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Overview on the intercalation of gold into graphite

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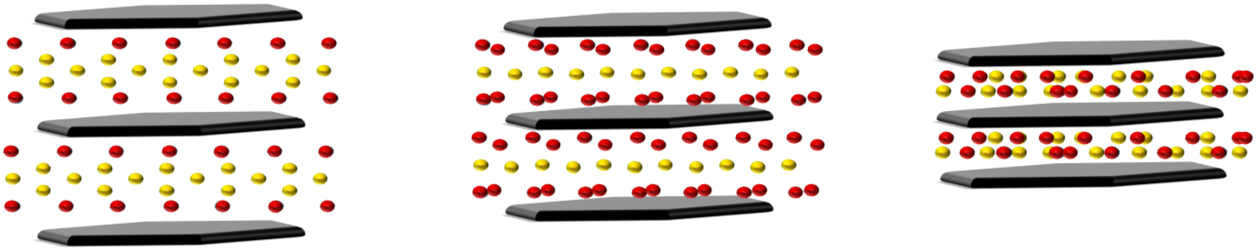
Graphical abstract



Abstract

The intercalation of gold into graphite has been achieved thanks to a solid-liquid method using potassium metal-based molten alloys. Numerous syntheses have been performed varying the reactive alloy composition, the reaction temperature and the reaction time. The optimization of these parameters allowed to synthesize three novel ternary graphite intercalation compounds (GIC) containing poly-layered intercalated sheets, called alpha, beta and gamma. Thanks to a specific X-Ray diffraction study, the *c*-axis stacking sequence of the three compounds was determined. The α-GIC, which is a metastable compound, contains mixed potassium and gold two-layered intercalated sheets. The β-GIC and γ-GIC contain very thick well ordered three-layered and five-layered sheets respectively. The very high repeat distances of the latter compounds are clearly explained by the amount of intercalated metals, precisely determined using nuclear microprobe analyses.

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1. Introduction

Graphite is a lamellar material that can accommodate various species in its Van der Waals's spaces, leading to graphite intercalation compounds. The first synthesized graphite intercalation compound is the KC_8 binary phase. It was prepared in 1926 by Fredenhagen and Cadenbach [1] by direct action of potassium vapor on graphite.

Then many other binary compounds with alkali metals [2, 3], alkaline earth elements [4] and some lanthanides [5-7] have been synthesized. Among these compounds, some have revealed remarkable physical and chemical properties [8-13] so that some of them lead to applications in everyday life [14, 15].

Subsequently, various ternary compounds have been also obtained. The first ternaries were synthesized by combining two alkali metals inside the intercalated sheets [16]. However, most of them are prepared using an alkali metal as an intercalation vector for the elements unable to intercalate by themselves. Indeed these elements can be intercalated into graphite only when they are associated with potassium or another alkali metal. The most known ternaries belonging to this class contain Hg, Tl, As, Sb, Bi [17] associated with K, Rb or Cs. On the other hand, electronegative elements as O, S, Se and Te were equally able to intercalate into graphite in association with potassium [18].

Gold can also be intercalated into graphite by this method [19] leading to three ternary phases, respectively denoted alpha, beta and gamma. This latter has been largely studied [20, 21] because it is the most stable of these compounds. In this paper, we present a full comparison between the chemical and structural properties of the three phases that are significantly different.

2. Experimental Section

2.1 *Synthesis*

The synthesis of potassium-gold graphite intercalation compounds has been performed using potassium as an intercalation vector in liquid potassium-gold alloys. The alloys are prepared in a glove box under a high purity argon atmosphere. Gold and potassium are precisely weighed and introduced in a stainless steel reactor. After obtaining a homogeneous liquid, the graphite sample is then immersed in the liquid alloy and maintained in it thanks to a tungsten sample-holder. Highly ordered pyrographite platelets (PyroGraphiteComprimé Carbone Lorraine - PGCCL) of

$8 \times 2 \times 0.5$ mm^3 in size are used. The reactor is then tightly closed and the intercalation reaction is performed outside the glove box at a given temperature for a precise reaction time. At the end of the reaction, the reactor is opened in the glove box, heated again and the sample extracted from the liquid alloy. The excess of alloy is eliminated using a coupled furnace-centrifuge device and the sample is transferred under argon into a glass capillary tube for X-ray examination.

2.2 *X-ray experiments*

All the samples were analyzed by X-ray diffraction with a Bruker D8 diffractometer using molybdenum Kα1 radiation ($\lambda K_{\alpha 1}$ = 70.926 pm) with a Lynxeye detector, or a theta/2 theta CGR diffractometer with the same radiation. Since the pyrographite platelets are highly oriented, the sample can be placed with the incident beam parallel to the *ab* planes so that the 00*l* reflections are isolated and can be studied separately in order to determine the *c*-axis stacking sequence. The *c*-axis experimental electronic density profile is retrieved from the 00*l* structure factors using the 00*l* intensities.

2.3 *Nuclear Microprobe Analyses*

Nuclear Microprobe Analyses (NMA) were used in order to determine the composition and the homogeneity of GIC [22, 23]. They are based on the interactions between a high energy ion beam in the MeV domain and the probed material. For the experiments with potassium-gold GIC, we used a proton beam of 3050 keV and a particle detector at 170° from beam axis. Rutherford Back Scattering is the only interaction to be taken into account for gold since no nuclear reaction occurs in these beam conditions. For this element, elastic scattering probability presents the advantage to be easily calculated. For carbon and potassium, the choice of this ^{1}H beam instead of the often used ^{4}He beam enables to benefit from enhanced scattering on carbon and potassium with so-called non-Rutherford cross-sections. Thus, a single spectrum leads to the whole information for potassium, carbon and gold on a sample surface of 200 x 200 µm², with a lateral resolution of a few micrometers (beam size 4 x 5 µm²).

Sample compositions are determined by adjustment between simulated spectra and experimental data. The core of the simulation code used, SimNRA [24] is the description of the interaction of the ions with matter such as nuclear interaction kinematics and related cross-sections data, electronic stopping power, etc… It assumes a layered target, each layer having a given composition and a given thickness. The simulated spectrum is then made up of the superimposed contributions of all the interactions, with each isotope of each element of each sublayer of the defined solid.

3. Results and Discussion

3.1 *Investigation of the graphite-potassium-gold system*

It is well known that for the synthesis of GIC using an alkali metal based alloy, the composition of the alloy and the reaction temperature are the main parameters to monitor. A tiny variation of them is able to significantly modify the nature of the obtained sample. Numerous experiments were carried out to explore the Au-K phase diagram between 300°C and 650°C with alloy compositions ranging from 5 to 50 gold at.%, for various reaction times (1 minute to 2 weeks). In all experimental conditions, we observed graphite intercalation compounds either as a pure compound or as a mixture of phases. Four compounds have been obtained, the well-known first stage binary KC_8 compound and three original potassium-gold GIC denoted alpha, beta and gamma according to their repeat distance. Among them, only one has been perfectly isolated. The best synthesis conditions were determined for each compound. The nature of the reaction products depending on the temperature and the composition of the reactive alloy is given Figure 1.

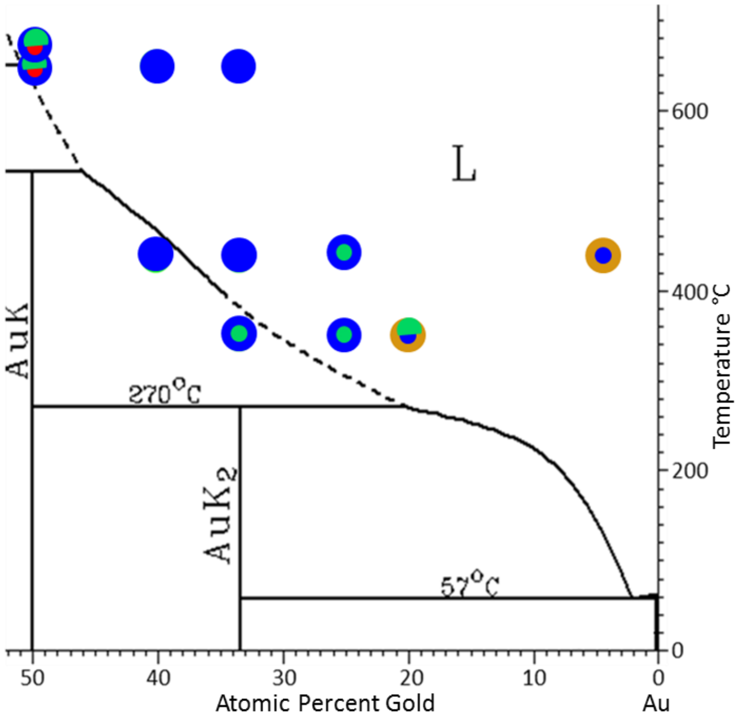

Figure 1: Part of the experimental Au-K phase diagram exhibiting the various GIC belonging to the graphite-potassium-gold system: ●α-GIC; ●β-GIC; ●γ-GIC;●KC$_8$, observed as pure phases or as mixtures.

The binary KC$_8$ compound can be observed in the potassium-rich domain of the phase diagram (≤ 20 Au at.%). It is associated with the γ-GIC for the alloys containing 5 to 20 gold at.%. When the alloy composition increases in gold, the binary KC$_8$ is no more obtained whereas the γ-GIC can be always synthesized in the range of 5 to 50 gold at.% whatever the temperature.

The α-GIC can be only obtained for an alloy containing at least 50 gold at.% at relatively high temperature. This compound is metastable. Indeed this compound seems amorphous after one week, however an annealing of only 4 minutes at 300° C in a glove box allows in part to recrystallize it. The compound denoted β, can be prepared using an alloy containing 20 to 50 gold at.%. This compound could never be isolated, it appeared during the study of the formation mechanism of the γ-GIC. The last compound, the γ-GIC, can be observed in a large range of alloy compositions but it was isolated in a reproducible fashion and a good cristallinity using a Au-K alloy containing 35-40 gold at.% at 440° C for 8 days. The best known experimental conditions for the preparation of ternary compounds are listed in Table 1.

Table 1: Optimal composition of alloy allowing the synthesis of first stage ternary GIC in the graphite-potassium-gold system.

Compound	**Alloy composition** *(Au at.%)*	**T** (*°C*)	**Reaction time** (*days*)
α	50	650	2
β	20 – 50	Not isolated	
γ	34	440	8

3.2 *Formation mechanism study of the γ-GIC*

The formation mechanism of the γ-GIC (Figure 2) has been studied by carrying out several reactions under the best synthesis conditions as indicated in table 1. Pyrographite platelets have been immersed in alloys containing 34 gold at.% at 440 °C for several reaction times (1 minute, 5 minutes, 30 minutes, 1 hour and 2 hours). Then samples have been studied by *ex-situ* X-ray diffraction analysis.

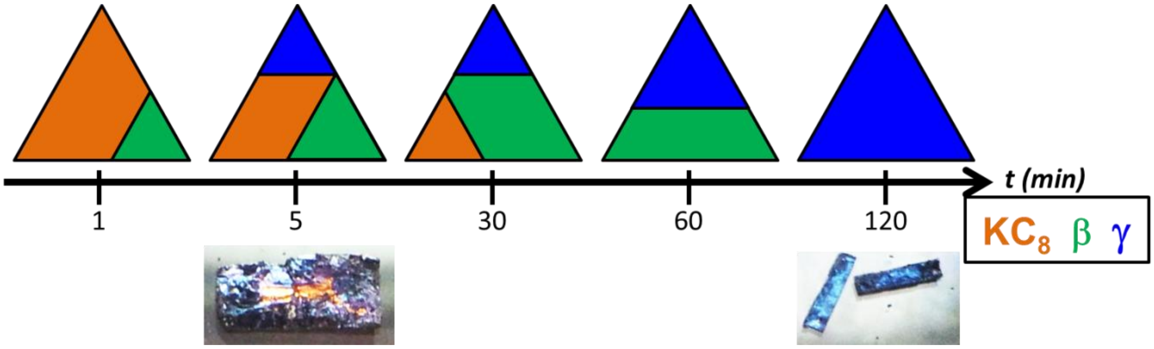

Figure 2: The formation sequence of the γ-GIC in an Au-K alloy containing 34 gold at.% at 440 ° C, for increasing periods of time: ▲ γ-GIC, ▲ β-GIC; ▲ KC_8.

After one minute of reaction, potassium and gold are intercalated into graphite. The result is a phase mixture consisting of the first stage potassium graphite intercalation compound, KC_8 and a first stage Au-K graphite ternary compound (β-GIC).

The γ-GIC is observed as soon as 5 minutes of immersion, the sample obtained is a mixture of 3 phases containing KC_8 and β- and γ-GIC . Increasing reaction time to 30 minutes, a mixture of these three phases is still obtained. Then, after 1 hour of reaction, a mixture of two phases (β- and γ-GIC) is observed. The pure γ-GIC is prepared after 2 hours. Finally, an increase in the reaction time (7 hours, 1 day, 4 days, 8 days) always leads to the synthesis of the γ-GIC.

3.3 *Structural study of the gold-based graphite intercalation compounds*

00*l* diffraction patterns have been recorded for each compound or mixture of phases.

From the 00*l* diffractogram relative to the γ-GIC (Figure 3), a repeat distance of 1311 pm for a first stage compound was determined. This very high repeat distance is characteristic of poly-layered intercalated sheets, probably five-layered ones. The quantitative study of these 00*l* reflections allowed to determine the experimental *c*-axis electronic density profile. The latter was compared to a calculated one; in the corresponding model (Figure 4), the intercalated sheet is five-layered, according to a K-Au-Au-Au-K stacking sequence for a $K_{1.3}Au_{1.5}C_4$ chemical formula [19]. The potassium layers are placed near the graphene planes and frame the gold sheets which is divided in three layers. In this configuration, since the graphene sheets are negatively charged, potassium planes are positively charged and consequently the center of the intercalated sheet made up of gold globally hold a partial negative charge. The experimental data used for the calculation can be retrieved in [20, 21].

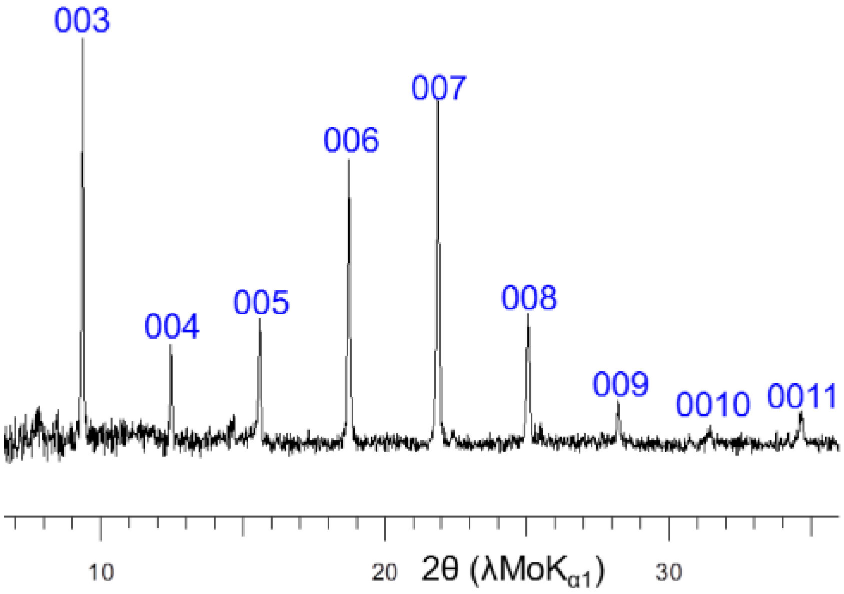

Figure 3: 00*l* X-ray diffraction pattern of the first stage γ-GIC (λMo$K_{\alpha1}$ = 70.926 pm).

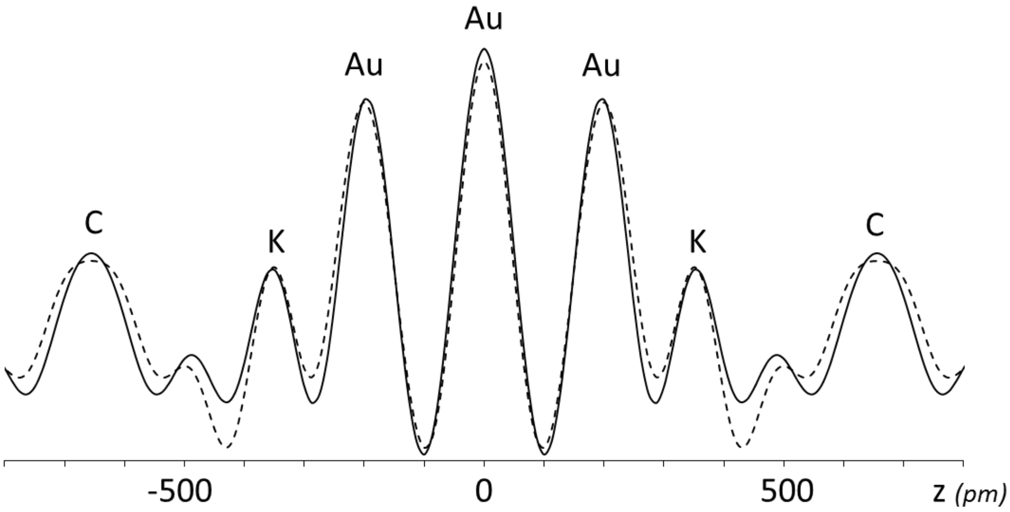

Figure 4: Experimental (dotted line) and calculated 1D electronic density profiles along the *c*-axis for the γ-GIC (R_F = 17.9%). Quantitative data can be retrieved in [20, 21].

During the study of the formation mechanism of the γ-GIC, the β-GIC has been observed. The diffractogram of the β-GIC presented Figure 5 can be indexed as a first stage ternary compound with a repeat distance of 960 pm. It is smaller than that of the previous one and the stacking sequence can be described with a three-layered intercalated sheet containing two superimposed potassium planes surrounding a gold layer. The quantitative study of the 00*l* reflections allowed to draw the *c*-axis electronic density profile (Figure 6). The experimental profile is compared with the one calculated from a model. The experimental and calculated data are reported in table 2. The best agreement between model and experiment is obtained for a model of intercalated sheets which can be described by a K-K-Au-K-K stacking sequence, the potassium planes being separated by 60 pm. The atomic amount and position of each species in the model are specified in table 3. The corresponding chemical formula is $K_{1.6}Au_{0.7}C_4$.The residual factor reaches then 5.8 %.

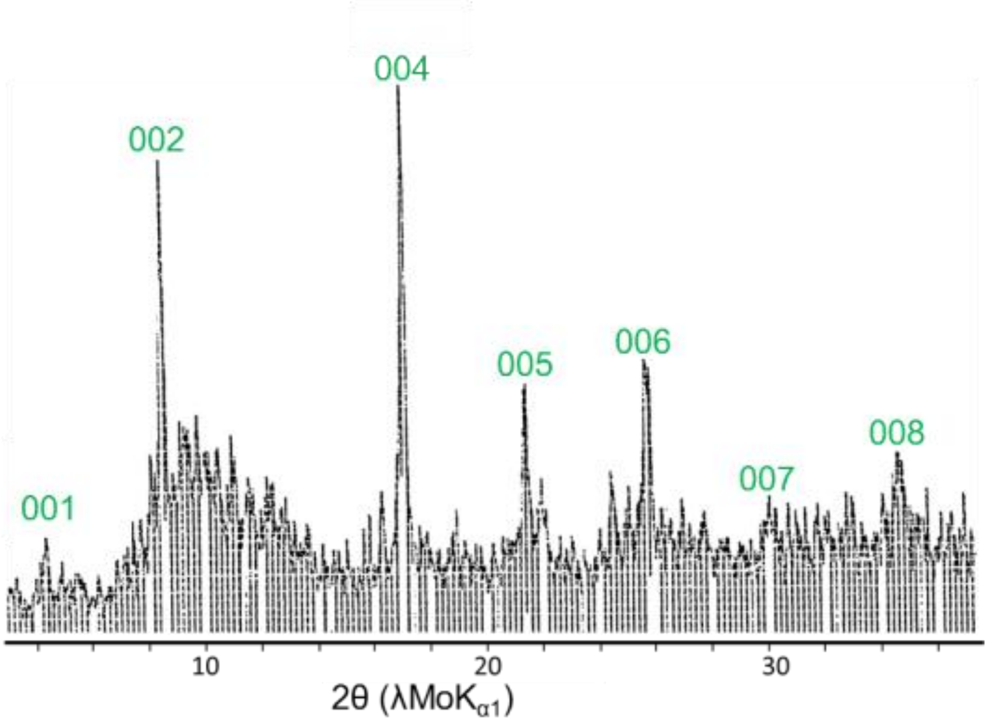

Figure 5: 00*l* X-ray diffraction pattern of the β-GIC (λMoK$_{\alpha1}$ = 70.926 pm).

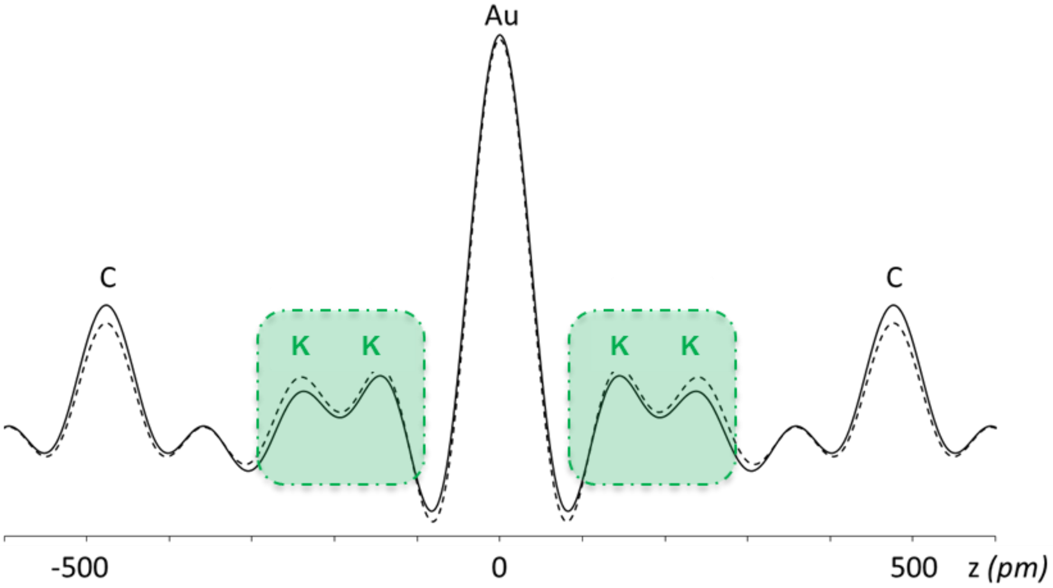

Figure 6: Experimental (dotted line) and calculated 1D electronic density profiles along the *c*-axis for the β-GIC. (R_F = 5.8%).

Table 2: Experimental and calculated intensities and structure factors of the 00*l* reflections of the β-GIC.

00*l*	**θ** (°,(λMoK$_{\alpha1}$))	**d$_{00l}$** *(pm)*	**I$_{00l}$ exp**	**I$_{00l}$ calc**	**F$_{00l}$ exp**	**F$_{00l}$ calc**
1	2.13	953	100	100	59	59
2	4.27	476	50.60	86.62	60	78
3	6.41	318	7.09	6.56	28	26
4	8.56	238	68.09	69.38	100	100
5	10.72	191	23.17	22.91	66	65
6	12.90	159	27.25	27.86	79	79
7	15.10	136	8.52	6.46	48	42
8	17.32	119	13.23	12.63	65	63

Table 3: The atomic amount and position of each species in the β-GIC.

Element	**C**	**K**	**K**	**Au**	**K**	**K**	**C**
Position (*pm*)	-476.5	-230	-170	0	170	230	476.5
Atomic content	2	0.34	0.46	0.70	0.46	0.34	2

When the amount of gold in the reactive alloy increases, the α-GIC can be obtained. It exhibits the smallest repeat distance of all compounds in this system. Typical 00*l* X-ray diffraction pattern (Figure 7) shows intense and individualized Bragg peaks which can be indexed considering a first-stage graphite intercalation compound with a repeat distance of 500 pm.

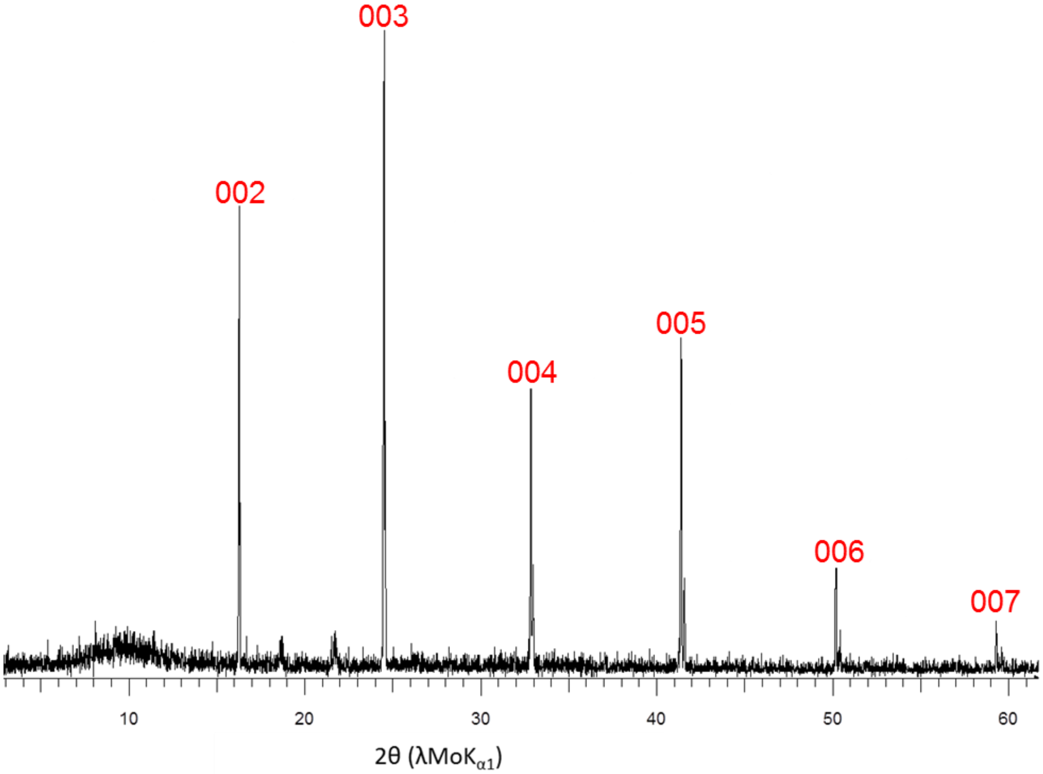

Figure 7: 00*l* X-ray diffraction pattern of the α-GIC ($\lambda MoK_{\alpha 1}$ = 70.926 pm).

The *c*-axis electronic density profile can be determined by the quantitative study of the 00*l* reflections. Taking into account the low interplanar distance (500 pm), a model containing a single layer seemed to be the best suitable but is was not possible to have a good agreement between experiment and any single layer model. In fact, the best obtained model (Figure 8) can be described as a two-layered intercalated sheet, each layer containing both gold and potassium for a chemical formula corresponding to $K_{1.03}Au_{0.68}C_4$. The experimental and calculated data are reported in table 4. Data relative to the atomic stacking model along the *c*-axis for the α-GIC are specified in table 5. The residual factor of 22% shows that this model might be perfectible, but corresponds to the best result obtained with our current data.

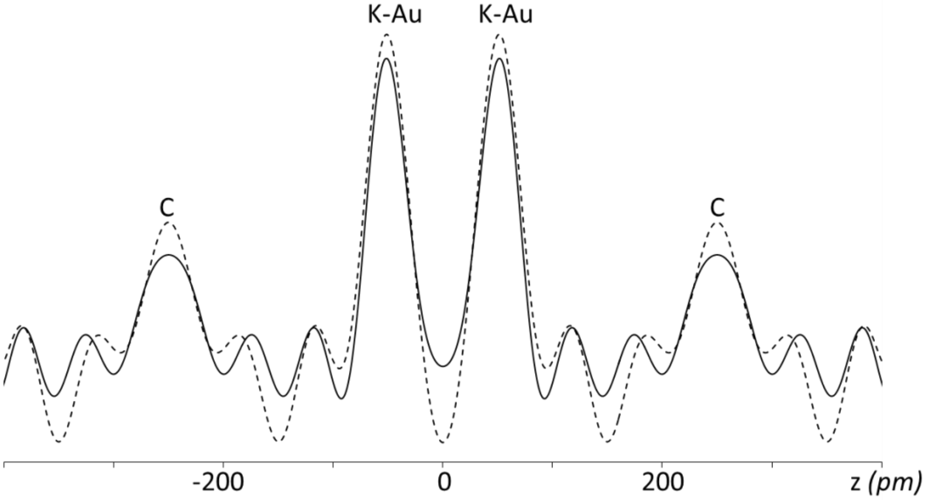

Figure 8: Experimental (dotted line) and calculated 1D electronic density profiles along the *c*-axis for the α-GIC. (R_F = 22%).

Table 4: Experimental and calculated intensities and structure factors of the 00*l* reflections of the α-GIC.

00*l*	θ (° *Mo* $K_{\alpha1}$)	d_{00l} (*pm*)	I_{00l} **exp**	I_{00l} **calc**	F_{00l} **exp**	F_{00l} **calc**
1	4.07	500	100	100	73	73
2	8.15	250	47.22	43.71	72	69
3	12.28	167	59.78	17.86	-100	-55
4	16.48	125	31.15	15.17	-86	-60
5	20.77	100	28.64	31.75	-95	-100
6	25.19	83	9.90	6.18	-63	-50
7	29.77	71	5.17	2.29	-51	-34
8	34.57	62	2.4	1.69	38	32
9	39.67	55	0.6	2.08	20	38

Table 5: The atomic amount and position of each species in the α-GIC.

Element	**C**	**K**	**Au**	**Au**	**K**	**C**
Position (*pm*)	-250	-50	-50	50	50	250
Atomic content	2	0.515	0.34	0.34	0.515	2

Thanks to these X-ray studies, it was possible to have a right approach of the chemical composition of these different compounds. To confirm these chemical formulas and to probe the homogeneity of the samples, ion beam analyses have been performed using nuclear microprobe.

3.4 *Chemical study of the gold-based GIC by ion beam analysis*

The nuclear microprobe analysis (NMA) appears as a powerful tool for analyzing graphite intercalation compounds. Unlike other techniques (as elementary analysis, EDX spectroscopy, thermo-gravimetric measurement or atomic absorption spectroscopy), it allows to simultaneously titrate all the elements contained in a same sample. The γ-GIC and α-GIC have been analyzed by this technique, the analysis of the β-GIC being not possible since it is not isolated.

3.4.1 *γ-GIC*

Several areas of each sample have been probed and several spectra have been recorded. After normalization of the spectra to the same number of incident ions, all of them can be superimposed (Figure 9), meaning a good reproducibility of the experiments and the homogeneity of the bulk GIC.

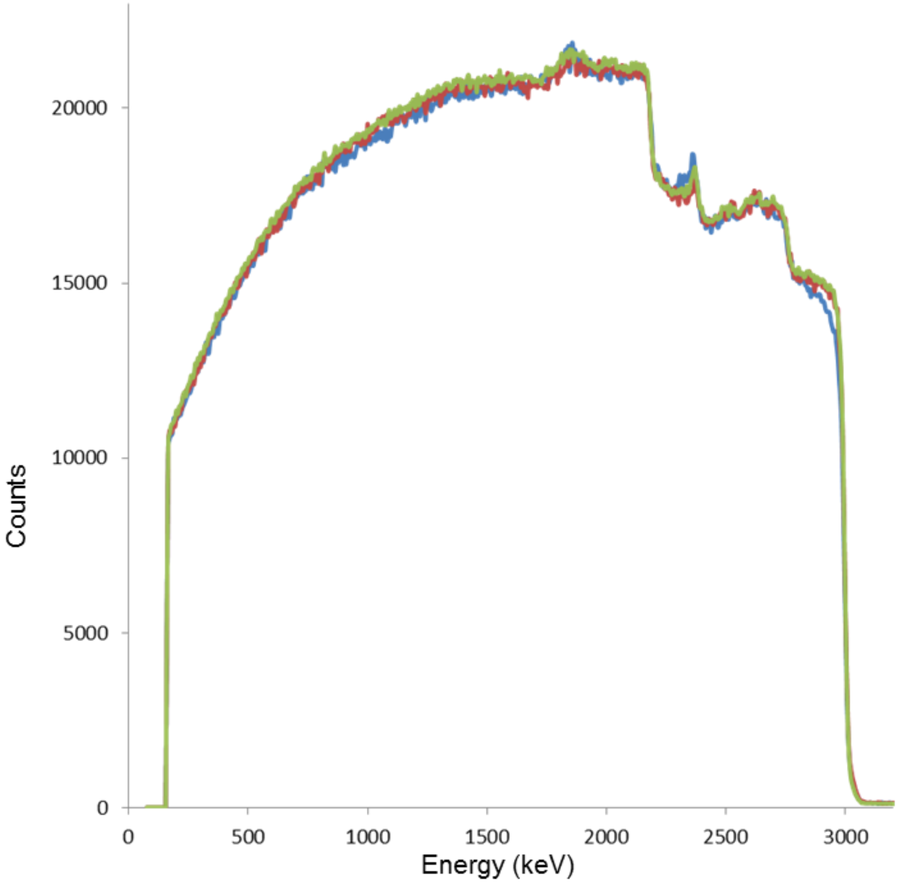

Figure 9: Comparison of three normalized RBS spectra of the γ-GIC recorded by scanning the beam on large areas (200x200 µm²).

From these spectra, elemental distribution maps can be extracted. Figure 10 represents a spectrum and its simulation with the contribution of each element.

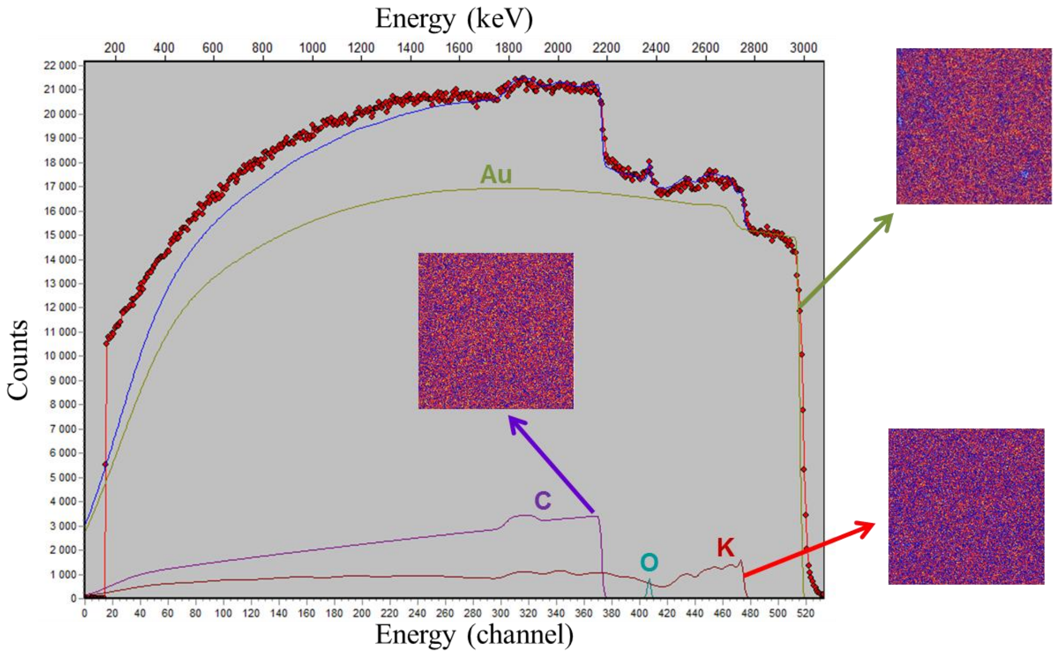

Figure 10: Recorded RBS spectrum (red dots) of the γ-GIC and its simulation (blue line) from [20, 21]. Each individual elementary contribution to the simulation is drawn. Square maps represent intensity contrasts within narrow region of interest in order to enhance K, Au and C contributions.

The individual contributions of the elements confirm the homogeneity of the samples in depth and laterally. This simulation allowed to determine a chemical formula corresponding to $K_{1.3}Au_{1.5}C_4$ for the bulk. The formula obtained by this technique is in agreement with the one deduced from crystallographic data. So, we can conclude that the γ-GIC synthesized using the solid-liquid method is a homogenous ternary compound which chemical formula is $K_{1.3}Au_{1.5}C_4$.

3.4.2 *α-GIC*

The α-GIC has also been analyzed by nuclear microprobe to confirm the chemical formula determined using crystallographic data and to probe the homogeneity of the compound. This phase is metastable and the synthesis conditions are more difficult to reproduce since the experimental parameters are very close to a solid-liquid biphasic domain in the Au-K phase diagram. Consequently, the analysis has been more complicated than for the previous compound. Indeed, this compound is very often obtained in the presence of a more or less significant amount of γ-GIC evidenced by X-ray diffraction. However, the measurements will give an approximate formula of this compound.

Multiple zones of various samples have been probed. As shown Figure 11 in the individual elementary contribution of gold, the samples are less homogeneous than those of the γ-GIC. In addition, the spectra for these maps are not always all superimposed, indicating the presence of in-depth heterogeneities.

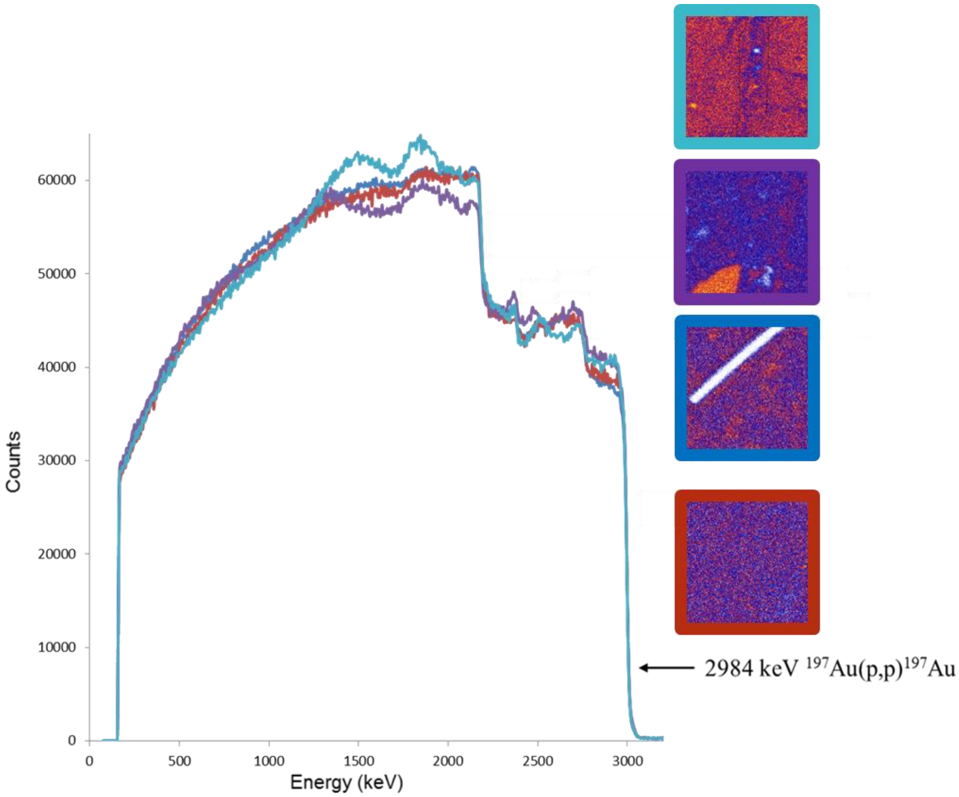

Figure 11: Comparison of four normalized RBS spectra of the α-GIC recorded by scanning the beam on large areas (200x200 µm²). The RBS spectra and their associated individual gold map are drawn with identical colors (one experimental spectrum corresponding to one mapframe).

From this individual elementary contribution, it is possible to select the desired fraction and generate a new spectrum to eliminate a possible overconcentration or any faults. After a selection of the data, a simulation was carried out (Figure 12).

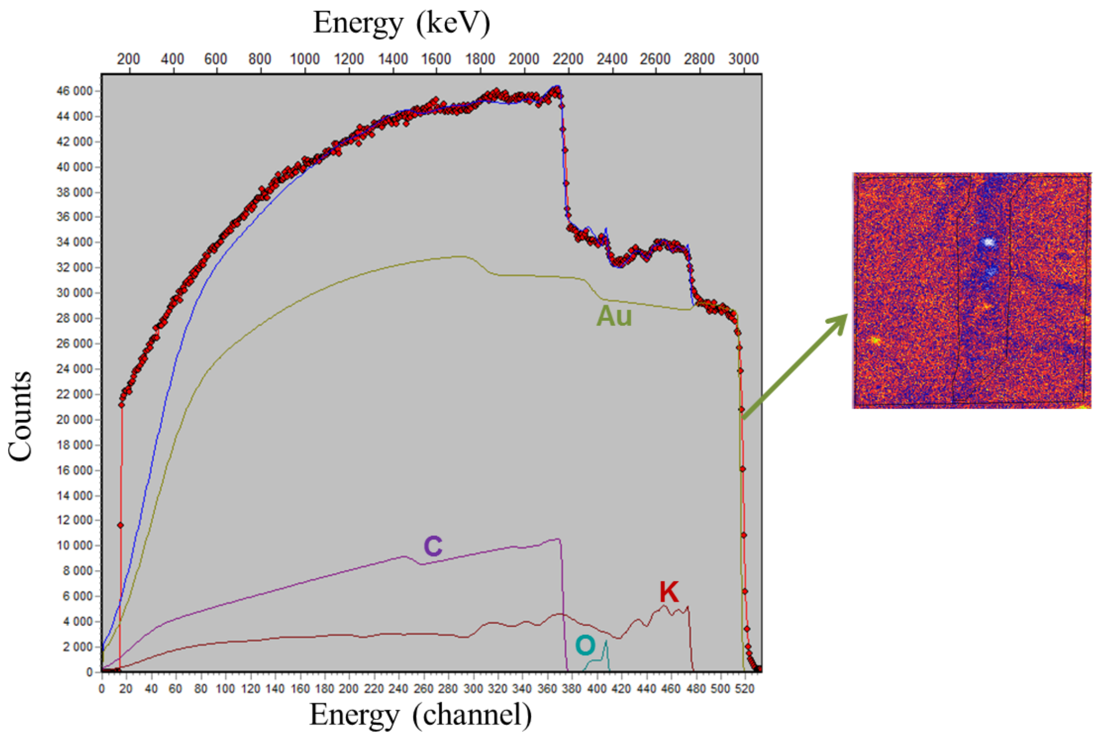

Figure 12: Recorded RBS spectrum (red dots) of the α-GIC and its simulation (blue line). Each individual elementary contribution to the simulation is drawn.

This sample is oxidized on the surface but the bulk of the sample corresponds to $K_{1.16}Au_{0.80}C_4$. The amount of metal in this formula is probably slightly overevaluated, this overestimation corresponding to the presence of a small amount of the γ-GIC.

Conclusion

Performing the extensive study of the graphite-potassium-gold system, this work shows that we have been able to synthesize three novel first stage ternary compounds in this system. Even if the the element associated with potassium is strongly electronegative, the obtaining of several ternary first stage GIC is usual in the case of donor type compounds. The first compound (α-GIC) exhibits a repeat distance of 500 pm, mixed Au-K two-layered intercalated sheets and a chemical composition corresponding to $K_{1.0}Au_{0.7}C_{4.}$ It is obtained for a high temperature and with a high amount of gold in the alloy; this compound reveals a metastable behavior. The second compound (β-GIC) is an intermediate phase which chemical formula is $K_{1.6}Au_{0.7}C_4$: its repeat distance reaches 960 pm and it possesses three-layered K-Au-K intercalated sheets. It has been evidenced during the study of the formation mechanism of the third compound. The exact experimental conditions leading to the pure phase still remain under investigation. The third compound (γ-GIC) has been easily isolated and it is reproducibly synthesized in a large range of temperature and reactive alloy composition. Its chemical formula is $K_{1.3}Au_{1.5}C_4$ with a repeat distance of 1311 pm, in agreement with five-layered K-Au-Au-Au-K intercalated sheets. This system is remarkable since it simultaneously presents the ternary GIC with the smallest and the highest repeat distances (dilations of 49% and 291% respectively).

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Table 1: Optimal composition of alloy allowing the synthesis of first stage ternary GIC in the graphite-potassium-gold system.

Compound	**Alloy composition** *(Au at.%)*	**T** (°*C*)	**Reaction time** (*days*)
α	50	650	2
β	20 – 50	Not isolated	
γ	34	440	8

Table 2: Experimental and calculated intensities and structure factors of the 00*l* reflexions of the β-GIC.

00*l*	**θ** (°,(λMoK$_{\alpha1}$))	**d$_{00l}$** *(pm)*	**I$_{00l}$ exp**	**I$_{00l}$ calc**	**F$_{00l}$ exp**	**F$_{00l}$ calc**
1	2.13	953	100	100	59	59
2	4.27	476	50.60	86.62	60	78
3	6.41	318	7.09	6.56	28	26
4	8.56	238	68.09	69.38	100	100
5	10.72	191	23.17	22.91	66	65
6	12.90	159	27.25	27.86	79	79
7	15.10	136	8.52	6.46	48	42
8	17.32	119	13.23	12.63	65	63

Table 3: The atomic amount and position of each species in the β-GIC.

Element	**C**	**K**	**K**	**Au**	**K**	**K**	**C**
Position (*pm*)	-476.5	-230	-170	0	170	230	476.5
Atomic content	2	0.34	0.46	0.70	0.46	0.34	2

Table 4: Experimental and calculated intensities and structure factors of the 00*l* reflexions of the α-GIC.

00*l*	θ *(° Mo $K_{\alpha 1}$)*	**d_{00l}** *(pm)*	**I_{00l} exp**	**I_{00l} calc**	**F_{00l} exp**	**F_{00l} calc**
1	4.07	500	100	100	73	73
2	8.15	250	47.22	43.71	72	69
3	12.28	167	59.78	17.86	-100	-55
4	16.48	125	31.15	15.17	-86	-60
5	20.77	100	28.64	31.75	-95	-100
6	25.19	83	9.90	6.18	-63	-50
7	29.77	71	5.17	2.29	-51	-34
8	34.57	62	2.4	1.69	38	32
9	39.67	55	0.6	2.08	20	38

Table 5: The atomic amount and position of each species in the α-GIC.

Element	**C**	**K**	**Au**	**Au**	**K**	**C**
Position (*pm*)	-250	-50	-50	50	50	250
Atomic content	2	0.515	0.34	0.34	0.515	2