Experimental report

13/02/2017

Proposal: 9-10-1489 **Council:** 10/2016

Title: Heavy Ions separation: neutron reflectivity and gamma-ray fluorescence at Liquid/liquid interface

Research area: Chemistry

This proposal is a continuation of 9-10-1436

Main proposer: Olivier DIAT

Experimental team: Ignacio RODRIGUEZ LOUREIRO
Ernesto SCOPPOLA
Bertrand BRAIBANT
Luc GIRARD
Olivier DIAT
Gary SIMPSON

Local contacts: Richard CAMPBELL

Samples: D2O
Nitric acid
Dodecane
Gadolinium Nitrate
Lithium Nitrate
C7F8
DMDOHEMA
Fluorinated Malonamide

Instrument	Requested days	Allocated days	From	To
FIGARO	4	3	20/01/2017	23/01/2017

Abstract:

Neutron reflectivity on liquid/liquid (LL) interface will be performed to study the distribution of gadolinium cation across the interface. There are two objectives: a first one that will allow to compare data with previous experiments but at different temperatures. The second is to couple reflectivity and
gamma fluorescence detection to better characterize the Gadolinium distribution at the LL interface.

Experimental report

15/09/2016

Proposal: 9-10-1489 **Council:** 2016-10

Title: Heavy Ions separation: neutron reflectivity and gamma-ray fluorescence at Liquid/liquid interface

Research area: Soft condensed matter

This proposal is a continuation of 9-10-1436

Main proposer: Olivier DIAT

Experimental team: Ernesto SCOPPOLA
Bertrand BRAIBANT
Olivier DIAT
Luc GIRARD

Local contacts: Richard CAMPBELL
Michael JENTSCHEL
Paolo MUTTI

Samples: C6F14
F-DMDBTDMA

Instrument	Requested days	Allocated days	From	To
FIGARO User-supplied	4	3	2016 january 20th at 8h30	to23rd at 9h00

Abstract:

Experimental Report

Solvent Extraction: Gadolinium at Fluroinated Oil/Water interface

The main purpose of the experiment was to study the perfluoro-toluene / water (LL) interface with a perfluorinated malonamide (also called diamide or extractant) in the organic phase and Gadolinium nitrate in aqueous phase. The system was chosen because of the high transmission of neutrons through perfluorinated solvents and to couple the reflectometry and the γ fluorescence for the localization of Gadolinium at the LL interface.
We ask for 4 days, we've got only 3 days. We asked to be at a beginning of a run to not lose too much time for the installation which was fully new and never tested with this new configuration (shielding of the gamma detector)

Investigated samples:
Due to the short time, we have chosen to study the LL interface between Perfluoro-toluene, at two concentration of a specific synthesized extractant (0 and 0.08M modified afterwhile to 0.05 for phase transition problem) and an aqueous solution with various concentration of gadolinium nitrate in 1 M of deuterated nitric acid (0.0 M, 0.05 M, 0.25 M). (a total of 5000 €)
For all these samples, we recorded reflectivity data at two angles (a different configuration than the previous tests where we collected reflected intensity at 3 angles), 0.62 and 2.7° and choosing a large wavelength distribution, [2-16 Å] for the first angle and [2-30 Å] for the second set.
The gamma spectra were recorded only at the first angle where part of neutrons creates an evanescent wave that can excite the Gd cation at the interface.

Results and issues.
As already mentioned we have got only 3 days and not 4 as requested. Then we decided to shortcut our proposal and to reduce the number of investigated samples. 4 days were requested because we knew that the installation and test of the fully shielded gamma detector and more especially its synchronization with the chopper triggering would be difficult and would require several checking.
This is why we ask also to get beamtime at the beginning of a run.
One of our co-workers was present during the week before to start the experiment in order to check with the detector support ILL team that the mechanical and electronic supports are ready. Tests with the neutron beam were not possible due to the 50th birthday of ILL.
Stating on a Friday we lost more than half a day for installation and then a couple of hours for alignment of the cells (two cells to gain beamtime).
The first data were collected during the night and to realize at the end of this first night that the gamma detector acquisition was badly configured in the parameter sets and no gamma spectra were saved. This was solved with the help of the detector support team on Saturday. We lost some samples and some time to clean the cells and restart all our experiments.
We succeed to get data from a minimum of different samples to really some differences with those where Gd ions are condensed or not at the interface.
Unfortunately we discovered at the end of the experiment that the trigger of the chopper was not correctly recorded within the gamma data and that it would be not possible to analyze the reflectivity data taken into account the information of the Gd concentration as a function of q-vectors.
Although the gamma integration as a function of energy appear as correctly recorded, it appears that a gamma detector saturation issues prevent the correct synchronization between the neutron detector and the gamma detector. This issue will have to be discussed with the detector group.
The data are under investigation at this stage and due to the missing information we are not able right now to show a time dependent gamma fluorescence curve.

Preliminary Results
Collected data show that thanks to the upgrade in the experimental setup (lead shielding around gamma detector) the signal to noise ratio increases, as shown in Fig. 1.

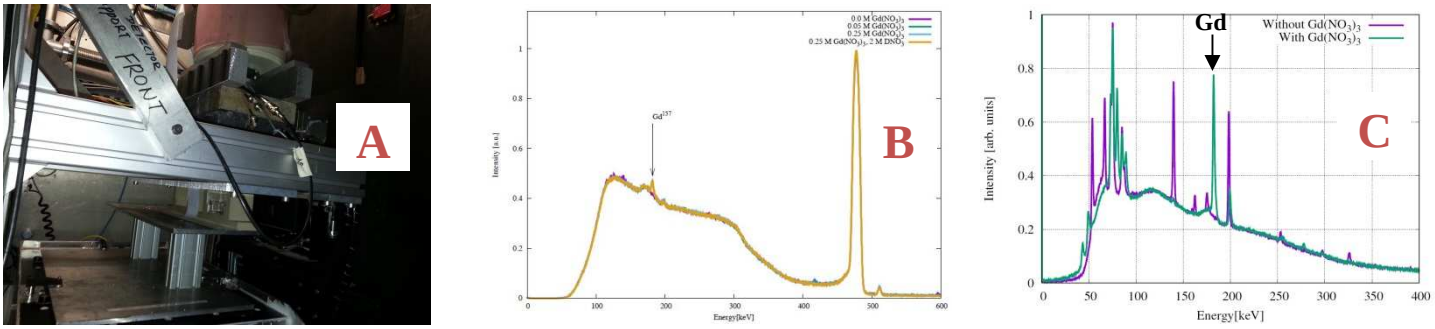

Figure 1. *(A) Picture of Experimental setup for lead shielding around the gamma detector. (B) Gamma intensity as a function of wavelength recorded during previous beamtime at ILL. (C) Gamma intensity as a function of wavelength during the last beamtime on FIGARO. Thanks to the lead shielding we can observe that the signal to noise ratio in the gadolinium emission line increases considerably.*

Regarding the collected reflectivity curve even if the analysis of the experimental data is still under process, we have observed the same features observed in the data collected during the previous beamtime (May 2016). By increasing the concentration of Gadolinium Nitrate in the aqueous solution, the reflected intensity decreases progressively below the critical edge, as shown in Fig. 2 right.

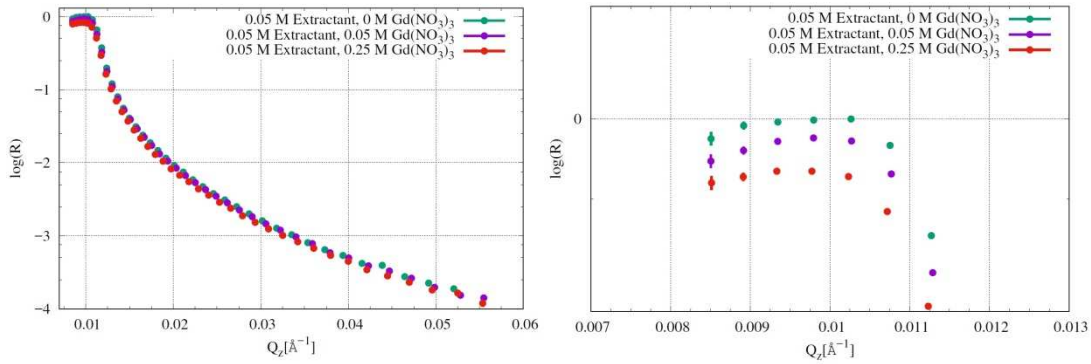

Figure 2. *(Left) Comparison between reflectivity curves collected at the 0.05 M of Extr. in F-Toluene contacted with a gadolinium nitrate (various concentrations) and nitric acid (1 M) aqueous solution. (Rigth) Zoom on the Q_z region below the critical edge. By increasing the concentration of Gadolinium Nitrate in the aqueous phase the "total" reflected intensity decreases due to the absorption of neutron by gadolinium atoms.*

Moreover, the comparison between reflectivity curves for samples loaded with different amount of gadolinium nitrate (Fig. 3) evidences modification on the interfacial structures that can be analyzed. Unfortunately the data analysis is still under process (Beamtime finished on the 23rd of January 2017) and we are not yet able to provide an final interpretation of the collected data.

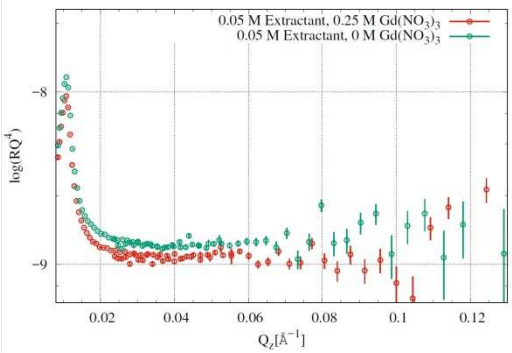

Figure 3. *Comparison between reflectivity curves collected at the 0.05 M Extr. in F-Toluene contacted with a (green) nitric acid (1 M) or (red) nitric acid (1 M) and gadolinium nitrate (0.25 M) aqueous solution.*