess of 70 meters. In the lower left (b) the scintillator minus fluors is highly transparent; at 440 nm it changes 10.4 meters to 8.0 meters (99.0 %T to 98.8 %T) in three years. In the upper right (c), a solution containing 5 g/l Gd is stable. After almost two years the absorbance change at 430 nm is about 0.0015 a.u. (30 m component). At 400 nm the change is only about 0.0043 (10 m component); there is no solid formation. The full scintillator mixture prepared by Buck [19], in the lower right (d), has an absorbance change at 430nm of 0.0022 (20 meter component) in just less than two years.

3.2.3 Radiopurity. One hundred kilograms of Gd-BDK were vaporized and re-crystalized into molecular crystals in order to achieve a high radiopurity. Table 1 shows the radiopurity results following these sublimations as measured in the MPIK germanium GeMPI detectors (GeMPI I, II, III) at the Gran Sasso Underground Laboratory. All numbers are within the radiopurity specifications.

Table 1. Radiopurity measurements of the Gd-BDK prepared for the neutrino targets of the Near and Far Detectors. Activity per detector in column three is calculated from column two.

Nuclide	Activity (mBq/kg)	Activity (mBq/detector)
Ra-226 (Rn-222)	<0.57	<26
U-235	<0.52	<24
Ra-228	<0.48	<22
Th-228 (2614 keV)	<0.54 (2 > detection limit)	<26
K-40	13.8 (7 > detection limit)	640

Eu-152	<1.0 (4 > detection limit)	<48

3.3 The TMHA carboxylic scintillator system

The TMHA complex of Gd has been studied over the course of five years (since 2003). Samples produced at 1 g/l for long term monitoring showed extremely good characteristics having good absorbance (>25 m, 440nm) and high light yield (64 % anthracene measured versus BC505). Samples up to 50 g/l Gd were readily produced; concentrated solutions at 10 g/l were made for for prototype tests. The procedure employs a pH controlled solvent extraction and remains pH stabilized. From these solutions, 100 liters of 1 g/l Gd 100 liters were produced for prototype testing in 2005. From various studies, it was found that initial degradation rates were approximately proportional to the Gd concentration at the start. Consequently a lifetime prediction of 15 meters to 5 meters absorbance was predicted over 5 years. A drawback of the TMHA system is the lability of the complexing anions suggesting its lower molecular stability relative to the BDK system, the absence of a purification mode from radiopurities via gaseous recrystallization and its dehydration characteristics. Consequently a long series of measurements (shown here for a sample observed for 4 years) indicates notable crystal formation during the 1000 and 1300 day interval.

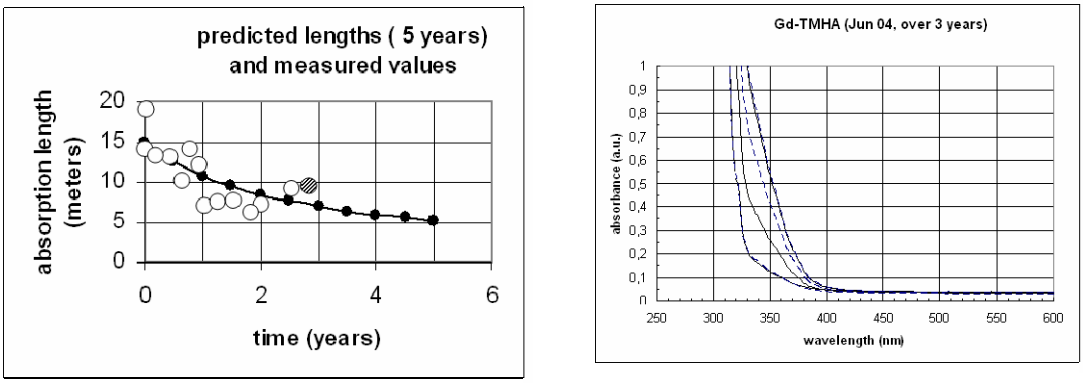
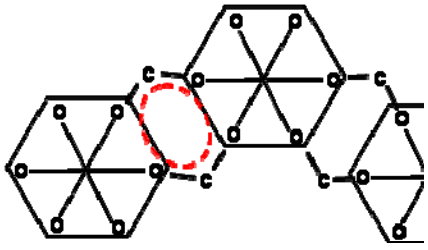


Figure 6. CBX system. After *ca*.1000 days (left) the system exhibits an instability attributed to changes in the molecular structure. Between 1000 and 1300 days the system was adjusted with a hydrogen bonding agent to see if it could be recovered. The system continues under study for 1500 days. Changes in absorbance (right) over the 1000 days are seen to occur (plotted at approx. 6 month intervals) and the absorbance slowly moves into the optical regime from "behind" the location of the UV absorption of the base fluor (here PXE). Nanogram to microgram crystals form after about 1000 days and are attributed to bridging. Afterwards, the UV spectrum is static.

Figure 7. Structural bridging. An explanation for the behavior of Gd-TMHA (CBX) degradation is the formation of bridging carboxylic acid anions which cross between Gd ions. Such bridging can occur (*ad infinitum*) due to the coordination sphere chemistry of Gd. Such bridging can lead to changes in the UV-vis properties due to electron delocalization. Perhaps (dotted line) aromatic effects can also enter.



4.0 Summary

The Double Chooz Project is in the assembly phase and scheduled to come on-line in 2009. In 1.5 y, the Far Detector should reach a sensitivity level of $\sin^2 2\theta_{13} = 0.057$, $\theta_{13} = 7.0$ deg. The Near Detector then comes on-line; after an additional 3.5 y the sensitivity level is $\sin^2 2\theta_{13} = 0.030$,

$\theta_{13} = 5.0$ deg. A "3 sigma effect" discovery potential [20] is 5.8 deg in 3 y. A very stable (>5 y) scintillator is extremely important to the measurement of θ_{13}.

In this paper, we present the first detailed information about the target and gamma catcher scintillator for the Double Chooz project. This is the first such full detector system based on beta-diketone chemistry. For this reason we have sought to summarize a number of references related to details of the work and to record references to some of the background work as well.

The neutrino target and gamma catcher in Double Chooz have the light yield and densities matched. A light yield model has been developed for multi-component liquids that allows us to understand the energy transfer properties and as a result we are able to tune the pulse shapes. This feature is afforded by choice of the liquid components which allow us an extra degree of freedom.

Extensive work has been made since 2000 with the CBX system based on TMHA whence we developed the pH stabilized carboxylate system. Detailed studies now extend for 4 years. The systems have known degradations which are modeled by us. After about 1000 days in existing samples, we see gradual changes in the UV and the onset of nanogram-scale crystal formation in conformance with what is predicted from concentration studies. Continued and ongoing investigation search lead us to conclude, for the purposes of the TMHA system, that bridging is playing a role such that the system is not usable by 3.5 years.

We thus concluded that the BDK systems are best capable of putting the right nucleus in the right place at the right time for Double Chooz. Future investigations in neutrino physics, for example [21], are focused on the use of beta-diketones and proceed in this direction. Parallel work with Nd, of interest to double-beta decay, proceeds as well using both TMHA and BDK systems.

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