Dear Reviewer 2,

thank you for reading our manuscript and providing valuable feedback. We addressed your comments and suggestions point by point as follows:

Comment / Suggestion:

The measurement is interesting, but a lot of laboratory work is still necessary to publish work.

A reasonable data analysis is missing, calibrations are missing and the paper conclusions are not supported by the sole measurements. Therefore it is not clear to me what the innovative results of this paper/measurement are in this status.

Reply:

We enhanced the manuscript by having improved our data analysis and by having made the description of our experiment, our results and what we take to be the calibration of our experiment much clearer. We further enhanced our paper by showing more clearly that the data presented in the manuscript does support the conclusions.

The innovate results are

- We show that Am-241 dissolved in an aqueous nitric solution produces light which we identify as radioluminescence
- We show that the light emanates evenly from the liquid
- We show that at the surface of the liquid is an increased production in radioluminescence, which we hypothesize can be attributed to air radioluminescence
- We present a fully resolved spectrum of the ‘liquid’ radioluminescence from 280 to 550 nm
- We investigate the intensity of the radioluminescence as a function of the Am-241 concentration and the acidity of the nitric acid solvent
- We discuss the possible mechanism that produce the radioluminescence

Comment / Suggestion:

As first I have a comment on the data handling. To remove the hot pixels due to 59.6KeV of Am-241 the image are "median blurred with a sliding window width of 7". This results in a very poor definition images that affects the subsequent analysis capability. However there is a very simple way to recognize and remove these hot pixel without any pixel blurring, since you have 30 different images. I suggest to do the pixelwise average, averaging pixel-by-pixel removing the largest, that is averaging the lower 29 values of the same pixel and always removing the value
with the largest measurement (that could be the X-ray). This unblurred-cleaned image would improve a lot the quality of the result.

Reply:

We redid our data post-processing by taking the pixel-wise median over all 30 images. This removed the unnecessary blurring and the hot pixels at the same time. We changed Fig. 2 to reflect this improved approach. We changed the caption to

> Demonstration of the post-processing using 30 images with exposure times of 60 s each. (a) A single capture, raw data. The cuvette and the liquid are clearly visible, hot pixels appear as red dots. (b) The pixel-wise median over a total of 30 images. The z-scale shows raw pixel values.

We changed the description of the post-processing in the text to

> All image material has been post-processed by taking the pixel-wise median of the 30 consecutive images. The so-constructed median image is largely free of hot pixels all the while leaving the features and resolution of the image unaffected. The procedure is illustrated in Fig. 2 with data from the experiments serving as example. For image processing we used python 3.6.3 as programming language and made use of the libraries OpenCV 3.3.1, matplotlib 2.1.0 and NumPy 1.13.3.

In addition to Fig. 2, Figs. 4, 5 and 6 and the quality of the content they present have changed as a direct result of improving the image processing.

Comment / Suggestion:

Here I summarize the statement/result that are in the paper and that are not fully convincing me:

1) “The light production in the liquid is evenly distributed throughout the sample volume having a slight increase on the surface of the liquid.” This statement in the result of the discussion of fig. 5.b.

-There a luminosity broad peak is labelled as “liquid bottom”,

-another peak is labelled “plume”

-and 3 small peaks are labelled ad “contamination”.

The peak labelled as “liquid bottom” is ascribed to be the “result of reflection from the cuvette holder”. This could be reasonable, since the optical effect of the top of the cuvette border is also clearly visible as two side spots also in the 2D color plot (left) of fig 5.b (or also better in unblurred fig 2.a) and this structure is producing the first peak of fig 5.b (right) (distance 2mm from top) that is wrongly labelled as “contamination”.

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In the improved image material in Fig. 5b) it will become clear that the peak which used to be labeled “liquid bottom” is created as a result of reflections from the holder. We amended Fig. 4 by a new sub-figure (e) that overlays the radioluminescence image of the cuvette with the background image. In the overlay image the influence of the holder can directly be seen. We further amended the text by

> It can also be seen there is an area of slightly increased pixel intensities at the lower end of the cuvette. The camera looks with a slightly tilted angle at the cuvette, making a part of the cuvette which is surrounded by the holder visible to the camera. It is those parts of the image that show increased pixel intensities. In the overlay image in Fig. 4 (e) this becomes even clearer. Thus it is very likely that the increased intensity in the radioluminescence image in this particular area is not created by an increased radioluminescence production but rather by reflection of radioluminescence photons from the sample holder towards the camera.

We also enhanced the discussion of the peaks that were previously labeled ‘contamination’. We amended our discussion by

> The local maxima to the left in Fig. 5 (b) correspond to the increased radioluminescence observed at the top end of the cuvette. This is very likely radioluminescence from dried liquid. The same cuvette was used to hold all liquid samples. Changing the sample made it necessary to empty and refill the cuvette. During refilling contact of the liquid with the walls was difficult to avoid, thus contaminating it. Even careful cleaning rarely removed all contamination, and it were the edges of the cuvette that proved most difficult to clean. It is this contamination that is seen in the radioluminescence image. Though the radioluminescence from contamination of the cuvette walls is somewhat of a nuisance for this particular work, it shows once again that radioluminescence imaging is a very efficient method to find alpha contamination in difficult to access areas.

We also changed the labels in Fig. 5 (b) to make our interpretation of the data clearer.

Comment / Suggestion:

The “plume” peak is instead ascribed to a physical effect: “The radioluminescence emanating from the cuvette in Fig. 4 (d) appears to be more intense at the surface of the liquid. This might be due to alpha particles leaving the liquid into the air and depositing their energy there, thus leading to more efficient radioluminescence production in air” This would support the other conclusion of the paper that alpha luminescence in air is more efficient that in liquid. However this interpretation of the plume is not convincing me, since, similarly to the first peak of “contamination” it could be that the “plume” is an optical effect of the liquid meniscus, that is clearly visible in fig. 4.b.

Reply:

The word “plume” was poorly chosen. The radioluminescence image only reveals that the liquid-
air boundary shows increased intensity, which we hypothesize to be caused by actual increased levels of radioluminescence (unlike in the case of the sample holder reflections). But it is certainly not a plume. We clarified our discussions of the radioluminescence image by changing the parts referring to the liquid-air boundary to

> The radioluminescence emanating from the cuvette in Fig. 4 (d) appears to be more intense at the surface of the liquid. We hypothesize this to be an effect of some of the alpha particles being created close to the liquid-air boundary not being fully stopped in the water [26]. They could then leave the liquid into the air, where they would create air radioluminescence. This hypothesis would further be supported by the observation that the shape of the area of increased intensity is very similar to the shape of the liquid-air boundary. The camera is slightly tiled towards the cuvette, which makes it possible to look on top of the boundary the liquid is having with the air. It is this area that shows the increased radioluminescence. The overlay image Fig. 4 (e) makes this easy to see.

Comment / Suggestion:

To solve this problem and to give solidity to the conclusions, I suggest to compare this measurement with another control one using a some known luminescent liquid (with known spectra).

Reply:

No knowledge of the spectrum of the radioluminescence is required to explain its spatial distribution.

We enhanced the manuscript by adding the overlay Fig. 4 (e). There it can be clearly seen that the increased pixel intensity at the bottom of the cuvette is due to reflections from the sample holder. The same image shows that the increased pixel intensity at the liquid-air boundary can not be explained by reflections. We rather explain it with an actual increase in radioluminescence production, which is caused by alpha particles leaving the liquid and producing air radioluminescence. This is a known phenomenon. We clarified this in the manuscript and added references to the appropriate works.

Comment / Suggestion:

Moreover I suggest to not use blurring to improve the geometry information in these figures (as suggested above) and I suggest also to superimpose to fig 5 a line-drawing of the cuvette border and of the curved liquid meniscus and to give the same information to the left plots of fig.5 adding more vertical lines (there only liquid surface is now shown).

Reply:

All image post processing has been redone using pixel-wise median as noise-reduction technique. In the new data analysis none of the images has been blurred.
We chose not to add lines to images of the cross sections. It made the images very messy and hard to comprehend. We instead added an overlay image of the radioluminescence from the cuvette with the background image in Fig. 4 (e). It provides the spatial information to the radioluminescence image as suggested. We also added information about the liquid surface to both the liquid sample images in Fig. 4 and to the curve of the spatial distribution in Fig. 5 (b).

Comment / Suggestion:

Moreover I would see superimposed to fig. 5 right plots 2 more plots of other slices (scaled to fit same height): e.g. fig. 5a a slice outside the liquid and a slice with liquid meniscus fig 5.b also left and right slices/band near the central one that is shown now.

Reply:

With Fig. 5 (a) we show that the radioluminescence emanates evenly from the liquid. The color map already shows this, the slice makes it even clearer.

With Fig. 5 (b) we show that there is increased radioluminescence at the liquid-air boundary, that reflections from the sample holder explain an increase at the lower parts of the cuvette and that the top parts of the cuvette are contaminated. The color map already shows this, the slice makes it even clearer.

The current slices highlight the mentioned key findings of the paper. We added the labels in Fig. 5 (b) to make it even clearer. More slices would shift the focus of the figure away from those the key results. However, the color map contains all information about the spatial distribution, additional details can easily be obtained from there.

Comment / Suggestion:

2) Radioluminescence spectrum.

The shape of fig 7.b is analyzed to infer that: “The steep decrease at the short wavelength side of the spectrum makes it unlikely that the observed light is Cerenkov light, bremsstrahlung or any other similar broadband emission.” It is not clear to me how the absorption coefficient of water/cuvette and readout system have been accounted for. In particular in liquid water a very strong absorption is expected exactly below 400nm as measured. Also in this case I would expect to see the correction for these absorption effects or to see the comparison/calibration of the measured spectra with a reference one using the same experimental set-up. Without similar quantitative analysis is meaningless the inference about the physical mechanism responsible for the observed luminescence.

Reply:

In both spectral measurements we take into account the effects of all elements in the optical
path. We clarified this by amending our description of the experiment with

> During post-processing the spectral data we accounted for effects that alter the measured spectrum. The cuvette, the light guide and the PMT have a flat response. They can change the intensity of the light they respond to but they cannot alter the shape of the spectrum of this light. The reflectance of the grating has been calibrated to give a flat spectral response. A 10 mm thick water column has a negligible absorption in the measurement wavelength range [24], thus it has no effect on the measured spectral shape.

We further enhanced the manuscript by adding the following clarification to the discussion of the spectrum:

> The correct spectral shape of the well-known radioluminescence of air in Fig. 7 (a) verifies that the calibration of spectral response has been successful and that the spectral shape of the liquid radioluminescence in Fig. 7 (b) can be trusted. The only additional elements in the optical path are the quartz cuvette and the water column, which both have negligible absorption at the measured wavelength range from 280 nm to 550 nm.

Sincerely,

Thomas Kerst
Corresponding author

Tampere University
Physics Unit
Photonics Laboratory
P.O. Box 692
FI-33101 Tampere, Finland
+358 50 300 5968
thomas.kerst@tuni.fi