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Influence of radioactive decay on actinide magnetic susceptibility measurements by the Evans method

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In order to explain the higher magnetic susceptibility of some aquo actinide ions than predicted by Hund's rules, the molar magnetic susceptibility of two americium's isotopes (²⁴¹Am and ²⁴³Am) were measured using the Evans method. Results obtained show a growing change in the magnetic susceptibility with α but also a β^- activity increase in solution. β^- particle effects appear stronger than radicals formed by α particles on the experimental values. A temperature dependence of Am(III) magnetic susceptibility has been observed but from experiments carried out here, it appears difficult to prove whether this effect arises from radicals or β^- . Finally, americium magnetic susceptibilities recorded in different media (HClO₄, HCl, HNO₃) have been compared to alpha and beta emissions' impact.

1. Introduction

Recently, a comprehensive examination of actinide paramagnetism in solution was undertaken in aqueous solution in order to obtain information about unpaired electron behaviours depending on their electronic states¹. In order to compare actinide ions in similar conditions, magnetic behaviour of actinide elements was studied in perchloric media (only water molecules are supposed to surround cations in the first coordination sphere) through bulk magnetic susceptibility (BMS) measurements using the NMR Evans method^{1,2}. Measurements of higher Am(III) magnetic susceptibility that predicted by Hund's rules and about two-fold that for americium compounds determined by SQUID magnetometer³ led to the consideration of a possible radioactivity influence on actinide cations magnetic susceptibility measurements in solution through the Evans' method. It is known that water radiolysis generates short life intermediates such as hydrated electrons (e^-_{aq}), hydrogen atoms ($^{\circ}H$) and hydroxyl radicals ($^{\circ}OH$). These are paramagnetic species and furthermore able to react with solutes to form secondary species (H₂; H₂O₂ ...) that can also lead to a slight chemical change of the solution^{4,5}. All species formed by water radiolysis can be summarized as follows:



Further, because the study was carried out in perchloric acid, the formation of other species by irradiation or reaction of radicals with perchlorate ions can contribute to a chemical change of the sample studied. The reaction of perchlorate ions with free radicals generated in water is known to be slower than for other anions such as nitrates or chlorides⁶. Moreover, it has been reported that degradation products are mainly formed by the direct action of radiations on perchlorates ions⁷. Thus, this secondary species formation (ClO₃⁻, Cl⁻) can lead to a composition variation of the working solution which is different from the reference and means a BMS drift as measured by the Evans method.

Radiolysis of an aqueous solution may have different effects depending on radiation type. These differences come from their linear energy transfer (LET) corresponding to the average amount of energy deposited per unit of track length (eVnm⁻¹). Heavy charged particles such as α particles (He²⁺) have short tracks and high LET, so they may lead to a significant radical concentration. On the contrary, radiations with a low LET (β^- , γ) have long distances and deposit only a small amount of energy in solution⁸⁻¹¹. Thus, for all analysed samples, it may be considered that the radicals' formation by radiolysis is mainly due to alpha emissions'. Radical life times are short because the reaction rate with solute¹¹, but they are continuously generated and so these species could account for a

significant increase in magnetic susceptibility measurements. This effect is particularly expected for cations with low BMS. The main objective of this study was to determine the radioactivity influence of these elements on magnetic susceptibility measurements by Evans' method. As explained above, the choice of Am(III) is justified by its weak susceptibility, one of the lowest among the paramagnetic actinide cations. Thus, the observation of an additional paramagnetism provided by traces of radicals or hydrated electrons should be facilitated. The two main americium isotopes available in the lab are ^{241}Am and ^{243}Am . They have very different radioactive decay rates ($T_{1/2} = 433$ years and $T_{1/2} = 7380$ years respectively). Regarding ^{243}Am , the ^{239}Np formed by radioactive decay must be removed to avoid any β^- emission sources. In order to prove the potential influence of radicals formed by radiolysis, a comparative study of these two americium isotopes, using recently purified samples, was performed varying time, Am concentrations and temperature conditions. In order to separate radicals and paramagnetic element effects from BMS measurements, the issue of magnetic susceptibility changes depending on alpha radioactivity, controlled by Am concentration in the solution, was addressed. Finally, it was considered interesting to observe the influence of β^- emission due to ^{239}Np formation. β^- particles are species assumed to be hydrated electrons that could skew magnetic susceptibility measurements. In fact, the study of ^{239}Np formation by ^{243}Am decay represents a starting point for further understanding of β^- emission impact.

2. Experimentals

^1H NMR spectra of ^{241}Am and ^{243}Am were recorded using 400 MHz Fourier transform spectrometers, Agilent DD2 and Avance III, set up for the study of radioactive samples. BMS were collected at every 5°C step in the $5 - 50^\circ\text{C}$ temperature range.

UV-vis spectra were recorded with a Cary 50 spectrophotometer using a 1 cm optical path quartz cell.

Determinations of actinide concentrations with γ spectrometry were performed using a Canberra spectrometer (electronic resolution: 100 keV – 2000 keV) with a germanium N-type detector.

2.1. Sample preparations

For magnetic susceptibility measurements, two americium samples with different isotopic compositions were used:

Sample 1: ^{241}Am (98.739%), ^{242}Am (<0.02%), ^{243}Am (1.251%).

Sample 2: ^{243}Am (96.6%), ^{241}Am (3.4%).

In order to separate Am(III) from neptunium formed by radioactive decay, a cationic resin DOWEX 50W-X8 was used to remove all cations with a lower charge than Am(III). Because Np(V) is the most stable neptunium oxidation state (NpO_2^+) the assumption was made that the neptunium formed in the Am solution is mainly at this oxidation state. The resin was firstly equilibrated in nitric solution at pH=1 before being introduced into the column. A 4 cm height of

cationic resin was carefully and evenly filled to avoid any preferential route during the americium elution. Am(III) fixation on the resin at low acidity (pH \approx 1): the Am stock solution was adjusted beforehand to the desired pH by adding 6 M sodium hydroxide dropwise, and then slowly added into the column to reach total fixation of americium. Neptunium elution was next carried out by successively adding small volumes of 0.1 M nitric acid. 24 mL were required to complete Np(V) elution (checked by dose rate measurements at the column contact). Then, Am(III) elution was performed at higher acidity using 15 mL of nitric acid ($[\text{HNO}_3] = 5$ M) and checked by dose rate measurements in contact with the eluate flask and the column. The americium hydroxide $\text{Am}(\text{OH})_3 \cdot x\text{H}_2\text{O}$ was precipitated by adding sodium hydroxide 6 M to the eluate solution and the supernatant then eliminated by centrifugation. The precipitate was washed three times with 0.1 M sodium hydroxide to eliminate the saline medium and avoid Am(III) loss by partial resolubilization during the water washing. The wet solid was dissolved in deuterated perchloric acid containing 0.1 M of t-BuOH, to obtain an Am(III) concentration of 0.1 M in 1 M HClO_4 . 300 μL (**Sample 1**) and 90 μL (**Sample 2**) of this solution underwent NMR analysis via a specific capillary tube while the remaining solution was set apart for americium concentration determination (γ counting and UV-visible spectrophotometry). Similar proceedings were carried out for Am(III) solutions in nitric and hydrochloric media.

2.2. Measurements

The molar magnetic susceptibilities of both americium isotopes were calculated by chemical shift difference $\Delta\delta$ between ^1H NMR signal of working (t-BuOH in) and reference (t-BuOH out) solution using the Evans method².

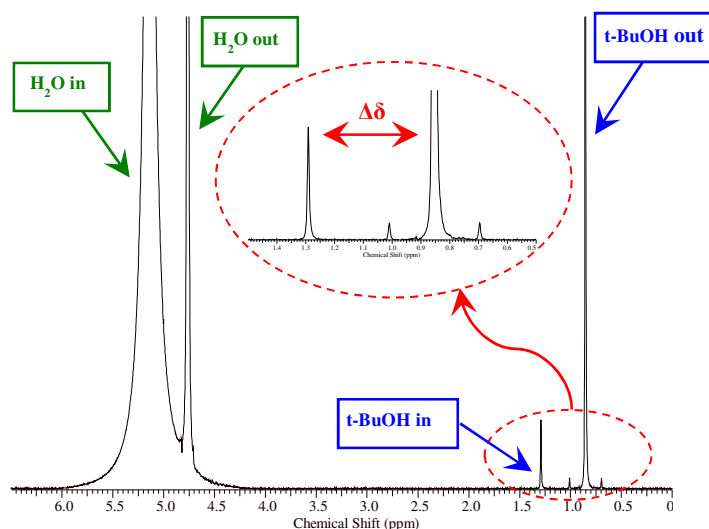


Figure 1 : ^1H NMR spectrum of t-BuOH (0.1 M) for magnetic susceptibility measurement of Am(III) in HClO_4 1 M (sample 1) at 25°C .

Uncertain values were estimated by taking into account $\Delta\delta$ accuracies of NMR spectra (**Figure 1**) and concentrations gained

from γ counting and UV-visible spectrophotometry (Figure 2).

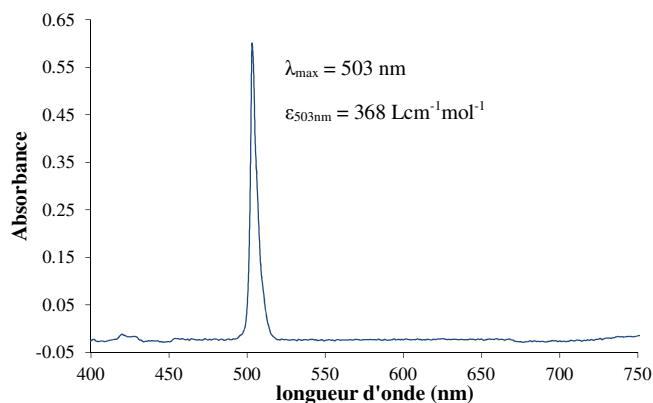


Figure 2 : UV-visible spectrum of Am(III) solution (dilution 60) in perchloric medium 1 M (sample 1).

3. Results and discussion

3.1. BMS of both Am(III) isotopes

One of first magnetic susceptibility measurements on Am(III) in solution at 25°C was made by Howland and Calvin¹² in 0.5 M HCl medium with a 0.01 M americium concentration involving a low α activity in solution $1.36 \cdot 10^{11}$ BqL⁻¹ (probably constituted of ²⁴¹Am: 41%, ²⁴³Am: 59%). In this study (0.096 M and 0.112 M), the α radioactivity in solution is $3.06 \cdot 10^{12}$ BqL⁻¹ and $3.99 \cdot 10^{11}$ BqL⁻¹ for sample 1 and 2 respectively. These values are thus three and twenty times greater than those in the Howland and Calvin¹² study.

Table 1 : Molar magnetic susceptibilities at 25°C in solution for several americium(III) concentrations. * just after purification

Sample	Concentration (molL ⁻¹)	Activity (BqL ⁻¹)	χ_M (10 ⁻⁸ m ³ mol ⁻¹)	Medium
*Sample 1	0.096	$3.06 \cdot 10^{12}$	1.408 (± 0.038)	HClO ₄ 1 M
*Sample 2	0.112	$3.99 \cdot 10^{11}$	1.163 (± 0.025)	HClO ₄ 1 M
Howland and Calvin ¹²	$1 \cdot 10^{-2}$	$1.36 \cdot 10^{11}$	0.905	HCl 0.5 M

Values determined at 25°C during this work are given in Table 1. These results show a growing magnetic susceptibility with increasing α activity in the solution. BMS measurements performed by Evans method are clearly sensitive to α activity.

The paramagnetic behavior study of both isotopes in the 5 – 50°C temperature range presented in Figure 3 was performed to investigate the origin of the deviations observed immediately after americium purification.

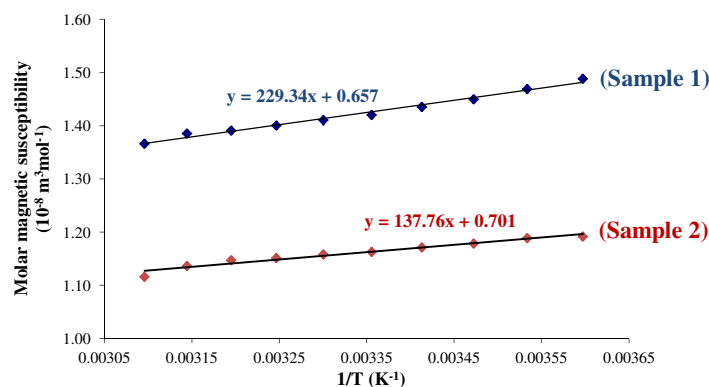
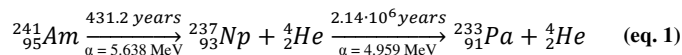


Figure 3 : Magnetic susceptibility fluctuation vs 1/T for samples 1 and 2 in perchloric medium 1 M (just after purification).

Molar susceptibility measurements (χ_M) performed on both Am(III) samples show their magnetic behavior differs with temperature, since the slope of the plot $\chi_M = f(1/T)$ is measurably greater for sample 1 (²⁴¹Am) than for sample 2 (²⁴³Am). Since radical formation is temperature independent, this trend can be directly related to α activity variations between the ²⁴¹Am and ²⁴³Am samples. This suggests that the radicals have a temperature dependent paramagnetism that leads to a change in the magnetic behavior of measured samples. Moreover, it is noteworthy that the intercepts are quite similar for both curves despite the low accuracy arising from the slope extent. Hence, the radioactivity effect does not seem to affect the temperature-independent part of the magnetic susceptibility.

3.2. α particle influence on BMS measurements

In order to more accurately characterize magnetic susceptibility change with solution radioactivity, measurements were carried out with different ²⁴¹Am concentrations since ²⁴¹Am decays through α -particle emission. Americium concentration adjustments were performed by successive dilutions of the stock solution which was the starting point of susceptibility measurements. The calculation of the α solution radioactivity after purification was achieved via the ²⁴¹Am concentration and the following radioactive decay:



The molar magnetic susceptibility vs solution activity plot (Figure 4) clearly shows that the Am(III) paramagnetic susceptibilities measured increase linearly with the α radioactivity, demonstrating the likely influence of radicals formed by radiolysis.

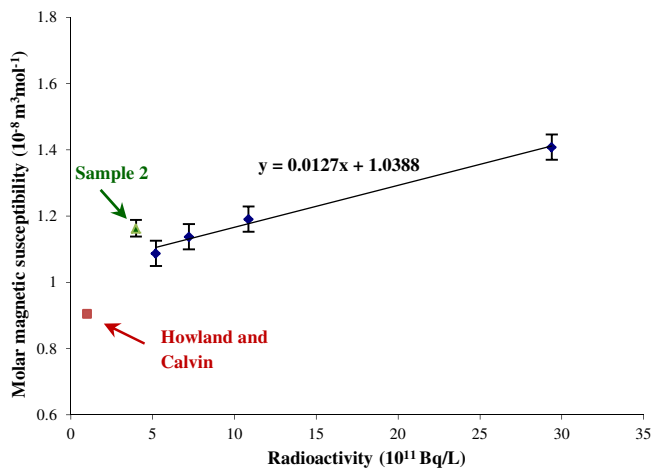


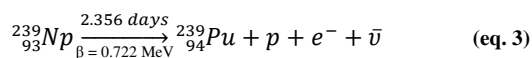
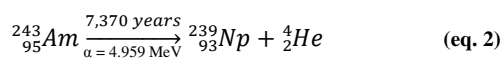
Figure 4 : Molar magnetic susceptibility of ^{241}Am (sample 1) with α solution radioactivity in HClO_4 1M at 25°C .

Assuming the curve follows a linear law, α radioactivity related to the americium concentration clearly increases Am(III) BMS measurements χ_M of $0.13 \cdot 10^{-23} \text{ m}^6 \text{ mol}^{-1} \text{ Bq}^{-1}$. Extrapolating this law to the sample radioactivity used by Howland and Calvin¹², the Am(III) magnetic susceptibility would be $1.06 \cdot 10^{-8} \text{ m}^3 \text{ mol}^{-1}$. This value is 17% higher than the published value ($0.905 \cdot 10^{-8} \text{ m}^3 \text{ mol}^{-1}$). This shift can be assigned either to the different apparatus used (a bifilar suspension method and the Evans method), experimental error (no uncertainties are reported for Howland and Calvin measurements) or, more likely, to a nonlinear BMS behavior at weak radioactivity.

Applying this extrapolating law to Sample 2 radioactivity ($3.99 \cdot 10^{11} \text{ Bq L}^{-1}$) leads to a $1.09 \cdot 10^{-8} \text{ m}^3 \text{ mol}^{-1}$ BMS. This calculated value is only about 6% different from the experimental result observed and can be ascribed to experimental error. Error on magnetic susceptibility measurements is mainly due to the americium concentration determined by UV-visible spectrophotometry and γ counting. Moreover, Am(III) concentrations measured with two different devices for Sample 1 and Sample 2 may have caused larger deviations.

3.3. Time study

To complete values obtained on alpha radiolysis impact, it was particularly interesting to study the effect of beta emissions on susceptibility measurements, as, this type of radioactive decay (β^-) leads to the emission of electrons in solution that can disturb magnetic susceptibility measurements by Evans method. The study of ^{239}Np formation (short period isotope) by ^{243}Am decay was carried out with this in mind.



The half-life of this element is relatively short ($T_{1/2} = 2.356$ days) and leads to a transient equilibrium 24 days after purification (Figure 5). Therefore, a BMS study of americium Sample 2 was performed over 37 days in order to observe any effect arising from β^- particle concentration on magnetic susceptibility measurements.

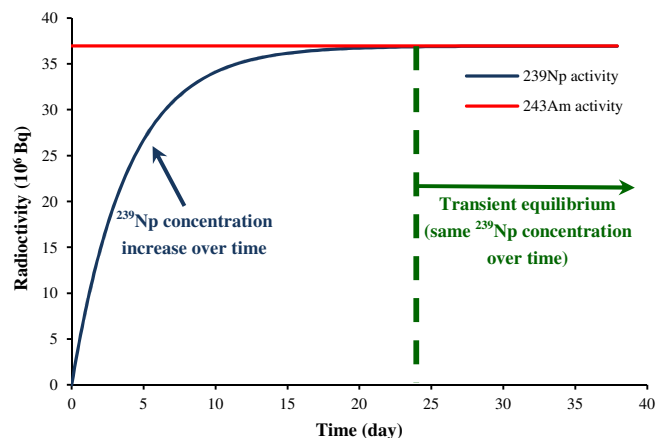


Figure 5 : Radioactivity evolution of ^{243}Am and ^{239}Np radio-isotopes over time.

The monitoring of the chemical shift difference ($\Delta\delta$) changing over time (Figure 6) shows two main areas. A first region (1) occurring within the first 21 days after purification exhibits a quick increase of the magnetic susceptibility, followed by a stabilization. It seems to coincide with gradual production of ^{239}Np in solution but could also be explained by an accumulation of alpha radiolysis secondary products. The second region (2) for $t > 25$ days shows a greater variation of the magnetic susceptibility over time which keeps on growing. These two trends of magnetic susceptibility as measured by the Evans method are discussed hereafter.

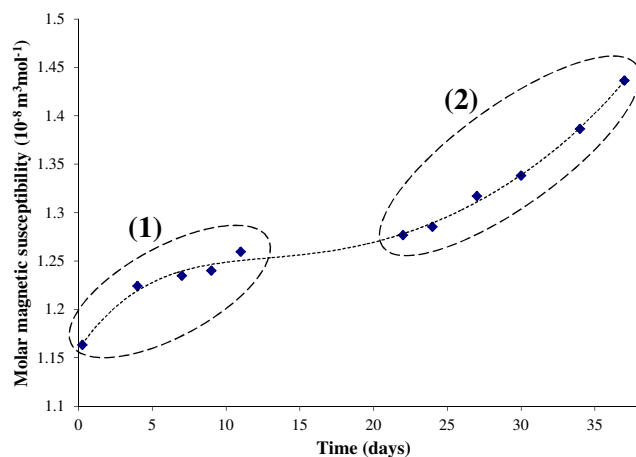


Figure 6 : Changes to Sample 2 (mainly ^{243}Am isotope) magnetic susceptibility χ_M over time after Am(III) purification at 25°C , according to the Evans method.

3.3.1. β^- emission influence

Considering only the first region (I) described above, it was possible to correlate the magnetic susceptibility measured in solution with the ^{239}Np radioactivity in order to characterize the β^- emission influence on susceptibility measurements by the Evans method.

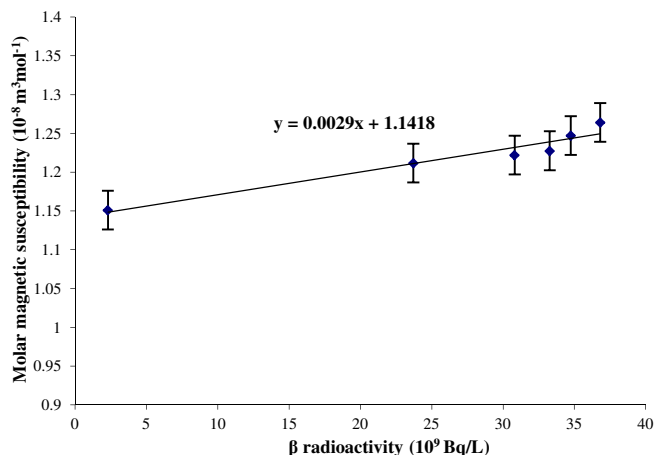


Figure 7 : Molar magnetic susceptibility of Sample 2 with β^- radioactivity in solution at 25°C.

The resulting plot **Figure 7** shows a linear variation between magnetic susceptibility and β^- activity increasing over time. The intercept provides a ^{243}Am magnetic susceptibility without solvated electrons (β^- emissions) of $1.154 \cdot 10^{-8} \text{ m}^3 \text{ mol}^{-1}$. From the slope of this straight line and similarly to α activity, it can be deduced that β^- radioactivity leads to a magnetic susceptibility increase of $2.9 \cdot 10^{-23} \text{ m}^3 \text{ mol}^{-1} \text{ Bq}^{-1}$. It was surprising to find out that the magnetic susceptibility increase due to β^- emissions is 23 times more efficient than for α particles at a given activity. This result was unexpected because α particles are more likely to interact with other atoms and lose their energy (about several MeV) than β^- particles because of their mass. The β^- efficiency observed could be due to direct effects on BMS measurements, in contrast to α particles which have to produce radical species in order to change BMS results. By subtracting the χ_M part related to the α activity from our ^{243}Am sample (Sample 2: $3.99 \cdot 10^{11} \text{ Bq L}^{-1}$) thanks to the results gained from ^{241}Am experiments (Sample 1), a magnetic susceptibility of $1.05 \cdot 10^{-8} \text{ m}^3 \text{ mol}^{-1}$ can be deduced for Am(III) at the same activity used by Howland and Calvin¹². This value is very close to the previous one obtained from the ^{241}Am experiment after α correction ($1.06 \cdot 10^{-8} \text{ m}^3 \text{ mol}^{-1}$).

Both approaches enabled the experimental determination of the Am(III) magnetic susceptibility to be assessed as approximately $1.06 (\pm 0.03) \text{ m}^3 \text{ mol}^{-1}$ at room temperature. This value is consistent with the results reported by Howland and Calvin¹² but nevertheless 17% higher. As mentioned previously (3.2), this difference might be explained by the fact that we considered a linear dependence that occurs whatever the activity level. It is not yet determined whether this holds at low activity levels.

3.3.2. Radiolysis influence

Beyond 25 days of study, an increase in magnetic susceptibility was observed (**Figure 6**). This second trend has been ascribed to the accumulation of secondary products (ClO_3^- , Cl^-) generated from radical recombination over time, leading to a sample that is far from its initial composition. This observation meant a reinterpretation of the first trend. It could be that the first evolution previously connected to ^{239}Np production in solution may also coincide with a gradual increase of H_2O_2 in solution, followed by a stabilisation of this species concentration by chemical and radiolytic degradation processes. It can be seen in **Figure 6** that this equilibrium was reached 10 days after americium Sample 2 preparation. In 1964, Katakis and Allen⁶ demonstrated that in HClO_4 medium, the amount of hydrogen peroxide was stable after 100 minutes of irradiation ($\approx 600 \mu\text{mol L}^{-1}$) with an intensity of $3.24 \cdot 20^{20} \text{ eV L}^{-1} \text{ min}^{-1}$. In our study, the intensity generated in solution being $5.87 \cdot 10^{19} \text{ eV L}^{-1} \text{ min}^{-1}$, the equilibrium between formation and destruction of H_2O_2 was expected to be delayed. However, the lack of intermediate data hampers a more accurate interpretation. In order to observe the effect of H_2O_2 production on t-BuOH signal, measurements were performed by the Evans method on a water sample containing H_2O_2 (3 M).

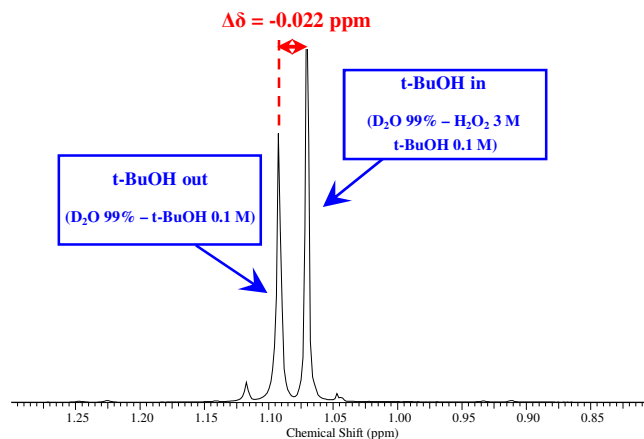


Figure 8 : ^1H NMR spectrum of t-BuOH (0.1 M) containing H_2O_2 (3 M) in the working solution (in) compared to the reference (out) at 25°C.

The NMR spectrum (**Figure 8**) shows that the t-BuOH signal shifted slightly at such a high H_2O_2 concentration and therefore, H_2O_2 production by water radiolysis is not the source of the first deviation observed.

The ClO_3^- and Cl^- formation after recombination of the solvent with free radicals formed by radiolysis was four times lower than the production of H_2O_2 ¹³. However, the production yield of these species is constant over time and no stabilisation is observed^{6, 13}. Thus, the BMS increase observed in the long term could be assigned to secondary product formation. In order to prove this effect,

measurements were made using the Evans method on water samples containing ClO_4^- , ClO_3^- and Cl^- ions ($\approx 4 \text{ M}$) respectively. The magnetic susceptibilities calculated for these ions are given in Table 2.

Table 2 : ClO_4^- , ClO_3^- and Cl^- magnetic susceptibility at 25°C in D_2O .

Ions	Concentration (molL^{-1})	χ_M ($10^{-8} \text{ m}^3 \text{ mol}^{-1}$)
ClO_4^-	4.05 (± 0.08)	-0.020 (± 0.001)
ClO_3^-	4.22 (± 0.08)	-0.021 (± 0.001)
Cl^-	4.31 (± 0.09)	-0.032 (± 0.001)

Whereas the magnetic susceptibility of ClO_3^- ions is similar to the situation for ClO_4^- ions, the Cl^- ions show a higher diamagnetic susceptibility. These experiments reveal a low effect of degradation products on magnetic susceptibility measurements by the Evans method except for Cl^- ions. Considering the reaction yields in perchloric acid 1 M given by Bugaenko and Maksimov¹³ and our dose rate, $[\text{Cl}^-]$ and $[\text{ClO}_3^-]$ degradation product concentrations were about 0.013 M and 0.025 M respectively. These concentrations lead to a $1 \cdot 10^{-3}$ ppm additional shift which is lower than the chemical shift observed on the Am(III) NMR spectrum. Thus, the effect of these ions is opposite to the effect observed on an Am(III) NMR spectrum, and cannot explain the shift seen on the NMR spectra.

This assumption is also supported by the decreasing quality of ^1H NMR spectra, given that splitting of the t-BuOH peak or shoulder peaks associated with the emergence of new ^1H signals can be observed. This feature corroborates the assumption of degradation product formation contributing to a medium change and consequently to a BMS variation. Integration measurements performed on the t-BuOH signal show a significant loss of this compound in the working solution compared to the reference. Thirty days later only 13% of the t-BuOH was available in the inner part to ensure BMS measurements. In other words, 87% of t-BuOH contributes to secondary degradation product formations. Among them t-BuCl, CH_3Cl or CH_3OH have ^1H chemical shifts in agreement with the main new peaks observed around 1.5 to 2.5 ppm. Diamagnetic susceptibility calculated from Pascal's constants¹⁴ exhibit t-BuOH BMS similar to t-BuCl one ($-0.073 \cdot 10^{-8}$ and $-0.080 \cdot 10^{-8} \text{ m}^3 \text{ mol}^{-1}$ respectively) but much larger than those of methanol or CH_3Cl ($-0.027 \cdot 10^{-8}$ and $-0.040 \cdot 10^{-8} \text{ m}^3 \text{ mol}^{-1}$ respectively). Progressive replacement of t-BuOH by less diamagnetic compound in the working solution leads to magnetic susceptibility change of the medium and would explain the chemical shift variation increase presented in part (2) Figure 6. NMR spectra showing the sample changes over time are given in the supplementary information.

3.4. Anion effects on Am(III) BMS

A change in the first coordination sphere of actinide ions due to complexing ions in solution can cause a change in the cation

electron cloud, which can be observed by the study of their magnetic behaviour. In order to characterize the anions' influence, it was considered useful to study Am(III) magnetic susceptibility with various Cl^- and NO_3^- ion concentrations. Therefore, preliminary experiments were performed in 1 M HCl and HNO_3 solutions. To overcome the influence of β^- emissions in solution, measurements were performed immediately after ^{239}Np removal from Sample 2.

Table 3 : Am(III) magnetic susceptibility at 25°C in HClO_4 , HNO_3 and HCl solutions. * just after purification

Actinide cation – oxidation state	Solution	$[\text{An}^{n+}]$ (molL^{-1})	χ_M ($10^{-8} \text{ m}^3 \text{ mol}^{-1}$)
*Am(III) (Sample 2)	HClO_4 1 M	0.112 (± 0.006)	1.163 (± 0.025)
	HNO_3 1 M	0.124 (± 0.006)	1.166 (± 0.026)
	HCl 1 M	0.116 (± 0.006)	1.131 (± 0.025)

From a general point of view, a comparison of the magnetic susceptibilities measured (Table 3) does not reveal any medium influence on Am(III) paramagnetic properties. For An(III) cation, the magnetic susceptibility values determined for the aquo ion (HClO_4), in nitric and hydrochloric solution indicate differences which are only in the magnitude order of experimental errors. This observation demonstrates that the magnetic susceptibility difference between the values found here and the Howland and Calvin value is not explained by a medium difference. Moreover, this very weak influence of anions is in good agreement with EXAFS and spectrophotometric experiments showing no change in the first coordination sphere for such concentrations of Cl^- and NO_3^- ¹⁵. Moreover, the constancy of magnetic susceptibility suggests no medium effect at long distance on the magnetic properties of these cations. In 2010, calculations performed by Danilo and al.¹⁶ regarding the effects of the first hydration sphere and solvent on the electronic spectra of U^{4+} , NpO_2^{2+} and PuO_2^{2+} cations revealed that the effects induced by the solvent are relatively low ($< 100 \text{ cm}^{-1}$) on all energy transitions. For Am(III), the magnetic susceptibility measurements using the Evans method appear more sensitive to radiation (especially β^-) than to the nature of the anion. It can be concluded that higher anion concentrations are required to be able to observe anion effects, and that the nature and the activity of radioactive emissions cannot be ignored if reliable measurements are to be obtained.

4. Conclusions

Magnetic susceptibility studies performed on two americium isotopes (^{241}Am and ^{243}Am) revealed the influence of α and β^- radioactive decay types on measurements obtained by the Evans method. Firstly, the study of these two samples with temperature showed a different behavior, attributed to the radioactive decay difference between these solutions. However it is impossible, at this

stage, to quantitatively prove that the Am(III) magnetic susceptibility dependence with $1/T$ only arises from the radioactive particles β^- and/or radical species. The molar magnetic susceptibility trend was observed as a function of ^{241}Am concentration, thus revealing α emission particles' influence on measurements. However the linearity has to be checked for weak activities. A second work based on the ^{239}Np produced by ^{243}Am radioactive decay showed a correlation between β^- activity and a magnetic susceptibility increase of the sample. This time study revealed two phenomena. The first was initially identified as the β^- emission influence from the ^{239}Np decay, although the assumption of H_2O_2 production has also been postulated. However, the second phenomenon is clearly related to secondary product formation by radiolysis. Thanks to the weak paramagnetic behavior of Am(III), it was proved that the radioactivity of the actinide cations can disturb BMS measurements performed by the Evans method. Quantifications made on α and β^- effects in activity units allow an assessment as to whether radioactivity decays can significantly disturb magnetic susceptibility measurements. These results shed light on why some magnetic susceptibilities found in the literature are slightly higher than expected, and why omitting radioactive decay effects has led to no relevant correlation whereas effects on magnetic susceptibility are expected.

In contrast, studies carried out with americium in different media (HCl and HNO_3) show differences in the range of experimental errors. No conclusion can be drawn regarding the influence of counter-ions except that more concentrated solutions will have to be taken into account for such a study. These results enable the reproducibility of the magnetic susceptibility values to be noted, and support the deviations previously identified in studies based on radioactive decay in solutions.

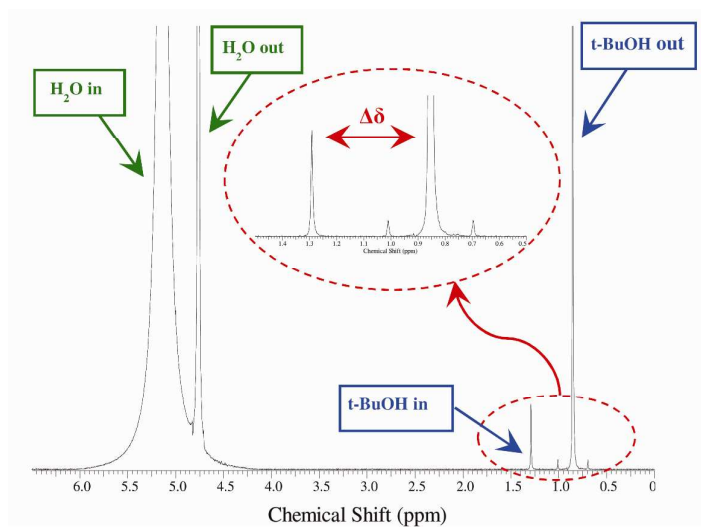
More NMR experiments at different temperatures will be required with both these americium isotopes in order to quantify the extent to which the magnetic susceptibilities arising from radicals and β^- particles could depend on the $1/T$ dependence.

Acknowledgements

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Magnetic susceptibility studies performed on two americium isotopes (^{241}Am and ^{243}Am) revealed the influence of α and β radioactive decay types on measurements obtained by the Evans method.