Studies of the impact of beta contributions on Cherenkov light emission by spent nuclear fuel

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Abstract

The Digital Cherenkov Viewing Device (DCVD) is one of the instruments used by safeguards inspectors to verify spent nuclear fuel in wet storage. The DCVD can be used for partial defect verification, where the inspectors verify that 50% or more of an assembly has not been diverted. The methodology is based on comparing the measured Cherenkov light intensity with a predicted intensity, calculated with operator information.

Recently, IAEA inspectors have encountered fuel assemblies for which systematic deviations between predictions and measurements could be observed, indicating that the prediction model did not take into account all sources of Cherenkov light production. One contribution to the Cherenkov light intensity that is frequently omitted is the contribution from beta decays, where energetic electrons exit the fuel material and enter the water with sufficient energy to directly produce Cherenkov light. The objective with this work was hence to study beta contributions and evaluate whether that could be the cause of discrepancy between predictions and experimental data.

By simulating the beta contribution for fuel assemblies where the discrepancy was experimentally observed, it was determined that beta decays were the cause. The fuel assemblies had fuel rods with relatively small radii, thin cladding, a short cooling time and an irradiation history that resulted in a relatively large beta contribution for assemblies that had a comparatively low burnup. Therefore, the beta contribution was significant, and caused 10-40% of the total Cherenkov light intensity. By including the beta contributions in the predictions, the RMSE of the deviation between prediction and measurement could be reduced from 20.7% to 11.6% for the available measurement data. The results highlight that the beta contribution can be significant and should be taken into account for accurate predictions.

Keywords: Nuclear safeguards, Cherenkov light, DCVD, beta decay, spent fuel verification

1. Introduction

Following international safeguards agreements, international inspectors from the International Atomic Energy Agency (IAEA) or EURATOM are tasked with verifying that nuclear material is not diverted from peaceful use. One form of nuclear material that must be verified is spent nuclear fuel. To help the inspectors to independently verify nuclear fuel assemblies, a multitude of instruments have been developed [1]. One of the deployed instruments is the Digital Cherenkov Viewing Device (DCVD), which measures the Cherenkov light emission by spent nuclear fuel assemblies in wet storage [2]. The DCVD is capable of performing gross defect verification, where the presence and qualitative characteristics of the Cherenkov light emissions are used to verify that an object is a spent nuclear fuel assembly, and not a non-radioactive dummy object. The DCVD is also used to perform partial defect detection, verifying that part of a fuel assembly has not been diverted. Two methodologies are in use for partial defect detection: one that uses image analysis to determine if rods in visible positions have been removed, and one that quantitatively measures the Cherenkov light intensity to verify that it is consistent with the expected intensity, based on operator declarations of the fuel [3].

1.1 Verification of spent nuclear fuel with the DCVD

For the quantitative Cherenkov light intensity verification, the Cherenkov light intensity is predicted based on operator-declared values of burnup (BU), initial enrichment (IE) and cooling time (CT), or the so-called BIC parameters. In general, these three parameters are sufficient to characterize the fuel assembly, though the irradiation history also has some influence in the abundance of fission products [4]. Once the inspector has completed the predictions of the emitted Cherenkov light intensity, the spent fuel assemblies are measured using the DCVD, which is typically mounted on the railing of a fuel-handling machine above the fuel pond.

In the analysis, measurement data is grouped according to fuel type and measurement campaign. Thus, the measurements of the fuel assemblies in each group can be directly compared with each other, since they have the same design and the measurements were taken under the same conditions. For each group, a multiplicative constant is

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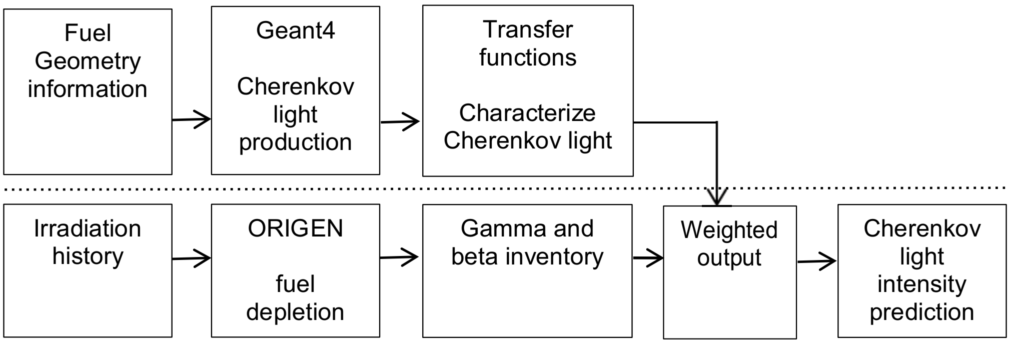

Figure 1: Schematic of the Cherenkov intensity prediction method. The top row contains the Monte-Carlo simulations to parameterize the Cherenkov light productions as a function of the radiation type and energy. The bottom row contains the calculations performed to obtain a prediction.

found to relate the predictions to the measurements using a least-square fitting. This multiplicative constant will compensate for effects that are identical for all assemblies in that group. Such effects include scattering and absorption of Cherenkov light in fuel assembly structures and the surrounding water, the optical efficiency of the detector system, the conversion of light to a measurable charge in the DCVD Charge-Coupled Device (CCD) chip, and the conversion of this charge to an image pixel value. Hence, after this calibration, the measured and predicted Cherenkov light intensities can directly be compared.

1.2 Motivation for this work

Recently, IAEA inspectors have encountered fuel assemblies where a systematic difference between predictions and measurements could be seen. The differences have been observed mainly for fuel assemblies with a CT of 2-5 years, and the magnitude of the difference was found to depend on burnup and fuel design. These systematic deviations indicate that the predictions do not accurately model the Cherenkov light production. The findings motivated this work, where the aim is to investigate whether the discrepancies are caused by the omission of direct beta-decay, and to assess/determine whether the predictions can be improved by including this contribution.

2. Cherenkov light intensity predictions

To predict the Cherenkov light intensity of an assembly, ORIGEN [5] is used to simulate the fuel depletion, either using the operator-provided irradiation history, if available, or using a default irradiation scheme otherwise. ORIGEN will then calculate the gamma-ray emission spectrum of the assembly, which is combined with a transfer function to obtain an estimate of the total Cherenkov light production of the assembly, and the abundance of beta-decaying isotopes. The methodology to predict the Cherenkov light intensity is summarized in Figure 1.

In the case of spent fuel stored in water, Cherenkov light is predominantly caused by gamma decays of fission products in the fuel [6]. Thus, to predict the Cherenkov light intensity, a model of both the gamma radiation emissions and the subsequent Cherenkov light production is needed. The Cherenkov light prediction model used in the latest version of the DCVD software is based on [7], and this method has been implemented and extended in [8]. As will be detailed later, the prediction model in the DCVD software does not take into account beta decays where the electron passes through the fuel and cladding, and enters the water with sufficient energy to directly produce Cherenkov light. This contribution will be referred to as the "direct beta contribution" in this publication, as opposed to the "indirect beta contribution" which is used to describe beta particles that produce bremsstrahlung in the fuel, which in turn produce high-speed electrons in the water through Compton scattering or photoelectric absorption.

In order to estimate the Cherenkov light intensity that is produced per gamma-quanta of a certain energy, Monte-Carlo simulations that consider the relevant fuel geometry are made. These results are used to create a transfer function that relates the gamma emission energy to Cherenkov light production, as described in [9]. This transfer function is then applied to the gamma emission spectrum of the fuel assembly (as calculated by ORIGEN), to predict the Cherenkov light production by gamma emissions.

In principle, beta-decays can be handled in the same way, but the beta emission spectrum is not calculated by ORIGEN and must be obtained in some other way. Reference [9] suggests simulating beta decays from selected isotopes, to assess their respective direct beta contributions. The results can be used to calculate another transfer function that relates either the isotope activity or isotope mass to a Cherenkov light production, which can then be added to the Cherenkov light prediction due to gamma rays. Note that ORIGEN can calculate the bremsstrahlung emissions due to beta decays being stopped in the fuel, which are included in the gamma emission spectrum. In the DCVD Cherenkov light predictions, the bremsstrahlung contribution is treated as gamma emissions.

Earlier studies showed that the direct beta contribution could approach 5% of the total intensity, in the case of fuel assemblies with long CT, thin rods and thin cladding [9], but that it would typically be closer to 1-2%. However, the experimental data available used to validate those predictions covered fuel assemblies with a CT of at least 5 years, and thus fuel assemblies with shorter CT were not evaluated. Furthermore, the fuel assemblies used to validate the model primarily consisted of fully burnt assemblies; hence, the performance of the prediction methods could not be thoroughly validated at lower BU values. For the fuel assemblies with a CT of at least five years used to validate the model, decays by Sr90/Y90 are the only significant source of beta decays. Due to its modest contribution, the direct beta component was not included in the prediction model available in the DCVD software. For many of the fuel assemblies where the IAEA inspectors have observed a discrepancy between predictions and measurements, the CTs are however shorter than for the measurements used to validate the model. Hence, additional beta-decaying isotopes may still be present, which could potentially contribute more significantly compared to Sr90/Y90.

2.1 Information about the experimental data

To investigate the cause of the systematic difference between predictions and measurements, a set of roughly 300 assemblies with operator provided irradiation history have been measured. The set of fuel assemblies were selected to have a short CT and varying BU, since such fuels have shown the greatest discrepancy between prediction and measurement. Based on the fuel type and irradiation history of the assemblies, a few general remarks can be made:

- The fuel assemblies had a BU in the range of 20-60 MWd/kgU, and CT of 2-5 years. The shortest cooled fuels had the largest range of BU values, while the longer-cooled fuels had typically reached their discharge BU.
- Most of the measured assemblies had smaller rod radii and thinner cladding compared to the fuel assembly measurements used to verify the prediction model [9].
- In general, the fuel assemblies experienced the highest power level during their first few cycles, followed by a varying number of low power cycles, and finally one or more medium power cycles before reaching the discharge BU.

These fuel assemblies thus have fuel parameters such as BU and CT that differ notably from the experimental data used to validate the prediction model [9]. The smaller fuel rod radii and thinner claddings mean that the direct beta contribution could be larger than in the previously studied cases, and the shorter cooling times means that additional beta-decaying isotopes beyond Sr90/Y90 may need to be taken into account.

The irradiation history of the assemblies in this data set also differed from previously considered irradiation histories where even, high power cycles were assumed for all but the last fuel cycle, which was assumed to be a low-power cycle. The differing power history will affect the abundance of short-lived isotopes present at the short cooling times of 2-5 years in this data set.

3. Methodology

3.1 Depletion calculations

The experimental data consist of DCVD measurements of roughly 300 assemblies of the same type. The assemblies have a wide range of BU and CT, and were selected since the systematic deviations in the predictions were pronounced for this group of assemblies. For each fuel assembly in the data, ORIGEN was run to determine the gamma emission spectrum and beta-decaying isotope contents. The depletion calculations accounted for the the operator provided irradiation history, simulating the correct per-cycle average burnup and the length of all cooling times. The ORIGEN fuel libraries “ge10x10-8” and “atrium10-9” have a similar rod configuration to the measured assemblies, and the Atrium library was chosen since it matched the number of short rods. The ORIGEN gamma spectrum also includes bremsstrahlung.

3.2 Simulating Cherenkov light production

To model the Cherenkov light production in a fuel assembly geometry, a Geant4 [10] based simulation toolkit has previously been created [7]. The results of these simulations were used to set up the transfer functions, relating gamma and beta particle emissions to Cherenkov light production. This code simulates the emissions of gamma and beta particles in the fuel material, their interactions in the fuel, cladding and water, and the production of Cherenkov light in the assembly. This code has been used to study the Cherenkov light production by both gamma and beta decays [6], including both direct beta contribution as well as bremsstrahlung, to identify their respective contributions.

This code package was used to simulate the Cherenkov light production for the fuel assemblies in the experimental data set. The modelled fuel assembly geometry is summarized in Table 1 and was selected to be representative of several different modern BWR fuels in [11]. Although the exact fuel dimensions were not revealed by the operator, the modelled dimensions match the Atrium fuel simulated by ORIGEN. The selected dimensions are also representative of modern BWR designs, matching the experimental data. However, the lack of detailed knowledge does introduce uncertainties in the simulations. As an example, for

Fuel density [g/cm3]	Fuel radius [mm]	Cladding inner radius [mm]	Cladding thickness [mm]	Pitch [mm]	Rod configuration
10.5	4.34	4.42	0.61	13.4	10x10

Table 1: Fuel dimensions used in the simulations.

the modelled cladding thickness, if the thickness were to be changed by 0.1 mm, the direct Cherenkov light production by Y90 may change by a factor of two [6], since the beta electrons are strongly attenuated by the cladding material. Since exact fuel dimensions is typically not available to an inspector, the prediction model should be general enough to be applicable even in the lack of such information, although uncertainties will be introduced by such assumptions when they do not match the assemblies to be measured.

Monoenergetic gamma emissions from the fuel material were simulated for various energies in the range 250 keV to 4 MeV, to determine the Cherenkov light production in the water as a function gamma initial energy. To simplify the simulations, fresh fuel material was assumed, since the difference in gamma attenuation between fresh and spent fuel is less than 3% for the photon energies that can produce Cherenkov light and the BU encountered in this work [12]. The initial gamma particles were distributed uniformly in the radial direction of each rod. Cherenkov photons forming an angle less than 3 degrees to the vertical axis were tallied in the simulations, since [13] notes that the vertical and total Cherenkov light components may behave slightly differently. Due to the measurement setup, the measured intensity will more closely follows the vertical intensity.

To determine the beta contribution, a separate set of simulations were run for each isotope identified to be of relevance for direct Cherenkov light contribution, taking into account the beta energy spectrum of the decay. The beta decay spectra were taken from [14]. Bremsstrahlung was disabled in the simulations, to account for only the direct beta contribution, and because bremsstrahlung is treated as a gamma emission in the prediction model. Uniformly distributed starting locations were used also for the beta simulations, though as discussed in section 3.4, the real distribution is more complicated but is unlikely to be known to an inspector performing a measurement.

3.3 Intensity predictions

Once the ORIGEN depletion calculations were done and the transfer functions were set up, the clip software package [8] was used to extract the gamma emission spectrum and the abundance of the beta-decaying isotopes for each simulated assembly. These were combined with the simulated Cherenkov light production as a function of gamma-ray energy, and the Cherenkov light production per decay for the beta decaying isotopes, to assess the direct beta Cherenkov light production. The direct beta production was then added to the gamma production, to obtain a total prediction that properly includes both components.

Since the direct beta contribution was handled separately, this allowed for estimating the magnitude of the direct beta contribution relative to the gamma contribution. It also enabled comparisons between measurements and predictions, where predictions either included or excluded the direct beta contribution.

3.4 Limitations of the source distribution assumption

The radiation source distribution in a rod is more complex than a uniform distribution, which does have an impact on the Cherenkov light production, as noted in [6]. Some elements such as Caesium migrate due to heat gradients, and fission product concentrations are higher on the pellet rim due to the high-burnup structure. Especially for beta decays, where only decays on the pellet rim can produce Cherenkov light, effects such as the high-burnup structure at the rim can noticeably enhance the beta contribution [6]. The situation is further complicated by the fact that the fissile material in the pellet rim may be depleted more quickly than the bulk fuel rod material to obtain the high-burnup structure at low rod average burnup. However, if the rim is depleted early in the fuel lifetime, this may relatively suppress the beta contribution at higher rod burnups, when the rim has been depleted for some time. In addition, cracking of the fuel pellets may result in that regions some

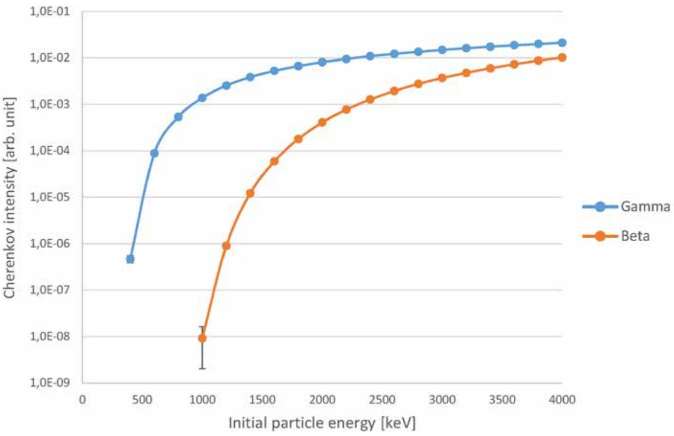

Figure 2: The average Cherenkov light production per gamma and beta particle emission. For the beta emissions, the contribution contains only the Cherenkov light produced directly by the beta decay and neglects any bremsstrahlung, which in turn can result in Cherenkov light production. The error bars refer to the Monte-Carlo statistical uncertainties in the simulations.

millimetres into the pellet have a free path to the cladding, which could allow beta decays from more interior locations of the pellet to contribute more to the Cherenkov light production. The development of a detailed source distribution model that is generally applicable based on the limited information available to an inspector is outside the scope of this work, hence the simplifying assumption of fresh fuel and uniform source distribution is made.

4. Results

4.1 Cherenkov light production by gamma and beta decays

The Cherenkov light production as a function of initial particle energy is shown in Figure 2, for both initial gamma and beta particles. Similar to the results of [6], beta particles require higher energy than gamma particles to produce comparable amounts of Cherenkov light. A kinetic energy of around 250 keV is required for an electron to produce Cherenkov light in water, though as seen in Figure 2, an initial kinetic energy of around 1 MeV is required for the electron to be able to penetrate the fuel and cladding to directly produce Cherenkov light.

4.2 Beta decaying isotopes of interest

Based on the results in Figure 2 and previous considerations, beta-decaying isotopes fulfilling the following characteristics may contribute to the Cherenkov light production at the cooling times seen in the experimental data:

- The fission yield should be high enough that the fission product isotope is abundant in spent nuclear fuel. Isotopes with a cumulative fission yield above 0.1% were included in this work.
- The half-life of the isotope should be short-lived to be active enough to contribute, but sufficiently long-lived to be seen in the experimental data. For this work, isotopes with a half-life between 1 month and 100 years were investigated
- The maximum beta particle energy should be at least 1 MeV according to Figure 2 to contribute at all to the measurable Cherenkov light intensity. The beta emission could come from either the decaying nucleus or a short-lived daughter.

Based on these criteria, three isotopes and their daughters were identified to be of relevance, as summarized in Table 2. For the three isotopes, the parent nuclei are sufficiently abundant and long-lived to be of interest, and the short-lived daughter emits high-energy beta particles. The simulated Cherenkov light production by these three isotopes is summarized in Table 2. The beta energy spectra used in the simulations were taken from [14].

4.3 Direct beta contribution in the experimental data

Using the Cherenkov light intensity prediction for gamma and beta decays, the fraction of Cherenkov light produced directly by beta decays to the total intensities were calculated. The results are shown in Figure 3. As can be seen in Figure 3, the direct beta contribution to the total Cherenkov light intensity is significant, above 10% in all cases for this data set and up to 40% for the low-BU assemblies. In part, the thinner cladding and smaller rod radii mean that beta decays are more likely to directly produce Cherenkov light compared to previously studied fuel types, which is one reason why their contribution is so significant here. However, part of the explanation is also the irradiation history of these specific fuel assemblies. In Figure 3, the fuel assemblies have been further subdivided into groups according to CT, with group 1 having the shortest CT and group 4 the longest. For group 1, a wide range of BU is present, and the effect of the irradiation history results in a wider spread. The irradiation history for all assemblies typically consisted of initially a few high-power cycles, until a burnup of 20-30 MWd/kgU was achieved. The high-power cycles were followed by several low-power cycles, typically until a burnup of 40-45 MWd/kgU was achieved. Finally, the assembly experienced some medium-power cycles before the assembly reached its discharge BU, of typically 55-60 MWd/kgU. The final irradiation cycle for group 1 fuels may thus be either a low, medium or high-power cycle. For group 2-4, the fuel assemblies had typically reached their discharge BU and experienced a final, medium-power cycle, resulting in a much less pronounced spread in the beta fraction within each group.

The relative fraction of the direct beta contribution by each of the three identified isotopes is shown in Table 3, for three selected groups of fuel assembly parameters. The build-up of Ce144 peaks at a BU of around 20-30 MWd/kgU in the ORIGEN simulations for this data set. Part of the reason for this concentration peak is that these fuel assemblies had just experienced high-power cycles; hence, the production of Ce144 is high. Another cause is the

Isotope	Sr90/Y90	Ru106/Rh106	Ce144/Pr144
Parent half-life	28.9 years	372 days	285 days
Daughter maximum beta energy	2.24 MeV	3.53 MeV	2.99 MeV
Cherenkov photons per decay	2.71E-5	3.87E-4	1.56E-4

Table 2: The three identified isotopes and daughters that contribute to the total Cherenkov light intensity. The Cherenkov light production takes into account the beta emission spectrum of the daughter, and neglects bremsstrahlung.

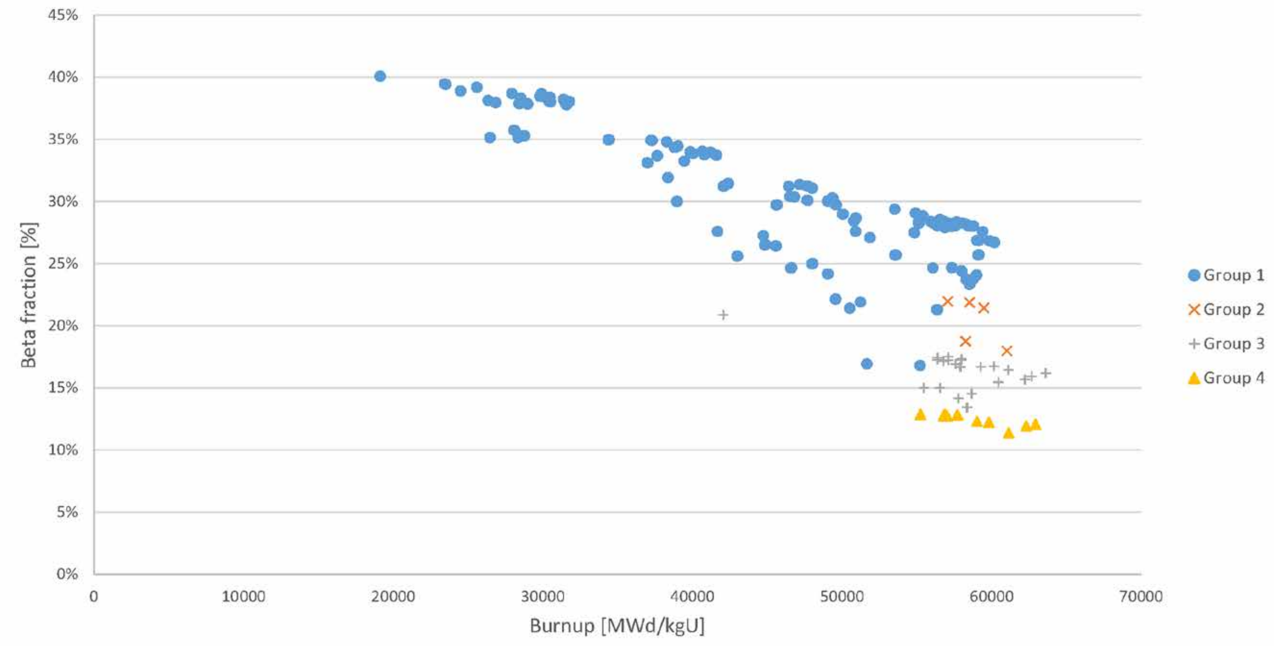

Figure 3: The fraction of total Cherenkov light production caused by direct beta decays, as a function of the fuel assembly BU. The remainder is caused by either gamma decays or bremsstrahlung due to beta decays. The fuels are grouped according to their CT, with group 1 having the shortest CT, and the CT increases with group number. The vertical spread in group 1 is predominantly caused by the differing irradiation history within the group.

cumulative fission yield of Ce144, which is higher for fission in U235 than in Pu239. Plutonium fissions contribute more to the energy release at higher BU than at lower BU since it builds up with BU, and consequently the Ce144 production is reduced with BU. For Ru106 the cumulative fission yield is significantly higher for fission of Pu239 than U235, hence its importance tends to increase with BU for the assemblies analyzed, although it was occasionally seen to decrease slightly during the low-power cycles. For fully burned assemblies, the importance of Sr90/Y90 increases with CT as it is comparatively long-lived, with the importance of Ce144/Pr144 decreases faster than Ru106/Rh106 due to its slightly shorter half-life.

4.4 Evaluation of prediction performance

A comparison is made between the measurements and the predictions, for predictions with and without the direct beta contribution, shown in Figure 4. Note that one fit is done to relate predictions without direct beta contribution to the measurements, and a second fit is done to relate the predictions including the direct beta contribution to the measurements. After the fitting, the relative deviations between predictions and measurements in the data sets were calculated and averaged over, to determine the RMSE values of the deviations. As seen earlier in Figure 3, the direct beta contribution varies significantly in this data set. That can be seen as a large spread in the difference between the prediction and measurement when the beta decays are not accounted for, as seen in Figure 4. In total, the predictions without the beta contribution had an RMSE between prediction and measurement of 20.7%. This was lowered to 11.6% when the direct beta contribution was taken into account; a value comparable to other measurement campaigns with long-cooled fuels where beta decay contribution was negligible, such as in [13]. Thus, when the direct beta contribution is included in the predictions, the predicted and measured values are a much better match after the new fit, judging by the RMSE values. This conclusion is valid when assessing the entire group of fuel assemblies in Figure 4, though note that individual predictions for a single fuel assembly may differ (i.e., improve or worsen) as the direct beta contribution is included.

In total, these results show that the direct beta contribution to the Cherenkov light intensity can be significant and should not be neglected for accurate prediction. The

Fuel	Sr90/Y90	Ru106/Rh106	Ce144/Pr144
CT 2 years, BU 20-30 MWd/kgU	5-10%	50-60%	30-40%
CT 2 years, BU 50-60 MWd/kgU	5-10%	60-70%	20-30%
CT 5 years, BU 50-60 MWd/kgU	30-35%	55-60%	10-15%

Table 3: The fraction of the direct beta contribution by each of the three identified isotopes, for three groups of fuel parameters in the experimental data. These groups represent combinations of high and low BU/CT that occur in the data set.

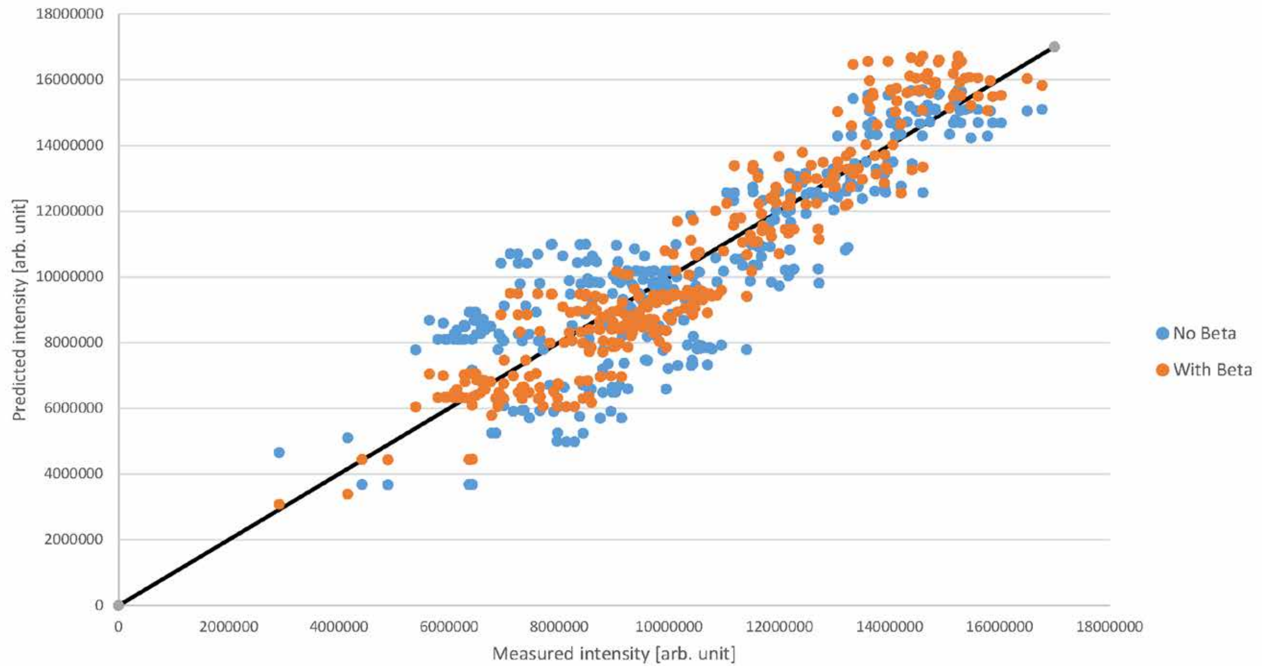

Figure 4: Comparison of the predictions of the two models, with and without the direct beta contribution, with the measured Cherenkov light intensities. The predictions of each model were scaled to match the measured intensity, as detailed in section 1.1. The black line is a guide for the eye, noting where the predictions and measurements match.

results also highlight that the abundance of several of the short-lived beta-decaying isotopes depends on the irradiation scheme used and should be accurately modelled for best results.

5. Conclusion and Discussion

Previous studies have identified that Cherenkov light is produced by beta particles that pass through the fuel material and cladding and directly produce Cherenkov light in the water. However, based on the available experimental data, it has not been clear when this becomes a significant contribution. In experimental measurements of fuel assemblies with cooling times of 2-5 years and burnups from 20-60 MWd/kgU, systematic effects were seen in the comparison between predictions and measurements, which largely can be explained by the direct beta contribution.

The model used to predict Cherenkov light intensities based on the fuel assembly gamma spectrum has previously been extended to also include beta decays explicitly. Since relatively few beta-decaying isotopes that significantly contribute to the Cherenkov light intensity are present, simulations were made for each isotope. This gives information about the intensity of the Cherenkov light produced per decay from each isotope. In turn, this can be combined with the isotope mass abundance in the spent fuel to obtain a prediction of the isotope-wise Cherenkov light production, which can be added to the total prediction. For the experimental data, adding the Cherenkov light contribution caused by these isotopes significantly reduced the systematic effects seen as a function of burnup and cooling time. The deviation between prediction and measurement was reduced from an RMSE of 20.7% to 11.6%, which is comparable to measurement campaigns with long-cooled fuels where beta decays had a negligible impact on the predictions. In the simulation work performed here, it was found that the relatively thin claddings and the irradiation history resulted in a much larger direct beta contribution than previous results have indicated, with the direct beta contribution varied between 10% and 40% of the total Cherenkov intensity. This shows that there exist fuel assembly populations where the direct beta contribution cannot be neglected in the predictions.

Based on these results, we recommend that the prediction methodology in the DCVD software should be updated to include the direct beta-contribution thereby making accurate predictions available to safeguards inspectors. The methodology itself has already been developed and is ready to be incorporated in the next DCVD software version. Implementing this will require simulations of the Cherenkov light production from identified beta-decaying isotopes for a number of fuel geometries. Should the DCVD be used for even more short-cooled fuel assemblies than those studied in this work, additional isotopes may need to be added to the model.

While this work have obtained improved predictions using a rather simple direct beta modelling, accurately modelling the beta contribution is more challenging, but could potentially further improve the results. Fuel rods feature a high-burnup structure on the pellet rim, as fully moderated neutrons do not penetrate deep in the fuel material. In addition, the high burnup structure may results in pellet cracking, allowing beta decays from slightly deeper within the pellet to have a free path to the cladding. The direct beta contribution was previously found to be sensitive to the cladding thickness, and effects such as cladding creep, oxygen and hydrogen pickup will likely influence the attenuation of beta particles in the cladding. Hence, for accurate modelling of the direct beta contribution, such effects should be included in the model, and especially how they change with time or burnup. Such models however need to be general enough that they can be applied based on the limited amount of data available to an inspector, in order to be useful in the field.

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