This version of the Electronic Supporting Information published 19 February 2024 replaces the original version published 14 February 2024. This version contains a higher quality version of the Syriac text on page 17.

Electronic Supporting information

"*What makes every work perfect is cooking and grinding*": the ancient roots of mechanochemistry

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Characterization of the reagents

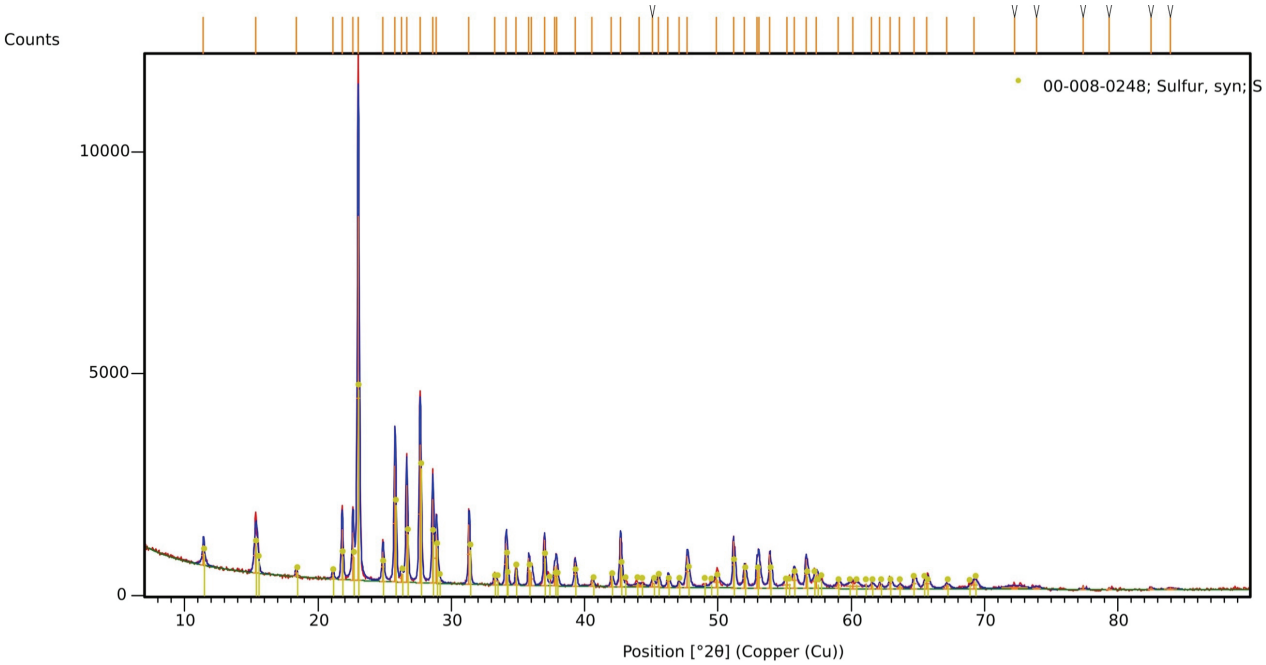


Figure S1. XRPD pattern of sulphur used for the synthesis of cinnabar. Phase identification was performed using the PDF 2 Release 2004 database.

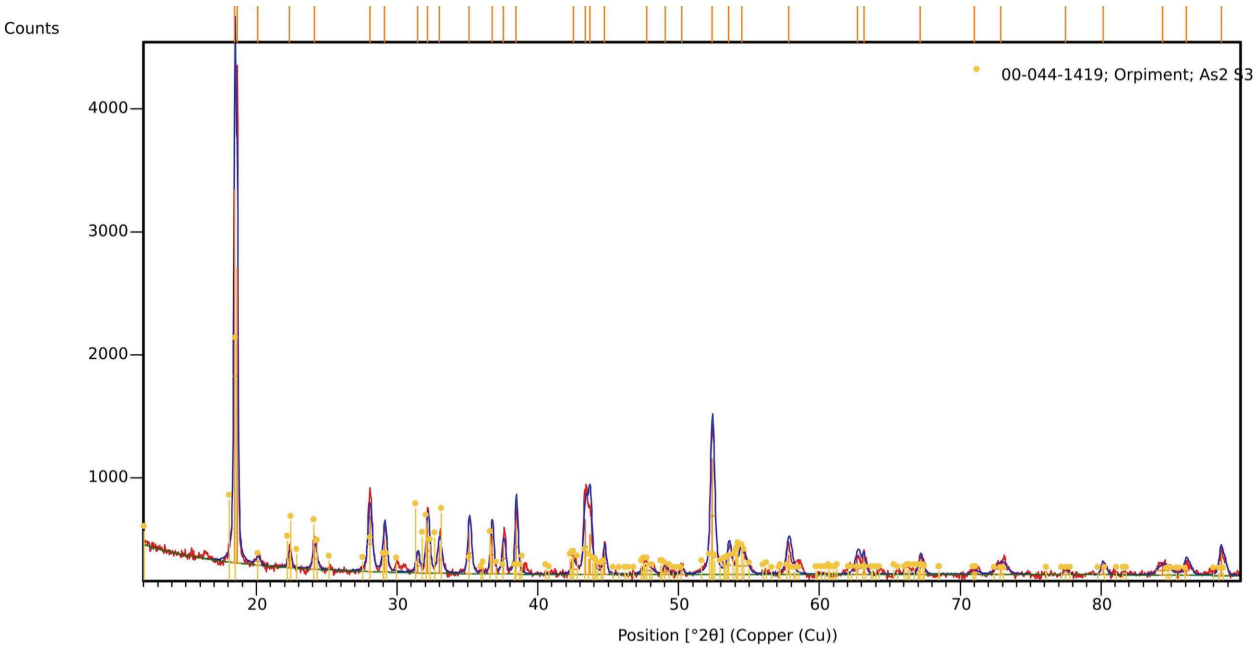


Figure S2. XRPD pattern of mineral orpiment used for the synthesis of cinnabar. Phase identification was performed using the PDF 2 Release 2004 database.

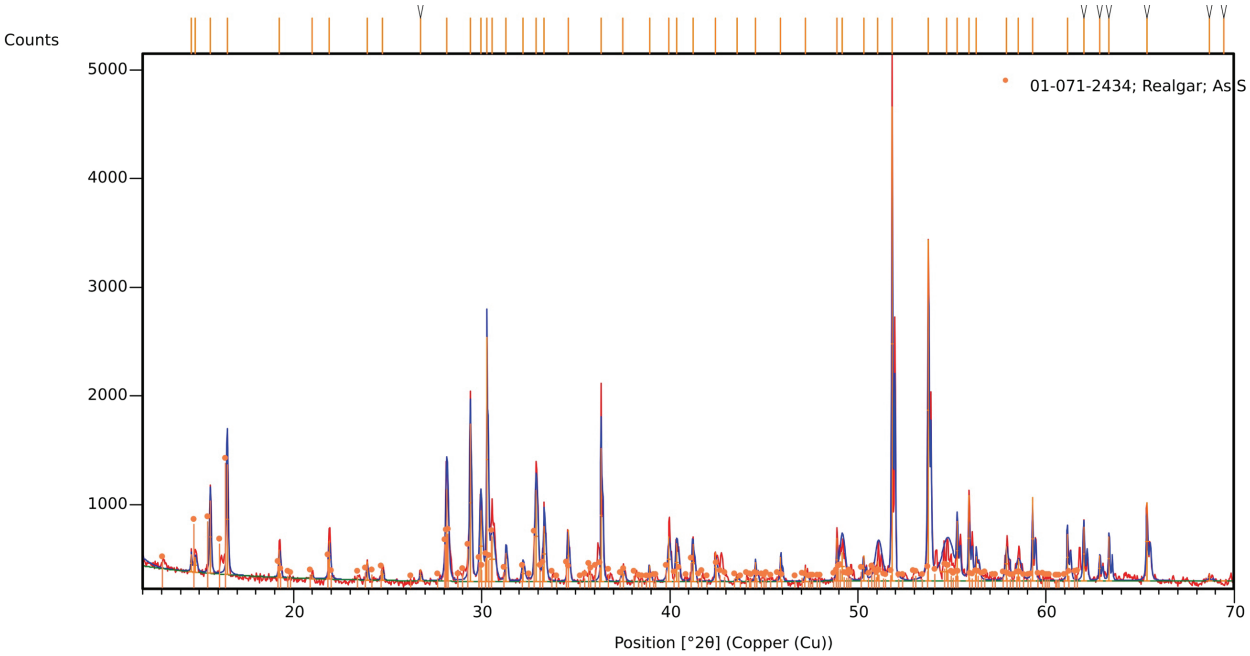


Figure S3. XRPD pattern of mineral realgar used for the synthesis of cinnabar. Phase identification was performed using the PDF 2 Release 2004 database.

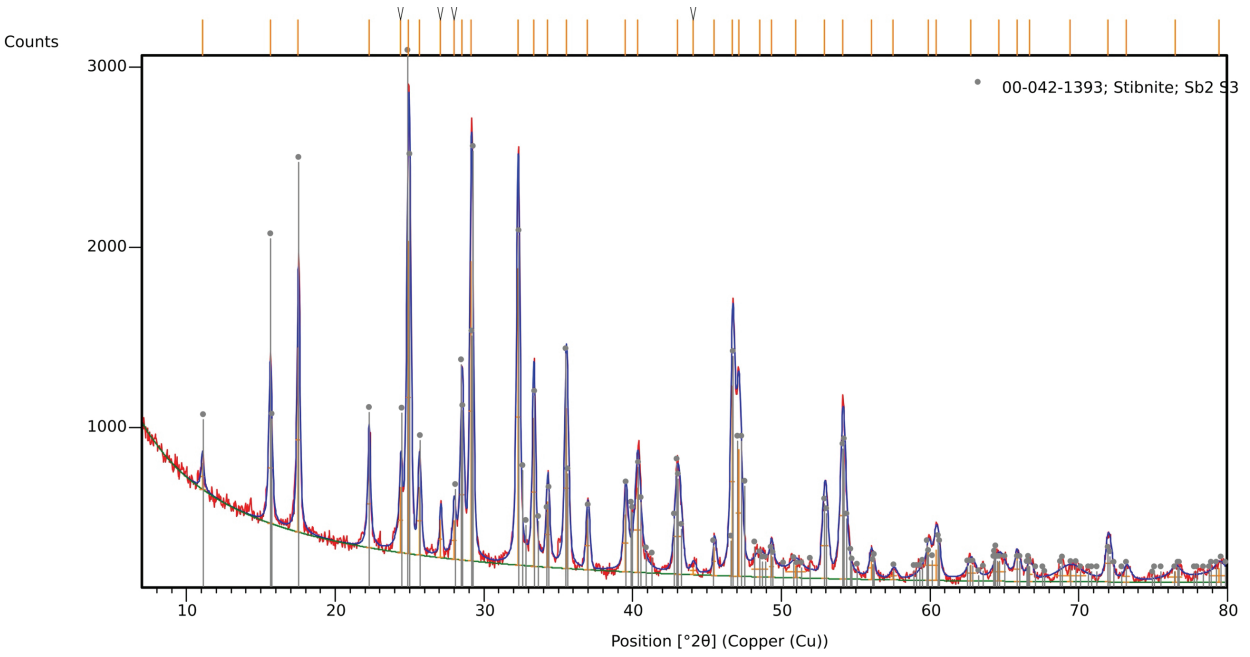


Figure S4. XRPD pattern of mineral stibnite used for the synthesis of cinnabar. Phase identification was performed using the PDF 2 Release 2004 database.

Synthesis of the cinnabar form sulphur and mercury

700 mg of mercury were ground with 350 mg of sulphur (S_8) which correspond to a molar ratio 3:1 S:Hg in a 1-mL steel jars that contained three steel spheres (Ø = 3 mm, 0.545 g) at 25 Hz. The reaction mixture was ground for six hours and then heated at 350°C for 24 hours. For the same batch, XRPD pattern was collected after three hours of milling, six hours of milling and after the thermal treatment (Figure S6).

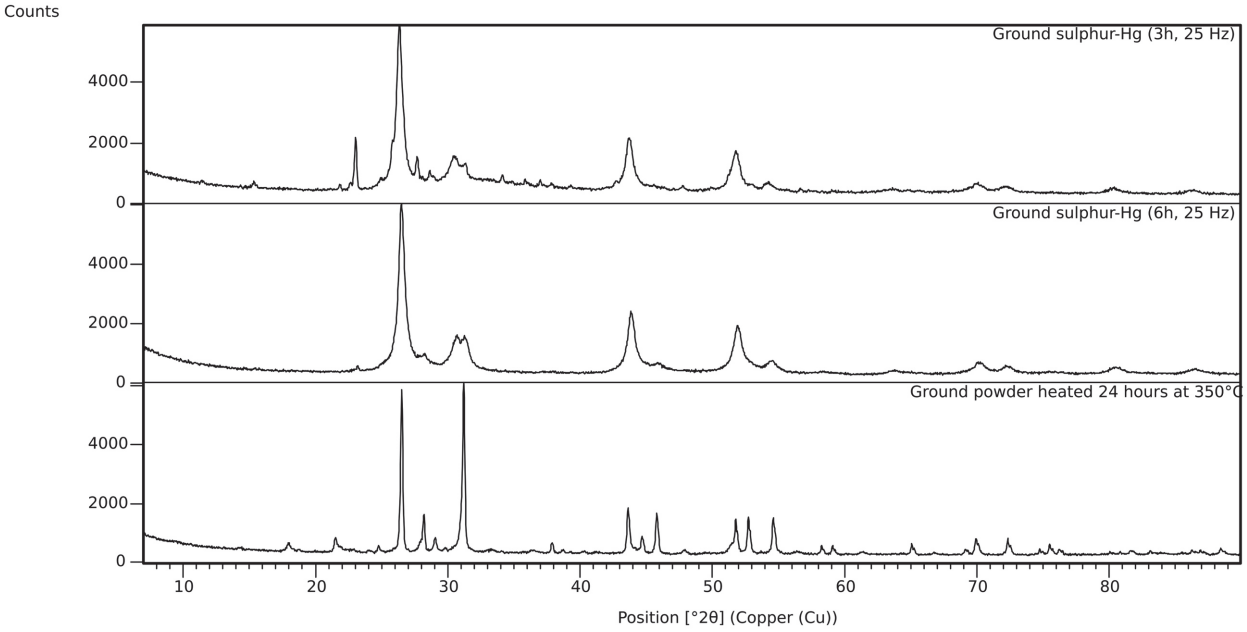


Figure S5. Comparison between XRPD pattern of ground powder obtained by milling sulphur and mercury three hours at 25Hz (up), six hours at 25Hz (middle) and after heating at 350°C (bottom).

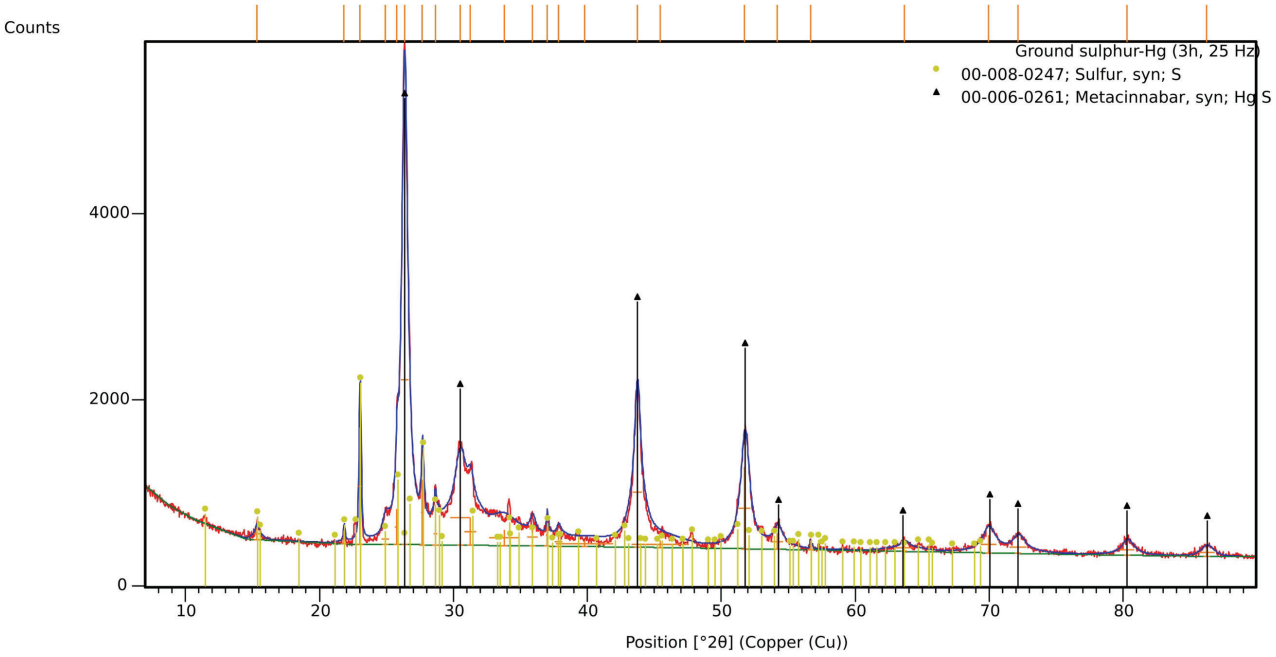


Figure S6. XRPD pattern of the ground powder obtained upon grinding sulphur and mercury for three hours at 25Hz which corresponds to a mixture of sulphur (yellow lines) and metacinnabar (black lines) and unreacted mercury (broad bump at θ=~ 30°). Phase identification was performed using the PDF 2 Release 2004 database.

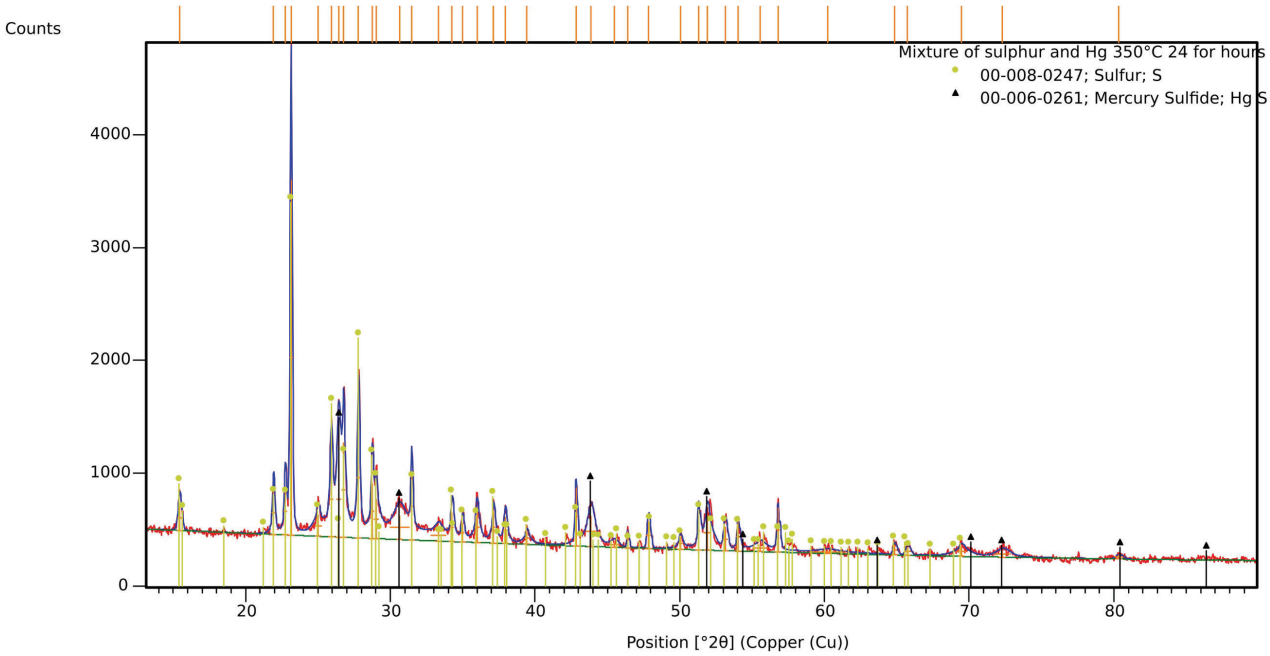


Figure S7. XRPD pattern of the residual powder obtained upon heating for 24 hours at 350°C a mixture of sulphur (yellow lines) and metacinnabar (black lines). Phase identification was performed using the PDF 2 Release 2004 database.

Figure S8.The custom-made furnace used for the synthesis of cinnabar. A thermocouple was used for monitoring the temperature.

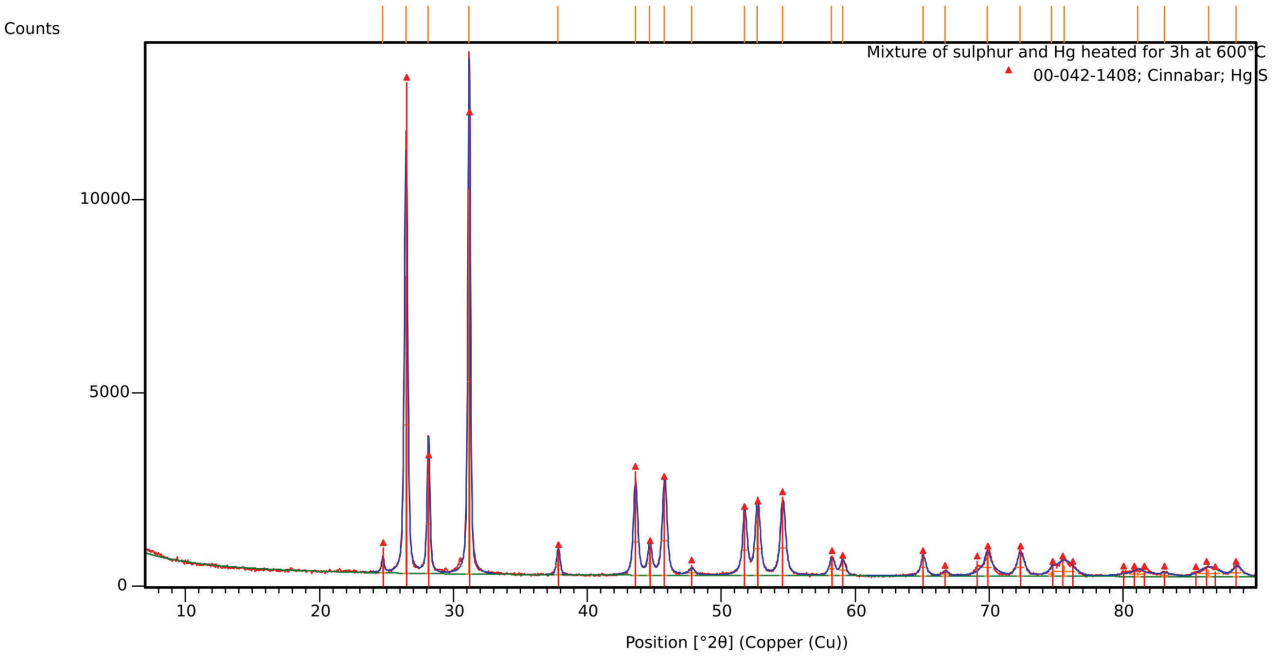


Figure S9. XRPD pattern of the red powder obtained by heating the mixture of sulphur and mercury for three hours at 600°C. Phase identification was performed using the PDF 2 Release 2004 database.

Synthesis of cinnabar from mercury and orpiment

700 mg of mercury were ground with 350 mg of orpiment (As_2S_3) which correspond to a molar sulphur:Hg 1.1:1 in a 1-mL steel jars that contained three steel spheres (Ø = 3 mm, 0.545 g) at 25 Hz.The reaction mixture was ground for six hours and then heated at 350°C for three hours. For the same batch, XRPD pattern was collected after three hours of milling, six hours of milling and after the thermal treatment (Figure S10).

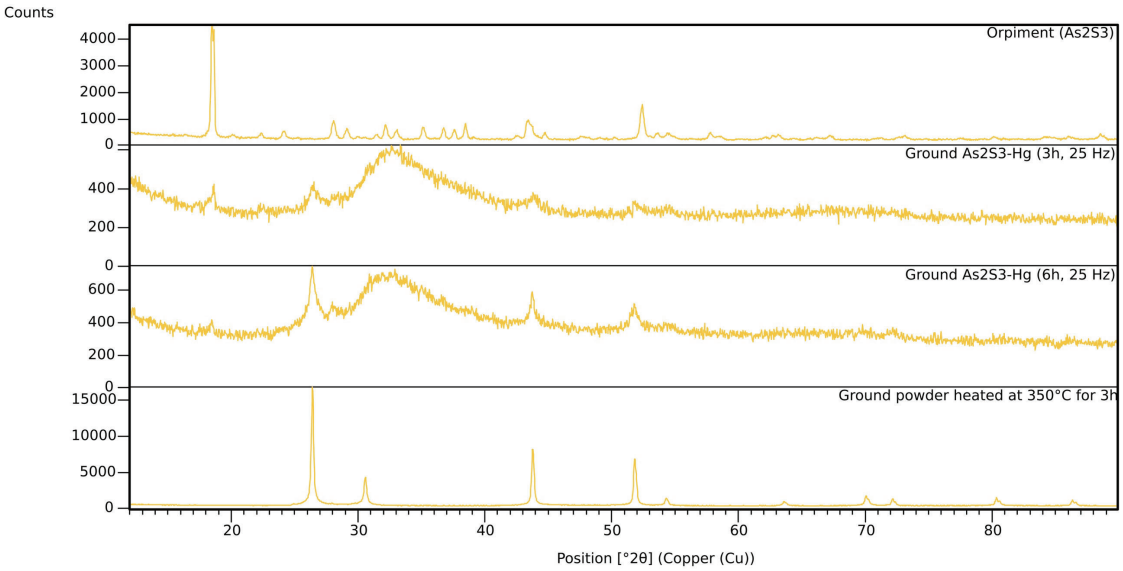


Figure S10. Comparison between XRPD pattern of reagent orpiment As_2S_3 (up), of the ground powder obtained from Hg and As_2S_3 (middle) and of the ground powder heating at 350°C (bottom).

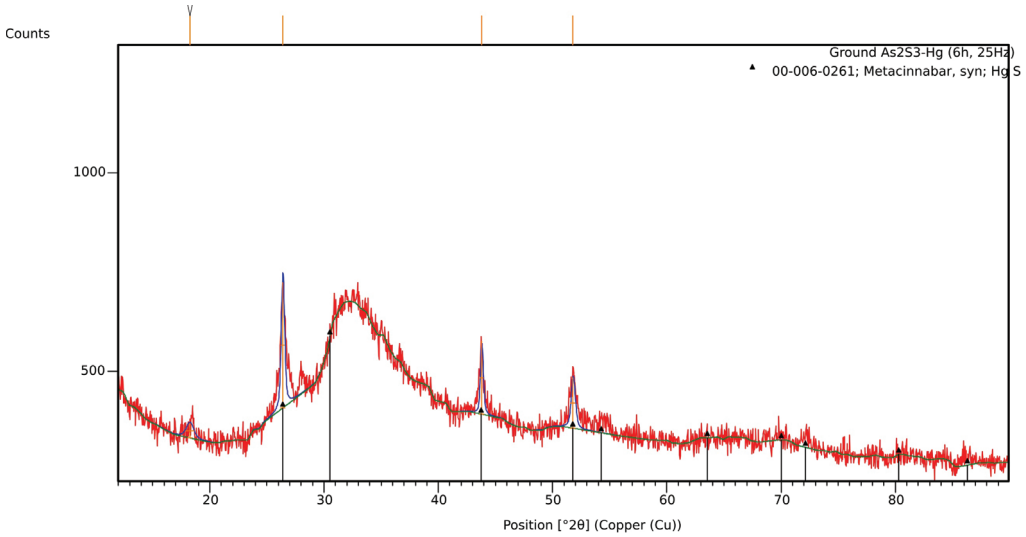


Figure S11. XRPD pattern of the ground powder obtained upon grinding orpiment and mercury for six hours at 25Hz which corresponds to a mixture of metacinnabar (black lines) and amorphous phases (broad bump at θ=~30°). Phase identification was performed using the PDF 2 Release 2004 database.

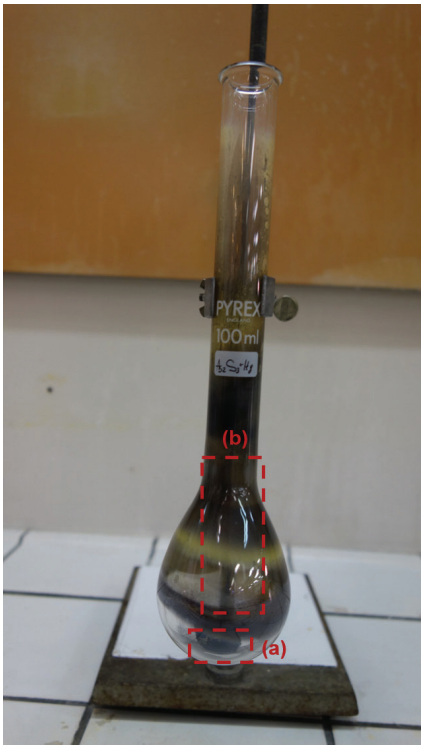


Figure S12. Kjeldahl flask used during the heating process. The rectangular figures indicate the two different samples of powder analyzed.

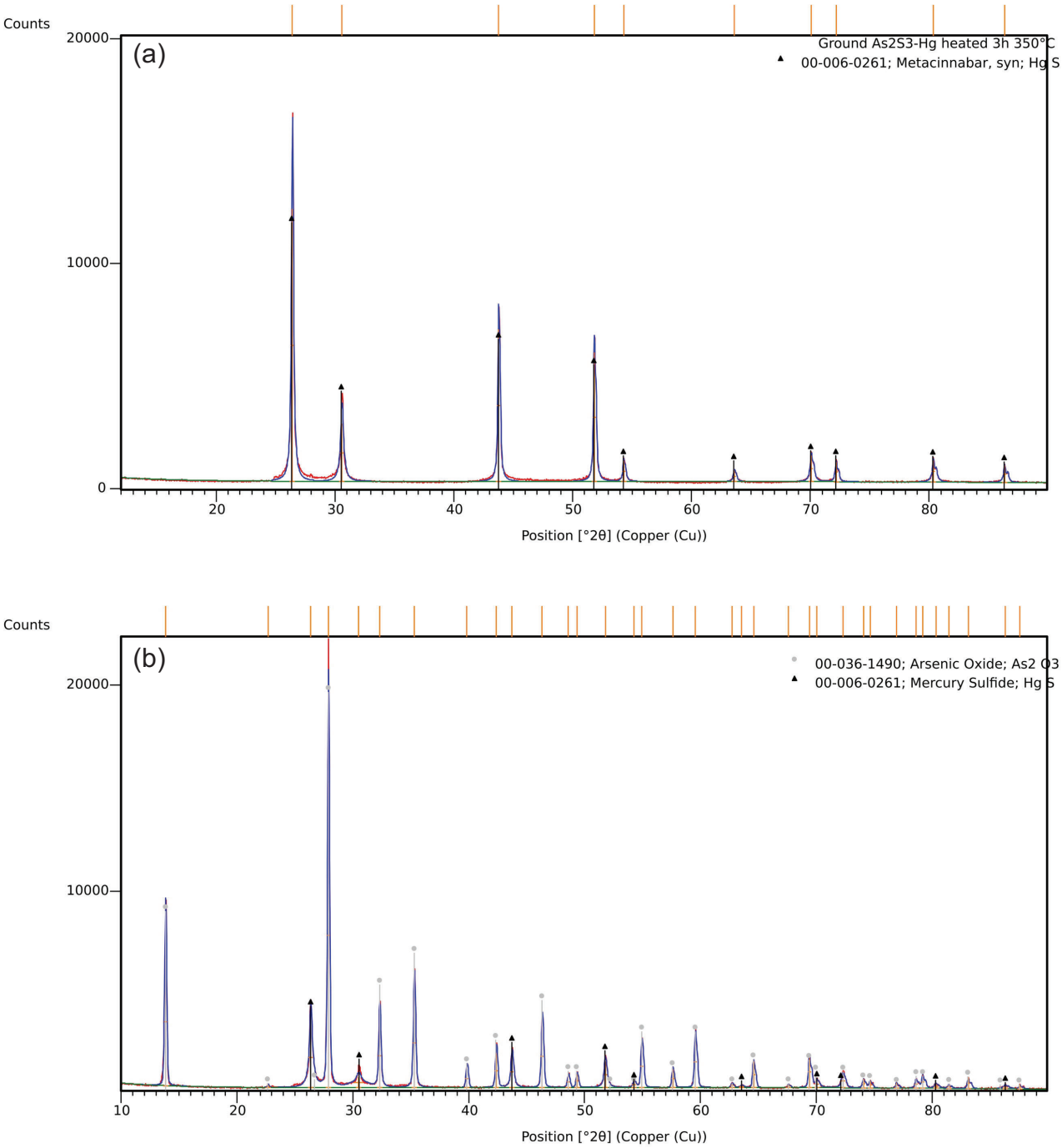


Figure S13. XRPD pattern of the residual powder (a. bottom of the flask; b. scratched from the glassware) obtained upon grinding As_2S_3 with Hg (three hours at 25Hz) and heating the so-obtained powder at 350°C. Phase identification was performed using the PDF 2 Release 2004 database.

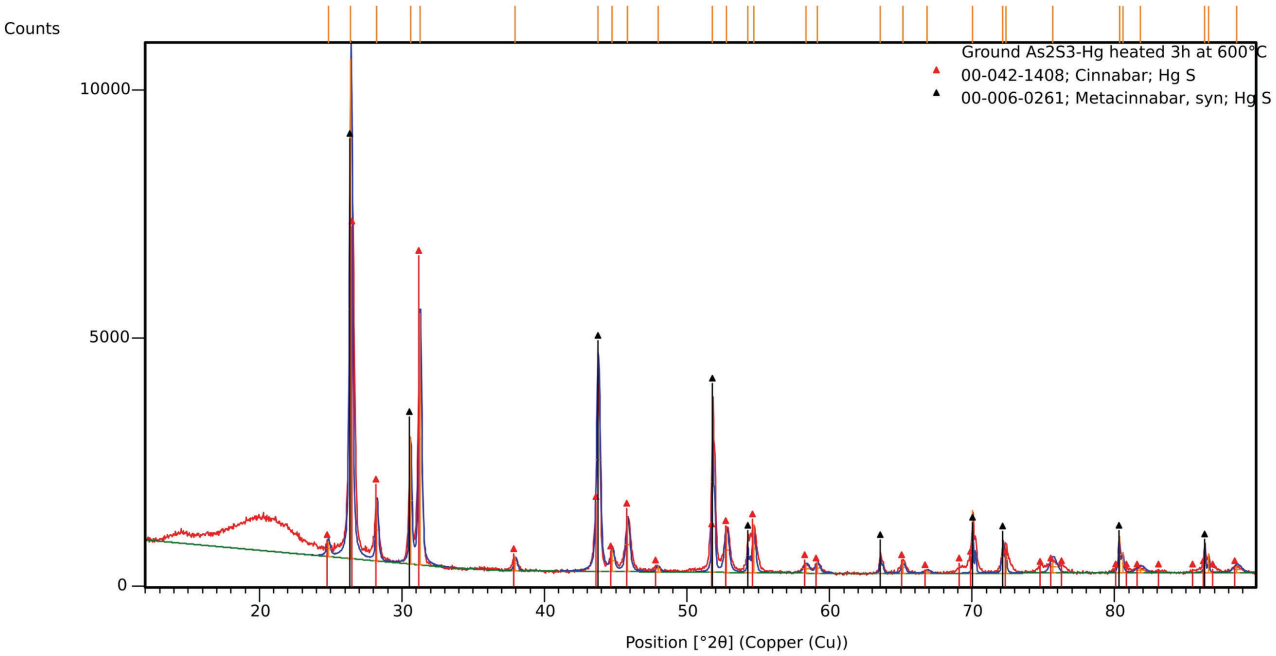


Figure S14. XRPD pattern of the residual powder obtained upon grinding As_2S_3 with Hg (three hours at 25Hz) and heating the so-obtained powder at 600°C. Phase identification was performed using the PDF 2 Release 2004 database.

Synthesis of cinnabar from mercury and realgar

700 mg of mercury were ground with 350 mg of realgar (As_4S_4) which correspond to a molar sulphur:Hg 1:1.1 in a 1-mL steel jars that contained three steel spheres (Ø = 3 mm, 0.545 g) at 25 Hz.The reaction mixture was ground for six hours and then heated at 350°C for three hours. For the same batch, XRPD pattern was collected after three hours of milling, six hours of milling and after the thermal treatment (Figure S15).

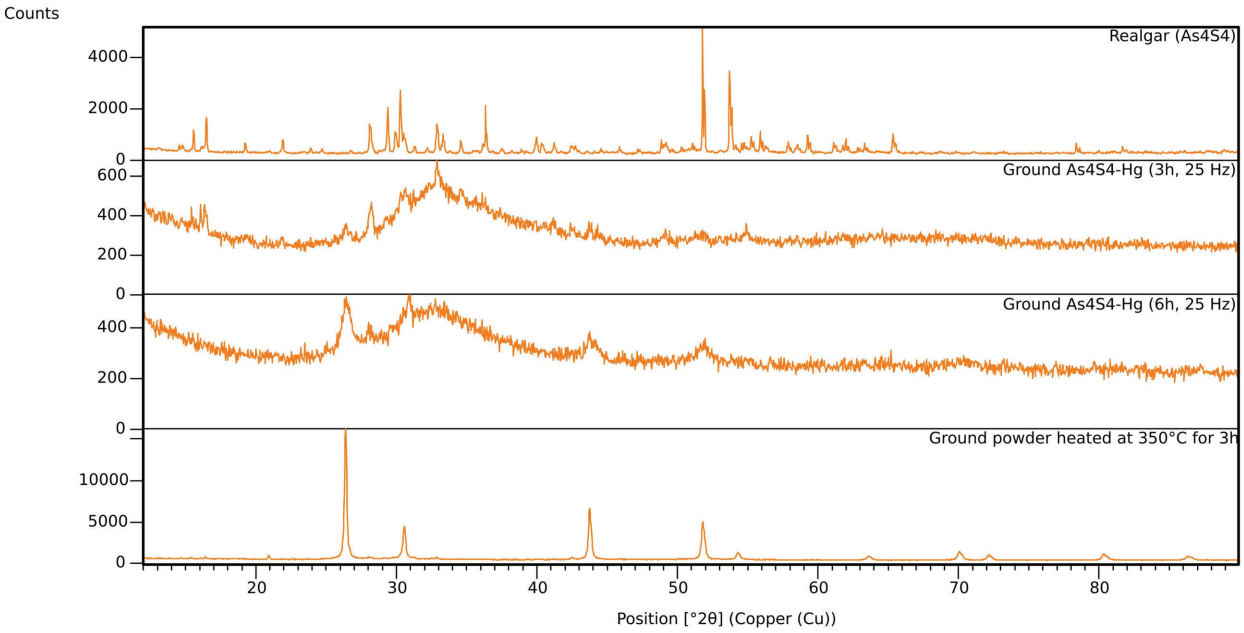


Figure S15. Comparison between XRPD pattern of reagent orpiment As_4S_4 (up), of the ground powder obtained from Hg and As_4S3 (middle) and of the ground powder heating at 350°C (bottom).

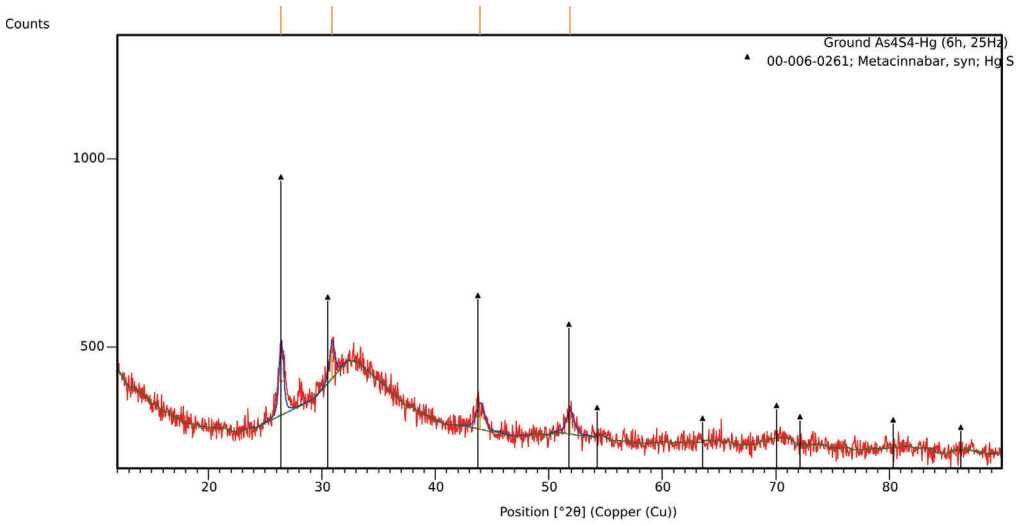


Figure S16. XRPD pattern of the ground powder obtained upon grinding realgar and mercury for six hours at 25Hz which corresponds to a mixture of metacinnabar (black lines) and amorphous phases (broad bump at θ=~30°). Phase identification was performed using the PDF 2 Release 2004 database.

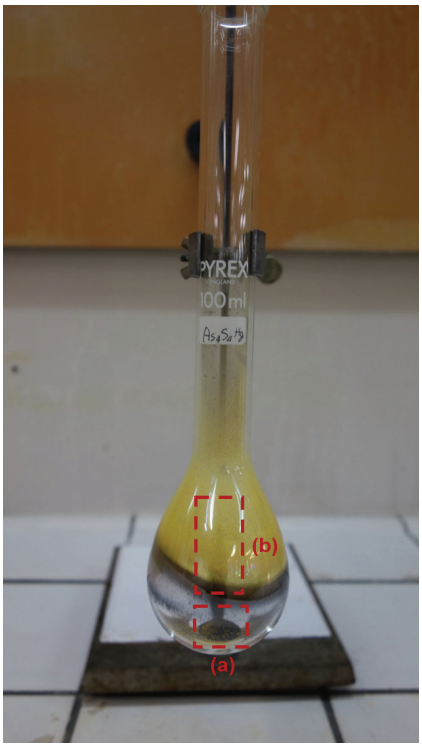


Figure S17. Kjeldahl flask used during the heating process. The rectangular figures indicate the two different samples of powder analyzed.

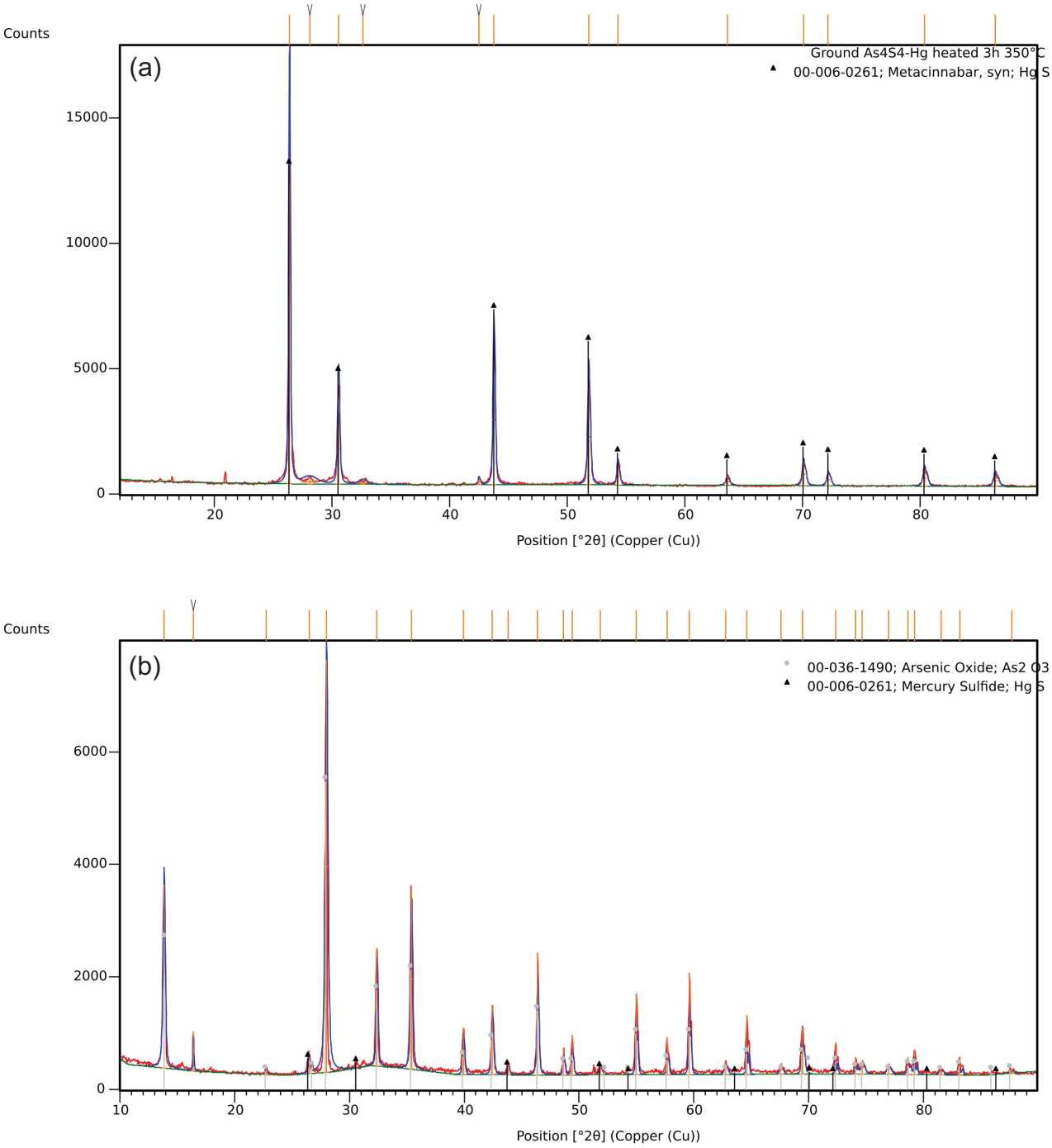


Figure S18. XRPD pattern of the residual powder (a. bottom of the flask; b. scratched from the upper part of the Kjeldahl flask) obtained upon grinding As_4S_4 with Hg (three hours at 25Hz) and heating the so-obtained powder at 350°C. Phase identification was performed using the PDF 2 Release 2004 database.

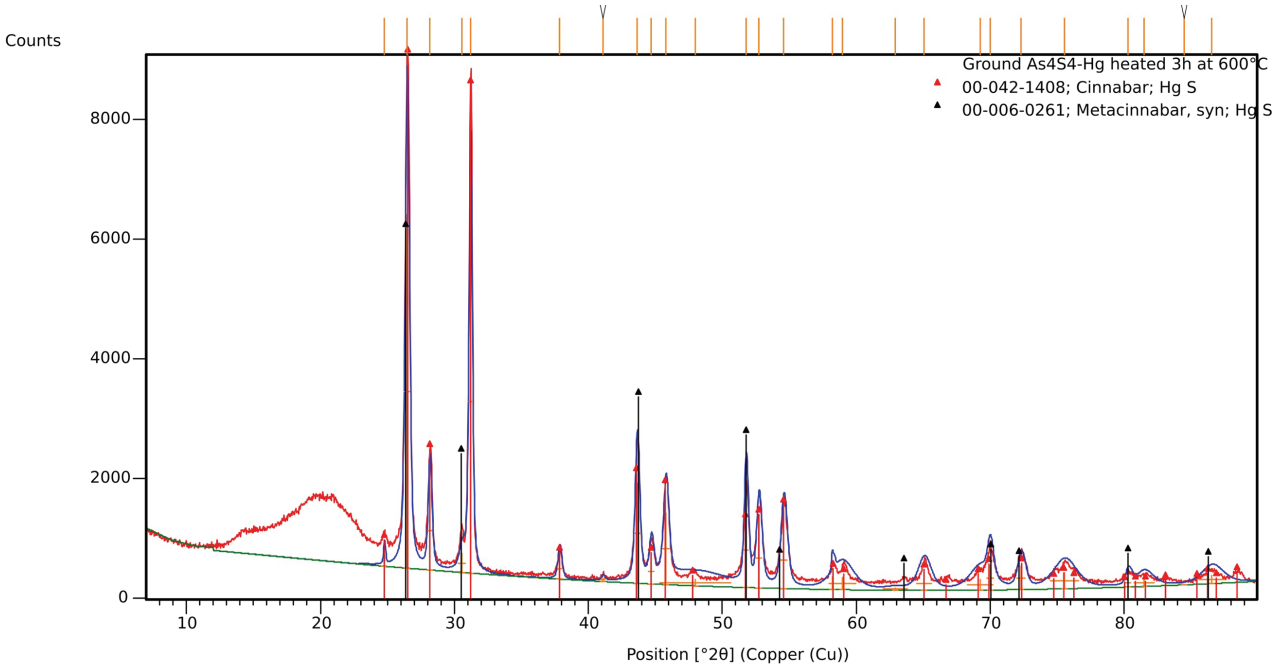


Figure S19. XRPD pattern of the residual powder obtained upon grinding As_4S_4 with Hg (three hours at 25Hz) and heating the so-obtained powder at 600°C. Phase identification was performed using the PDF 2 Release 2004 database.

Synthesis of cinnabar from mercury and stibnite

700 mg of mercury were ground with 350 mg of orpiment (Sb_2S_3) which correspond to a molar sulphur:Hg 0.9:1 in a 1-mL steel jars that contained three steel spheres (Ø = 3 mm, 0.545 g) at 25 Hz.The reaction mixture was ground for six hours and then heated at 350°C for three hours. For the same batch, XRPD pattern was collected after three hours of milling, six hours of milling and after the thermal treatment (Figure S21).

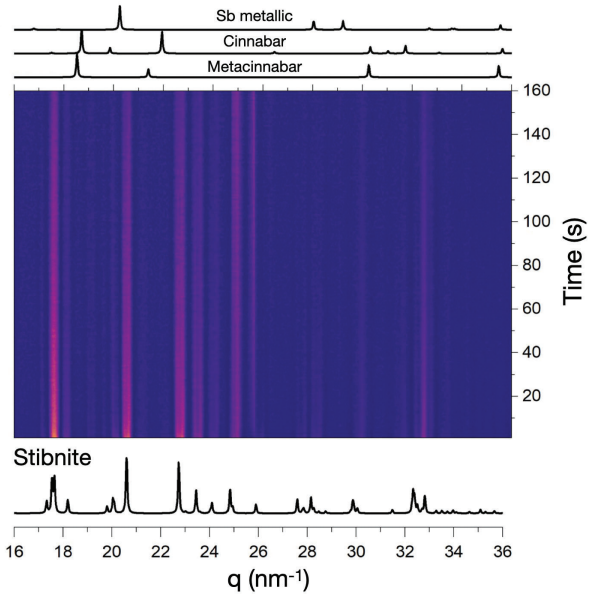


Figure S20. Time evolution of the milling reaction between mercury and stibnite. Milling conditions: 50 Hz for three hours with three 4 mm steel milling balls. No reactions occur.

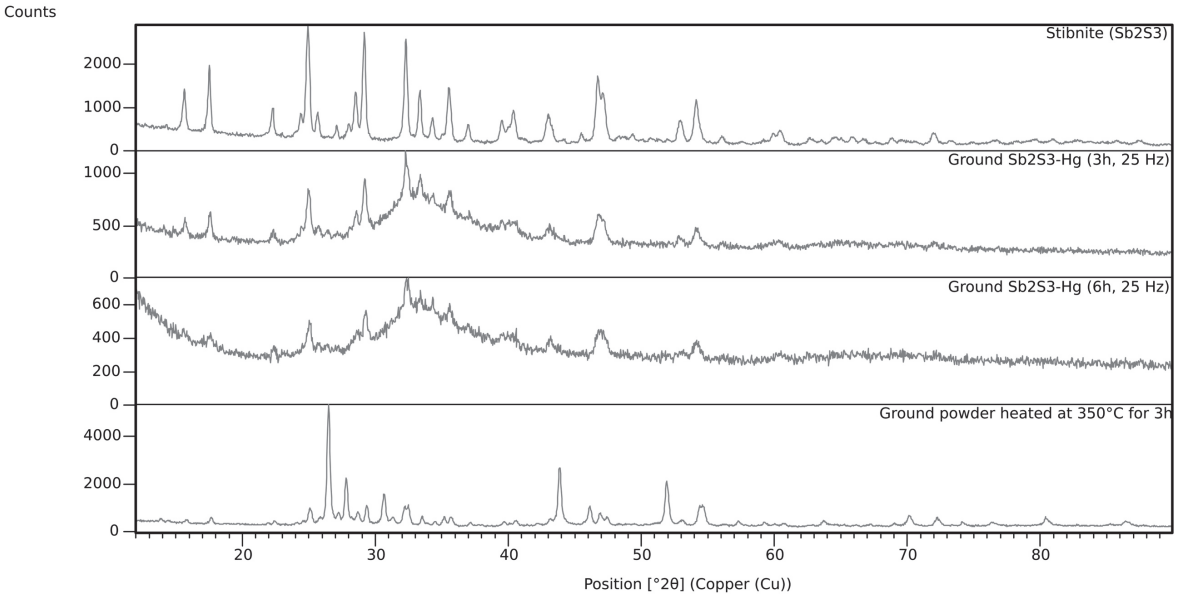


Figure S21. Comparison between XRPD pattern of reagent stibnite Sb_2S_3 (up), of the ground powder obtained from Hg and Sb_2S_3 (middle) and of the ground powder heating at 350°C (bottom). Phase identification was performed using the PDF 2 Release 2004 database.

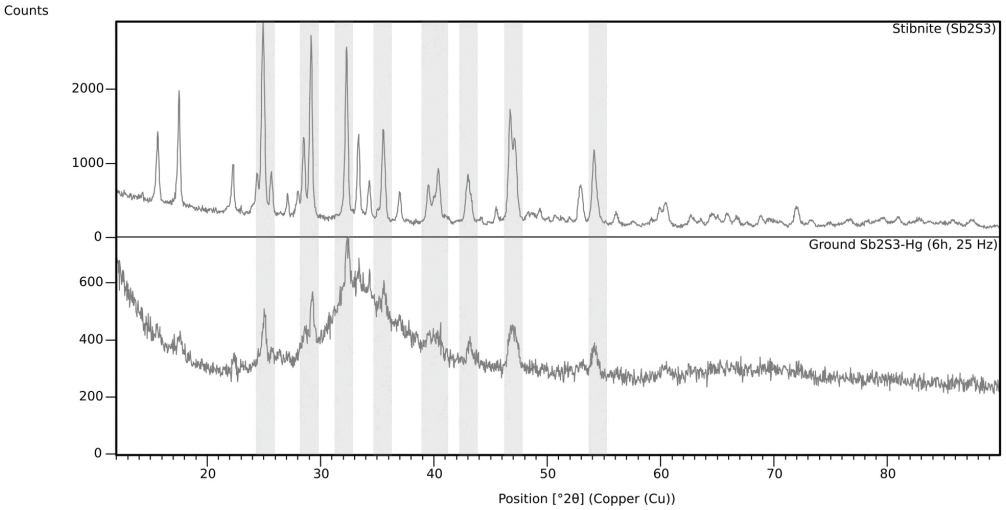


Figure S22. Comparison between XRPD pattern of the ground powder obtained upon grinding stibnite and mercury for six hours at 25Hz and mineral stibnite. The broad bump at θ=~ 30° can be ascribe to the presence of mercury.

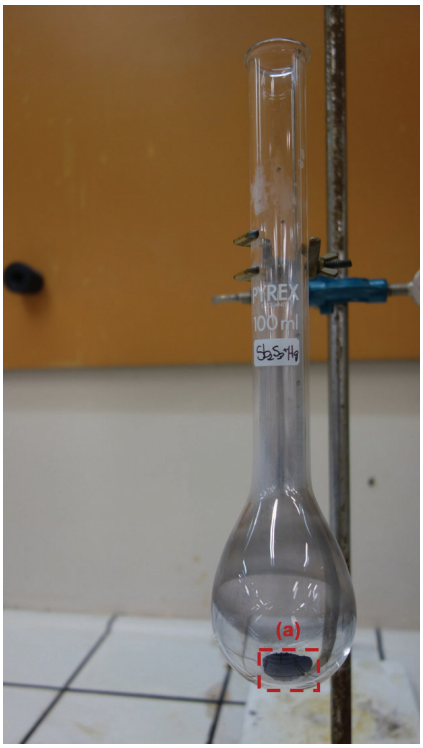


Figure S23. Kjeldahl flask used during the heating process. The rectangular indicates the sample of powder analyzed.

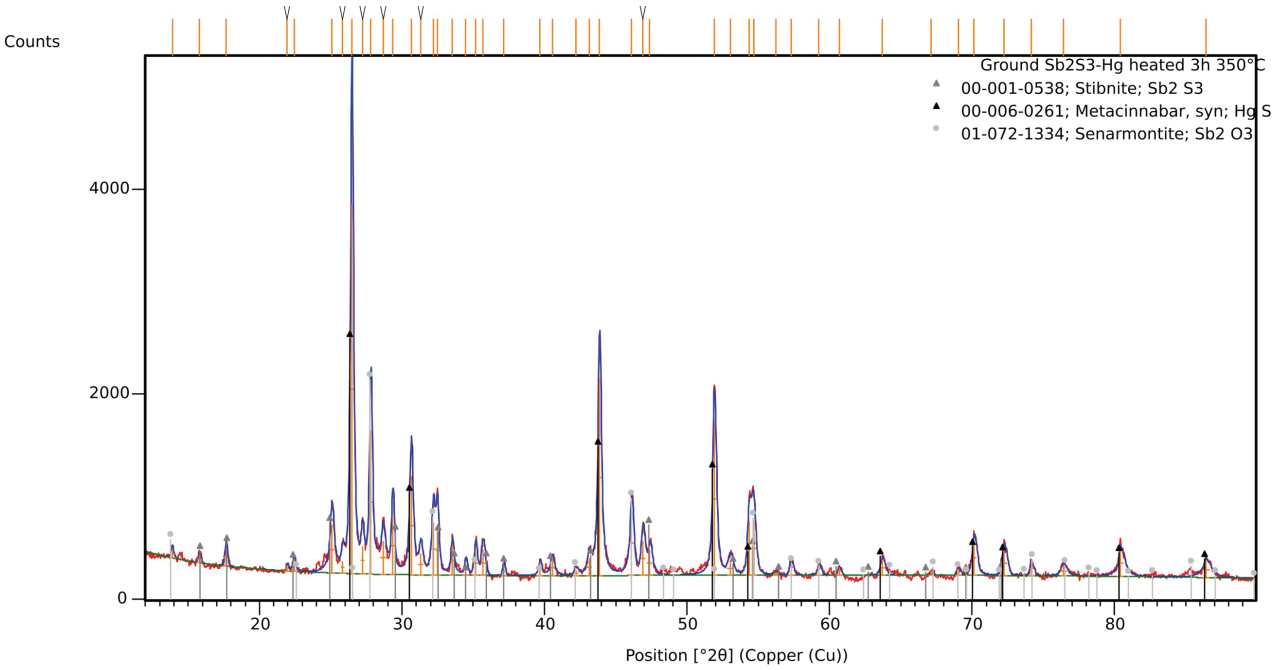


Figure S24. XRPD pattern of the residual powder obtained upon grinding Sb_2S_3 with Hg (three hours at 25Hz) and heating the so-obtained powder at 350°C. On the glassware condensed only unreacted mercury. Phase identification was performed using the PDF 2 Release 2004 database.

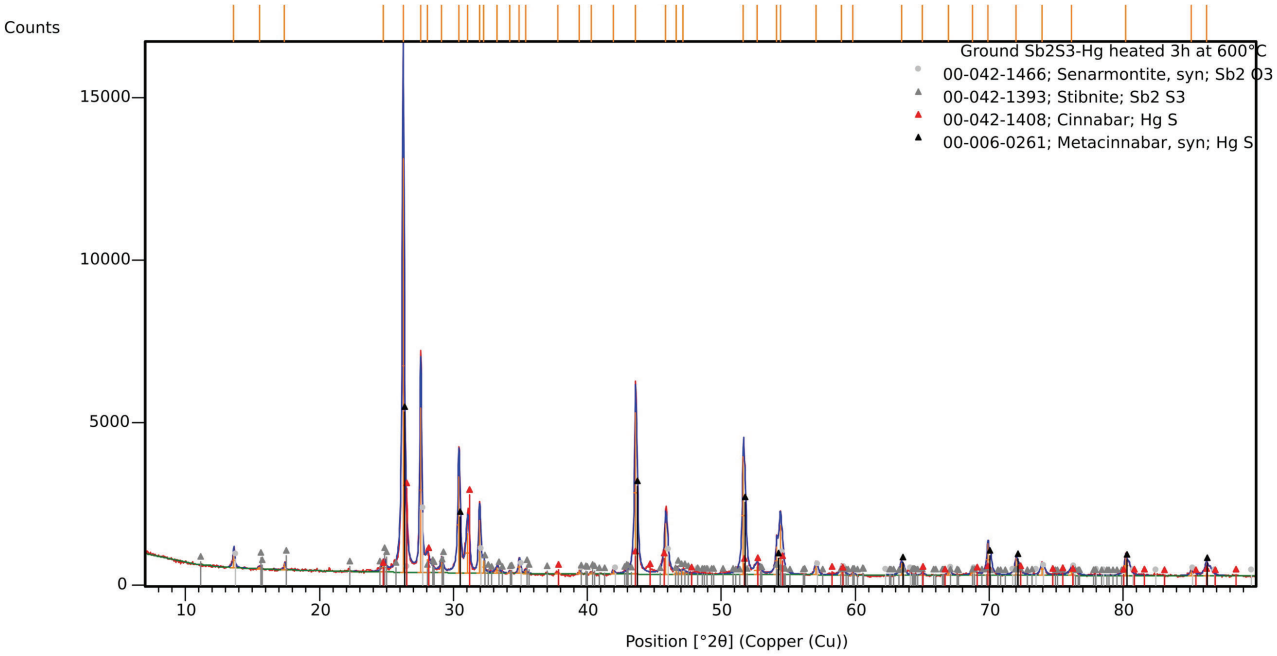


Figure S25. XRPD pattern of the residual powder obtained upon grinding Sb_2S_3 with Hg (three hours at 25Hz) and heating the so-obtained powder at 600°C. Phase identification was performed using the PDF 2 Release 2004 database.

Ancient Sources

Text 1: Anonymous Byzantine recipe. Edited by R. Halleux, *Traités des arts et métiers*, Paris 2021, p. 118.

Ποίησις κινναβάρεως. Δεῖ ἐμβαλεῖν εἰς θυίαν θείου ἀπύρου λίτραν α΄ καὶ ὑδραργύρου λίτρας β΄ καὶ τρίψας ἀμφότερα εἰς τὴν θυείαν ἡμέραν μίαν καὶ ἔμβαλε αὐτὰ εἰς βῆκον ὑέλινον καὶ φιμώσας τὸ στόμα αὐτοῦ μετὰ πυριμάχου πηλοκαρβούνου ἔχοντος πάχος δακτύλους γ΄ καὶ ἔμβαλε αὐτὰ εἰς αὐτόματον πῦρ ἐπὶ ὥρας ς΄ ἢ θ΄ καὶ εἶθ' οὕτως ἐκβαλὼν εὑρήσεις αὐτὰ βωλοποιηθέντα σιδηροειδῆ. Τοῦτο τρῖψον εἰς ἥλιον μετὰ ὕδατος πολλάκις. Ὅσον γὰρ τρίβεις αὐτὰ τοσοῦτον ξανθὰ γίνονται. Τὸ γὰρ θεῖον ἄπυρον τὰ φευκτὰ ἄφευκτα ποιεῖ.
The making of cinnabar. You must put unburnt sulphur, one ounce, and mercury, two ounces, in a mortar. After grinding them both in the mortar for a day, put them in a glass flask and seal its opening with a three-fingers thick fireclay made of mud and coal. Put them on a fire for the spontaneous digestion for 6 to 9 hours, then take it out and you will find an iron-coloured mass. Grind it several times in the sun with water. In fact, the more you grind, the more it turns yellow. Indeed, unburnt sulphur makes volatile substances fixed.

Text 2: Anonymous Byzantine recipe. Edited by A. Colinet, *Recettes alchimiques (Par. Gr. 2419; Holkamicus 109) Cosmas le Hiéromoine, Chrysopée*, Paris 2010, pp. lxxxviii-lxxxix.

Κινναβάρεως σκευασία. <Λαβὲ> ὑδραργύρου μέρη β΄, καὶ θείου ζῶντος λελειωμένου <...> οὔρου καθαροῦ μέρος α΄. Καὶ λαβὼν βικίον καθαρὸν δυνατὸν, καὶ ἄνευ καπνοῦ τῶν δυνάμεων βαστάσαι τὴν πυράν, βάλε τὴν σκευὴν εἰς αὐτό· μὴ γέμει δέ, ἀλλὰ μᾶλλον ἵνα ἐστὶ κενὸν ὅσον δάκτυλα β΄ ἢ γ΄. Καὶ ἀνάμιξον πάντα, καὶ ποίησον καμίνιον οἷον τοῦ ὑελοψοῦ· ἔστω δὲ τοιοῦτον βικίον εὐρύχωρον· καὶ ἄφες τόπον ὅσον θέλεις εἰσελθεῖν τὸ βικίον, καὶ χώρισον κάλαμον καὶ μετὰ ταῦτα ἄναψον τὸ καμίνιον. Ἔασον δὲ καὶ ἑτέραν θυριδίτζαν μικρὰν ὅθεν μέλλει εἰσελθεῖν τοῦ πυρὸς λάβρα κύκλωθεν. Τὸ δὲ σημεῖον τῆς ἐψήσεως τοιοῦτόν ἐστι· τήρησον τὸ κένωμα τοῦ βικίου, καὶ ἐὰν ἴδῃς ἐξερχόμενον καπνὸν ὡσεὶ πορφύρας σχῆμα ἔχοντα, καὶ τὴν θερμότητα (*fortasse lege* χρόαν) κινναβαρίζουσαν, ἰδοὺ γέγονεν. Κατάλειπε πλέον τοῦ ἐκκαίειν τὸ ὑέλιον· εἰ γὰρ τούτου γενομένου πλέον ἐθέλει ἐκκαῦσαι, ῥήγνυται τὸ ὑέλιον.
Preparation of cinnabar. Take mercury, two parts, naturally occuring sulphur (sulphur vivum) that has been ground, (a part), pure urine, a part. Take a clean, hard flask, which is resistant to a smokeless fire, and put the mixture in it. Do not fill the flask, but rather let it remain empty for two or three fingers. Mix all the ingredients together and set up an oven similar to the one used by glassmakers. The flask must be large. Leave enough room (in the oven) to fit the flask, split the reeds, and light the oven. There must be a little window where the flame can escape all around. This is the sign that the mixture is cooked: look at the empty part of the flask, and if you see a rising smoke that looks purple and has the colour of cinnabar, you must know that it is done. Do not allow the glass to be heated any longer. In fact, glass will break if you continue to heat it up too much.

Text 3: the Arabic *Book of the Keys of the Work* attributed to Zosimus of Panopolis, *Dār al-kutub*, MS Kīmiyā' 23, fol. 51r5–6.

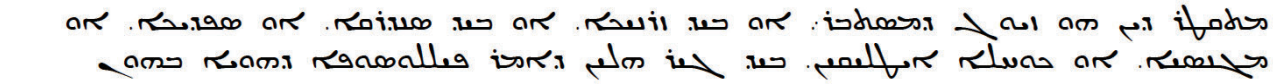
علمتك ان احكام العمل كله الطبخ والسحق. فان اردت الحق فاعلم ان الزئبق هو الذي يقلب الطبائع فيه تحجز وبه.

I taught you that what makes every work perfect is cooking and grinding. If you seek the truth, know that mercury is what transforms natures (that) are confined/fixed in it and through it.

Text 4: Pseudo-Democritus, *On the Making of Gold*, § 2 (that is, the fifth section of *Physika kai mystika*). Edited by M. Martelli, *The Four Books of Pseudo-Democritus*, Leeds 2013, p. 86.

Λαβὼν ὑδράργυρον, πῆξον τῷ τῆς μαγνησίας σώματι, ἢ τῷ τοῦ Ἰταλικοῦ στίμεως σώματι, ἢ θείῳ ἀπύρῳ, ἢ ἀφροσελήνῳ, ἢ τιτάνῳ ὀπτῷ, ἢ στυπτηρίᾳ τῇ ἀπὸ Μήλου, ἢ ἀρσενικῷ, ἢ ὡς ἐπινοεῖς.
Take mercury and make it solid (lit. "fix/freeze it") with the body of magnēsia, or the body of Italian stibnite, or with unburnt sulphur, or with moon foam, or with roasted lime, or with alum from Milos, or with orpiment, or according to your knowledge.

Text 5: Zosimus of Panopolis, Syriac *Book of Mercury*, Cambridge University Library, MS Mm. 6.29, fol. 57r20–57v3.



The mercury that we have detected is solidified by orpiment or realgar or white lead or *magnēsia* or Italian stibnite. It is indeed solidified by those substances in which the philosopher (i.e., Pseudo-Democritus) said that it is contained.

Text 6: the Arabic *Tome of Images*, book 2, attributed to Zosimus of Panopolis, İstanbul Arkeoloji Müzeleri Kütüphanesi (Istanbul Archeology Museum Library), MS 1574, fol. 50r15–50v1

الا ترين الحكيم كيف قال ان جعلتم محه الزئبق القنباري كان منها سر عظيم.

قلت. فما ذلك السر الدي يكون؟

قال. خذهما فاخلطهما. بعد ان تخلطهما حتى يغلظا فانك ستجدين الزئبق قد غلظ والذكر قد صار رماداً مخفاً في الزئبق.

Do you not see how the sage said: 'if you put the mercury from cinnabar with it (i.e. 'the male'), then a great secret belongs to them'? She said: 'And what is that secret?'. He said: 'Take the two and mix them. After mixing them until they thicken, you will find that the mercury becomes thick and the male turns into ashes hidden in the mercury.