Report on the outcomes of a Short-Term Scientific Mission[1]

Action number: CA20129

Grantee name: Leo Sala

Details of the STSM

Title: Ion-induced decomposition of iron pentacarbonyl clusters

Start and end date: 13/12/2021 to 23/12/2021

Description of the work carried out during the STSM

Description of the activities carried out during the STSM. Any deviations from the initial working plan shall also be described in this section.

(max. 500 words)

The goal of the STSM is to study the fragmentation of $Fe(CO)_5$ clusters upon collision with various projectile ions. As such, projectiles with different masses and charge states were used covering various interaction regimes as available during the beamtime: $^{3}He^{2+}$ (16 keV), $^{20}Ne^{4+}$ (40 keV), $^{40}Ar^{8+}$ (64 keV), and $^{20}Ne^{+}$ (6 keV) covering electronic stopping, nuclear stopping, and electron capture dominated ionization mechanisms. The first couple of days of the mission were dedicated to obtaining stable target cluster beams by first assembling an extension to the existing gas line to deliver the vapor from the liquid $Fe(CO)_5$ source into the aggregation chamber where the formation of the cluster beam is facilitated by cold He carrier gas. Parameters were then optimized and within the most stable condition identified, we were able to generate huge cluster sizes of up to ~60 units even at ambient temperature. Only the low mass fragments (up to ~4 cluster units) produced from the big clusters could be accurately resolved by the mass spectra so we focused on this region.

For each projectile, we then recorded the mass spectrum of $Fe(CO)_5$ at different extraction delays to resolve peaks in the low and high mass regimes and whenever conditions allow, we also performed ion-ion coincidence measurements and mapping in the hope of detailing the mechanisms of ion-induced cluster fragmentation. In general, the main goal was achieved although several planned projectiles utilized in gas phase [1] and surface experiments [2] on $Fe(CO)_5$ for direct comparison were not explored due to time and technical constraints. However, most of the interaction regimes are already covered by

[1] This report is submitted by the grantee to the Action MC for approval and for claiming payment of the awarded grant. The Grant Awarding Coordinator coordinates the evaluation of this report on behalf of the Action MC and instructs the GH for payment of the Grant.

the chosen projectiles and ample mass spectrometric data were gathered warranting further analysis in the coming days.

[1] Indrajith, S. et al. J. Phys. Chem. C, 123, 16, 10639–10645 (2019)

[2] Stanford M.G. et al. J. Vac. Sci.Tech. B 35, 030802 (2017)

Description of the STSM main achievements and planned follow-up activities

Description and assessment of whether the STSM achieved its planned goals and expected outcomes, including specific contribution to Action objective and deliverables, or publications resulting from the STSM. Agreed plans for future follow-up collaborations shall also be described in this section.

(max. 500 words)

During the STSM, we acquired plenty of mass spectra of $Fe(CO)_5$ clusters upon collision with various ions. We were able to cover various interaction regimes using different projectiles and this could already give sufficient data to explain various mechanisms of ion-induced decomposition of $Fe(CO)_5$ clusters. Representative spectra at the low and high mass regimes are shown in Fig. 1.

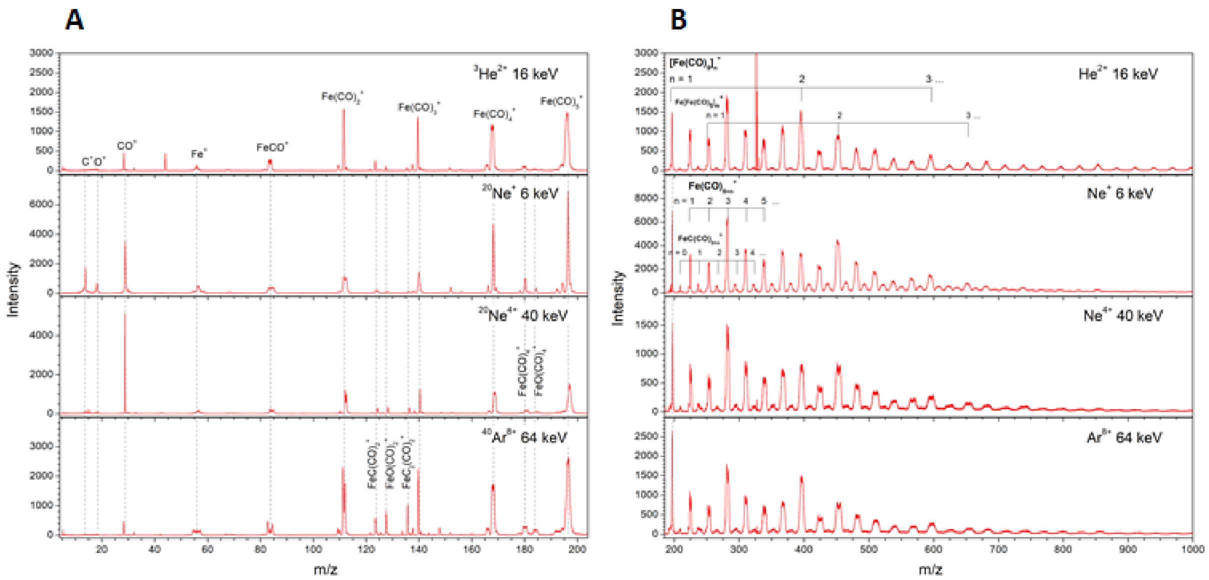

Figure 1. Mass spectra of $Fe(CO)_5$ cluster fragmentation upon impact of various projectiles at the low mass (A) and high mass (B) regions.

At the low mass regime, the peaks observed in the ion fragmentation of isolated $Fe(CO)_5$ [1] are present with varying relative intensities dominated by the parent cation, CO^+, and fragments arising from the loss of CO ligands. A faint $^{56}Fe^+$ peak can be observed which could mean that it remains with the ligands and clusters after impact. Peaks corresponding to C/O attached to the CO loss series $[Fe(CO)_n]$ can also be observed for some projectiles especially $^{40}Ar^{8+}$ which is not evident in the case of the isolated molecule [1]. The high mass regime shows evidence of clustering, and the spectra are dominated by multimer cations and their $(CO)_n$ loss series with similar patterns across all projectiles. Peaks corresponding to C attached to the monomer/multimer + $(CO)_n$ series also appear in between the intense peaks with the + O counterpart also often contributing as a shoulder. To further deconvolute the broad peaks and properly attribute them, we need to resort to spectra at longer extraction delays which we have already at our disposal. The coincidence maps are yet to be carefully analyzed although for the moment, and with the resolution that we have, occurrences of metastable states are not obvious.

Due to the volume and complexity of data gathered, further time is needed for data processing and analysis to eventually decide whether further experimentation is needed to construct a more complete picture that explains the fragmentation of $Fe(CO)_5$ clusters by ions in relation to results of the isolated molecule and surface studies. Nonetheless, we are planning similar experiments using a low energy ion gun to directly compare results with surface studies, and this could also be done in GANIL. With these data, we envision a publication that discusses the implications of these results on nanolithography, specifically in FIBID-related processes which is among the research areas of interest identified by the action.

Research coordination between the Aribe facility and our group in J. Heyrovsky Institute is reinforced and continued during the mission. Bulk of the data analysis will be done in our group with frequent consultations from our collaborators in GANIL. We will also seek collaboration with theoreticians involved in the cost action like the Solov'yov's group whose multiscale approach especially on FEBID/FIBID related systems might help explain the experimental results from this work and relate them to those occurring in actual FIBID systems with the goal of gaining a deeper understanding and eventual improvement of the technique especially in terms of obtaining pure deposits with precise dimensions.