

Exploring the ancient chemistry of mercury

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This paper explores the chemistry of mercury as described in ancient alchemical literature. Alchemy's focus on the knowledge and manipulation of natural substances is not so different from modern chemistry's purposes. The great divide between the two is marked by the way of conceptualizing and recording their practices. Our interdisciplinary research group, composed of chemists and historians of science, has set off to explore the cold and hot extraction of mercury from cinnabar. The ancient written records have been perused in order to devise laboratory experiments that could shed light on the material reality behind the alchemical narratives and interpret textual details in a unique perspective. In this way, it became possible to translate the technical lore of ancient alchemy into the modern language of chemistry. Thanks to the replication of alchemical practices, chemistry can regain its centuries-long history that has fallen into oblivion.

mercury | cinnabar | alchemy | extraction | replication

Alchemy is a difficult word. It encapsulates the long history of this discipline, which features Greek sources first translated into Arabic and then from Arabic into Latin. Alchemy stems from the Latin *alchimia*, a loanword that combines the Arabic article al- with chimia, which, in turn, can be traced back to the Greek chymeia (or chemeia). This term refers either to the action of smelting or to the ancient name of Egypt, the cradle of this practice. Interestingly, the same word chimia-and the related chimicus or chemicus—are at the basis of terms like chemistry and chemical (1). Chemistry, indeed, is the modern discipline most closely akin to alchemy, and a better understanding of the latter is critical to better reconstruct the history of chemistry (2). If alchemy often evokes almost mythical procedures, like the making of the philosophers' stone that can transform base metals into gold, the earliest alchemical texts produced in the Graeco-Roman Egypt (first to fourth century CE) deal with a vast variety of technical and, to some extent, experimental procedures. They mainly include recipes, which represent an invaluable source for understanding how our ancestors described, conceptualized, and manipulated natural substances. Such recipes were often dismissed as nonsense or even interpreted as mere allegories for spiritual practices of selfpurification. On the contrary, we detect in these recipes the actual description of chemical practices carried out in ancient workshops.

Our research adopts an interdisciplinary outlook through collaboration between chemists and historians of science to investigate one of the most intriguing elements in the history of alchemy, that is, mercury. A corpus of ancient sources was scoured in search of information on the extraction of mercury from the mineral cinnabar. A careful textual analysis is essential to interpret this rich variety of sources and translate ancient recipes into modern renderings.

In this study, we propose a suite of experiments that apply a circular hermeneutical approach to ancient recipes. We read the sources, propose chemical interpretations, test these hypotheses in the laboratory, and return to the sources (3). We followed the instructions of ancient recipes and used synthetic reagents, which enabled us to characterize the products and detect the roles played by the different ingredients. Then, we reproduced the recipes using mineral ores to confirm the possibility of the reaction with substances closer to those that were used by the ancients. In terms of equipment, we did not opt for historical reconstructions. Without affecting the results of the tested reactions, we adapted modern labware, thus significantly reducing the experiments' durations and conforming to modern safety protocols.

Background

Western alchemy emerged in Graeco-Roman Egypt, where, as early as the first centuries CE, a varied repertoire of alchemical texts was in circulation. Two Greek recipe books written on papyrus in the third century CE have come down to us (the so-called Leiden and Stockholm papyri). Other texts have survived in later copies transmitted by

Significance

Our research approach in the field of History of Science combines textual investigations with experimental replications of ancient alchemical procedures. Alchemy is often described as a pseudoscience, whereas it would be better described as a protoscience; indeed, we have reported procedures that never found their way into modern laboratories along with the formalization and translation of these procedures into the modern language of chemistry. We strongly believe that it is important to let the chemistry community know its history, which has deep roots in the past.

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Byzantine manuscripts or, in some cases, translated into Syriac and Arabic. Some texts were ascribed to authoritative figures: Maria the Jewess (the legendary designer of the heating method known as bain-marie), Chymes (the eponymous hero of alchemy that was called *chymeia* at that time), and the philosopher Democritus (who was credited with four alchemical treatises, a pseudonymous work now referred to as Pseudo-Democritus' books). Other key sources were produced by more historical figures, such as the Graeco-Egyptian alchemist Zosimus of Panopolis (third to fourth century CE). All these texts feature a close interest in mercury, whose mechanochemical properties captured the attention of early practitioners. After all, they conceptualized mercury as a common constituent of all metals. Zosimus reports that the Egyptian alchemist Pebichius (first to second century CE) stopped at the market and shouted, "All the (metallic) bodies are mercury" (4).

A proper investigation of the set of procedures that ancient alchemists followed to extract mercury from cinnabar may shed light on the experimental basis of this idea. Some of these techniques were described in earlier texts by authorities in different fields, from Theophrastus to Vitruvius, Dioscorides, and Pliny the Elder. These authors open an invaluable window onto operations that were applied in different branches of technical knowledge, such as ancient painting and pharmacy, and entered early alchemical literature. Ancient alchemists inherited and further developed this body of technical knowledge, which they condensed into recipes.

Indeed, ancient alchemical procedures have been transmitted over centuries as written instructions that scribes copied onto manuscripts hundreds of times. In this study, we trace the transmission and transformation of ancient instructions on how to process cinnabar from their earliest records in Theophrastus or Vitruvius up to the foundational texts of Graeco-Egyptian alchemy—that is, the Leyden papyrus, Pseudo-Democritus' recipes, and Zosimus' works, both in Greek and in Syriac translation—some of which are here edited and translated for the first time (for the full set of sources used for this study, see *SI Appendix, Supplementary Information Text*).

However, written instructions that deal with practices may remain obscure without their words being turned into deeds. Only by reenacting these practices in modern laboratories can it be possible to anchor such ancient texts to the chemical reality that they were intended to encapsulate.

Using this cross-disciplinary approach, we explore the ancient chemistry of mercury by investigating, in particular, Greek, Latin, and Syriac sources that describe its cold and hot extraction processes from cinnabar. Following the sources, we replicated the recipes and explored the viability and feasibility of the reactions as well as the technical underpinnings of the texts to disclose the practical dimensions of ancient alchemy and an unexpected variety of reactions. This study rectifies some explanations that have been commonly proposed for ancient technical knowhow and leads us to a better understanding of how ancient practitioners conceptualized and processed mercury.

Exploring Ancient Methods for Mercury Extraction

Ancient recipes describe many procedures to extract mercury from cinnabar. On the basis of the treatment of this mineral (either grinding or heating), we can cluster these procedures in two groups, namely, "cold extraction" and "hot extraction". **Cold Extraction.** The earliest known procedure for extracting mercury was recorded by the natural philosopher Theophrastus, who stated in his work *On Stones* (fourth century BCE), "Mercury is produced by grinding cinnabar with vinegar in a copper mortar with a copper pestle" (5, *SI Appendix, Sources: Ancient recipes on the extraction of mercury, text 1*).

In the 1920s, the chemist and historian of science Kenneth C. Bailey made the first and only attempt to replicate this procedure by grinding cinnabar and copper turnings with vinegar within a mortar (6). The cinnabar turned black—a chromatic change that indicates the presence of copper sulfide—whereas the surface of the copper turnings became covered in a mercury–copper amalgam (HgCu), which was also featured in our replication (*SI Appendix*, Figs. S1 and S2).

After Bailey's work, no further experiments were carried out, and the procedure fell into oblivion until Takacs cited Theophrastus' recipe as being the earliest witness to a mechanochemical reaction (7).

Modern chemists immediately recognize copper-that is, the metal from which pestles and mortars are made-as a key reagent. However, this procedural leap from instruments to reagents is not self-evident when observed from a historical perspective. Theophrastus' method was again reported by Pliny the Elder in the first century CE (8). A few centuries later, interesting variations were recorded in the work of the Graeco-Egyptian alchemist Zosimus of Panopolis. His writings include recipes that instruct how to use lead mortars and pestles to grind cinnabar in water or vinegar or in the juice of an unidentified plant (9, 10, SI Appendix, Sources: Ancient recipes on the extraction of mercury, text 3). Moreover, in another passage, Zosimus traces back the use of this equipment to first- to second-century authoritative sources. He claims that Maria the Jewess and Chymes used to grind cinnabar with pestles and mortars made of various metals, namely lead and tin (11, SI Appendix, Sources: Ancient recipes on the extraction of mercury, text 4). These sources may have conveyed the idea that metals constituted a key ingredient in the procedure. Indeed, in a recipe only preserved in Syriac translation, Zosimus prescribes the grinding of cinnabar with copper scrapings and vinegar until droplets of mercury become visible. What is left in the mortar is then heated in a closed vessel (12, SI Appendix, Sources: Ancient recipes on the extraction of mercury, text 5). Here, two points deserve special attention: 1) the metal is added to the procedure as an independent reagent, and 2) the second part of the procedure is performed in a closed vessel, as we shall better explain below.

We initially tested this set of cold extraction techniques by manually grinding synthetic cinnabar (HgS) and copper (Cu in powder) together with some drops of glacial acetic acid. The powder slowly turned black, and after 1 h of grinding, droplets of mercury, never recorded in any previous experimental report, were clearly visible (Fig. 1).

By ball milling cinnabar and copper together with some drops of glacial acetic acid, after 6 h, the reaction was complete. The X-ray powder diffraction pattern (XRPD) is characteristic of a metastable tetragonal form of chalcocite (13) (Cu₂S; *SI Appendix*, Fig. S4), meaning that the reaction was composed of reaction 1 (14, chalcocite exists as different crystal forms as well as different stoichiometry Cu_{2-x}S. For sake of clarity, we refer to chalcocite as Cu₂S.),

$$HgS + 2Cu \rightarrow Cu_2S + Hg.$$
 [1]

It is likely that the mercury obtained in this way is, to some extent, contaminated with copper; mercury, however, maintains its liquid phase.

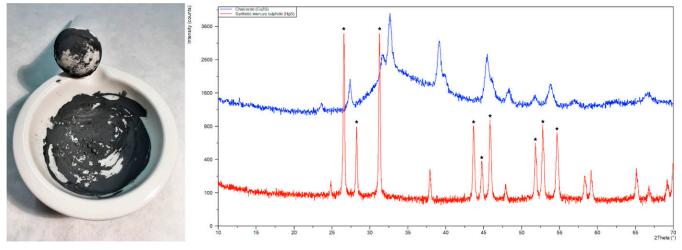


Fig. 1. *Left*, ceramic mortar in which HgS and copper powder were ground—droplets of mercury are visible at the bottom. *Right*, comparison between the XRPD pattern (blue solid line) of the residual powder obtained from the reaction of synthetic mercury sulfide and copper powder in the presence of acetic acid (reaction mixture ground with a ball mill for 4 h at 25 Hz; the high background in the pattern is due to the presence of the amorphous phase of metallic mercury in the final powder) and the XRPD pattern of synthetic mercury sulfide (red solid line). The asterisks in the red XRPD diffractogram indicate the peaks assigned to cinnabar. Phase identification was performed using the PDF 2 Release 2004 database.

If Takacs' article identifies the product of the reaction with mercury and generic copper sulfide, when this study began to be cited, other researchers limited the identification of different possible sulfides to "probably CuS" without conducting further analyses (15–17). Eventually, in recent papers, the reaction was formalized as reaction **2**,

$$HgS + Cu \rightarrow CuS + Hg.$$
 [2]

Our experiments make it clear that Cu_2S is the reaction product and not the incorrectly reported covellite (CuS) (18). This suggests that the reaction has only been cited in the scientific literature, without being reproduced and characterized. In fact, as in the case of the transmission of ancient recipes, which have been repeatedly copied in various manuscripts, modern explanations, when cited from one scientific paper to another, can be misinterpreted and their inaccuracies amplified over time.

All ancient sources stress the key role played by vinegar or water in the procedures. In our experiments, the mechanochemical reaction with copper and cinnabar was performed with glacial acetic acid, 6% acetic acid (as in vinegar), or water or under dry conditions. Droplets of mercury always became visible, but it is clear that the acid plays a catalytic role, perhaps by removing the oxide coating on the metal surface.

With respect to the use of metals other than copper, the replications confirmed that lead can reduce mercury. The XRPD pattern (SI Appendix, Fig. S5) indicates the presence of lead sulfide, but no droplets of mercury are visible owing to the formation of a powdery HgPb amalgam. When the reaction occurs with tin, the formation of the amalgam (HgSn) is clearly obtained, but the product of the oxidation of tin, probably tin sulfide, is hardly observed in the XRPD (SI Appendix, Supplementary Information Text). This amalgam, from which mercury can be easily recovered by heating, is a soft silvery paste (SI Appendix, Fig. S7), which ancient alchemists may have easily identified as being a kind of mercury. Indeed, in his writings, Zosimus seems to classify different types of mercury in accordance with the metals used in its extraction. In a long list of code names used for mercury, he also refers to it as "water of copper" and "water of lead", and he mentions various names given to "mercury from copper", "mercury from lead", and "mercury from tin" (19, 20), as if the metals used in the

cold extraction technique were conceptualized as sources of mercury along with cinnabar.

It is noteworthy that iron is not mentioned in this list. If we consider the standard reduction potentials of iron(0) to iron(II) or iron(III), we would expect the redox reaction to take place spontaneously, as it did with copper, lead, and tin. Therefore, we investigated the cold extraction of mercury in the presence of iron powder, but the reaction did not occur. This result further underlines the key role of replications, which avoid misinterpretations of ancient recipes based on pure chemical speculation. Experimental tests are always necessary, even for reactions that are chemically possible. It might not be coincidental that we have almost no record of ancient recipes on the cold extraction of mercury that mentions iron. Ancient alchemists might have tried to grind cinnabar with the metal and realized that it did not work.

Indeed, so far, we have detected only a single recipe that makes use of iron scrapings. After the above-mentioned recipe that explicitly introduces copper scrapings as a key ingredient to be mixed with cinnabar, Zosimus adds another procedure: "Take pure leaves of iron and make use of a mortar. Make scrapings as it is explained above (in the section) on copper (scrapings) or as it is explained by Pebichius, and (mercury) is produced" (21, *SI Appendix, Sources: Ancient recipes on the extraction of mercury, text 6*).

The instructions are incomplete here. Zosimus seems to refer back to the previous recipe, where copper scrapings were ground with cinnabar, and, in a second phase, the mixture was heated in a closed vessel. However, by simply grinding iron, no redox reaction occurs. On the contrary, only when cinnabar and iron are heated together in a closed vessel do they easily yield mercury. This procedure, which seemed to have been already known to the first- to second-century alchemist Pebichius, introduces us to hot extraction techniques.

Hot Extraction. According to ancient sources, the hot extraction method of processing mercury from cinnabar can be divided into three procedures: 1) simple heating of cinnabar, reported by Vitruvius; 2) heating cinnabar in a closed vessel in the presence of iron, a procedure described by Pliny, Dioscorides, and the Leyden papyrus; and 3) heating cinnabar in the presence of so-called "natron oil" in a closed vessel, as reported

by Pseudo-Democritus. Up until now, this variety has escaped the attention of modern historians who simply refer to the well-known reaction of cinnabar in the presence of oxygen at high temperatures (reaction 3) (22),

$$HgS + O_2 \rightarrow SO_2 + Hg.$$
 [3]

This procedure, which primarily entails the roasting of mercury ores, became standard after the publication of Agricola's De Re Metallica (1556) onward, whereas classical sources are less precise about the process. The Roman architect Vitruvius (first century BCE) only records an extemporary observation, noting that to produce the pigment, the ancients used to dry moist clods of cinnabar in ovens. When the fumes condensed, mercury could be found at the bottom of the oven and on the clods (23, SI Appendix, Sources: Ancient recipes on the extraction of mercury, text 7). Mercury is thus produced either by reaction of cinnabar with oxygen or by distillation. Indeed, if the ores encase droplets of mercury, this is simply distilled out when the ores are heated. As we shall see, since the first century CE, recipes on hot extraction techniques specify the use of closed vessels to facilitate the recovery of mercury and protect ancient alchemists from its fumes, whose toxicity was already well known in antiquity (24). However, because the procedure involves atmospheric oxygen, it was probably less efficient in a closed environment. We tested this by heating the cinnabar ores within an alumina crucible that was covered with a lid. In a crucible, the cinnabar sublimes and partially reacts with atmospheric oxygen, yielding mercury that condenses on the lid. The amount of oxygen estimated to be inside the crucible is insufficient to complete the reaction. Indeed, residual cinnabar and black metacinnabar (polymorphically stable at high temperatures) (25) were always observed, even after prolonged heating.

To overcome this hurdle and make the extraction technique more efficient, we can assume that ancient alchemists tried to add other ingredients before roasting cinnabar in closed vessels. Iron seems to have been used, at least according to Zosimus' recipe discussed at the end of the previous paragraph. Indeed, the crucial role of this metal is confirmed by Pliny the Elder (8, *SI Appendix, Sources: Ancient recipes on the extraction of mercury, text 8*) and Dioscorides (26), who describe the same method. Moreover, Dioscorides's recipe (first century CE) is also copied in the final section of the Leyden papyrus (third century CE), where it reads:

"They put an iron shell containing cinnabar in an earthenware vessel and enclose it with a convex lid smeared on with clay; then they light a fire upon (the vessel) with charcoal. The vapor that settles on the lid, when wiped off, is mercury" (27, *SI Appendix, Sources: Ancient recipes on the extraction of mercury, text 9*).

We replicated this procedure by placing the cinnabar on a small iron plate within an alumina crucible that was then covered with a lid (Fig. 2). Upon heating, cinnabar reacts with iron—and not with oxygen—according to reaction 4 (22),

$$HgS + Fe \rightarrow FeS + Hg.$$
 [4]

The reaction swiftly reaches completeness, and the residual powder characterized by XRPD confirms the presence of iron sulfide in the form of hexagonal troilite (28) (FeS; *SI Appendix*, Figs. S13 and S15). Despite the crucial role played by iron as a reagent, this has been overlooked in the modern studies of ancient extraction technologies (29, 30).

The third procedure was described by the alchemist Pseudo-Democritus (first century CE), as reported by a reliable Byzantine author called Christianos (sixth to seventh century CE). Democritus is said to have ground cinnabar together with natron oil and placed the mixture in a "double vessel" in order to capture all of the vapor, that is, the mercury sitting in the cinnabar (31, *SI Appendix, Sources: Ancient recipes on the extraction of mercury, text* 10). Byzantine manuscripts also transmit the recipe

"You must know that the transformation of cinnabar happens by means of natron oil: in this way cinnabar is melted by a light fire, as you know" (32, *SI Appendix, Sources: Ancient recipes on the extraction of mercury,* text 11).

The expression natron oil translates the Greek nitrelaion, which merges the terms nitron and oil (Gr. elaion). What is in fact meant by this expression is elusive and remains a matter of debate. We can exclude that this name refers to nitric acid, because the earliest known texts describing its production date back to the 13th century CE (33). This compound, indeed, was first attested in the Arabo-Latin tradition of the Jabirian corpus. The Greek term nitron probably refers to the mineral natron (sodium carbonate decahydrate) that was extracted from an ancient mining site known today as Wadi el-Natrun-a nearly dry lake in Egypt (34). However, its association with oil is uncertain. The alchemists probably referred to a mixture prepared by dispersing natron in a liquid. After all, they believed that liquid substances could react more promptly than solid ones, as had already been asserted by Aristotle (35). If we consider the compounds that were known at the time, this mixture likely constituted a solution of sodium carbonate dissolved in water, oil (e.g., castor and linseed oil), or vinegar. This supposition is supported by a Byzantine alchemist who identified natron oil as the "vinegar of natron" (36).

In a closed vessel, the role of sodium carbonate is pivotal, as demonstrated from the following experimental evidence. We mildly heated the same amount of cinnabar with and without sodium carbonate, avoiding the addition of any liquid. In the former case, the cinnabar fully converted into mercury; in the latter, unreacted cinnabar remained (Fig. 3).

To confirm the role played by sodium carbonate, we performed the extraction under vacuum to prevent the cinnabar reacting with atmospheric oxygen. Under this condition, mercury was obtained, and sodium sulfate (Na₂SO₄) was identified by an XRPD analysis as one of the products (*SI Appendix*, Fig. S43).

In the contemporary literature, this reaction was reported only once in a fairly old paper by Berthier, in which the residual powder is described as a mixture of sodium sulfate and sodium sulfide (Na₂S) (37). However, in all our experiments, no sodium sulfide was detected using XRPD or qualitative analysis by means of sodium nitroprusside solution (38), whereas sodium sulfate was always observed.

Once the key role of sodium carbonate (i.e., natron) in the hot extraction of mercury had been confirmed, its oily nature and role remained to be determined. We prepared saturated solutions of sodium carbonate in either water or vinegar, whose viscosity actually resembles that of oil. It is well known that when mixed with vinegar, sodium carbonate reacts, forming sodium acetate and carbon dioxide bubbles; this fact might have been recorded by the Byzantine alchemist Christianos when he claimed that natron effervesces when placed in a liquid (39). However, the low acetic acid concentration in vinegar does not fully consume the sodium carbonate.

To carry out the extraction, we used sodium carbonate dissolved in water or vinegar. The water (also contained in the vinegar) evaporates, and sodium sulfate and mercury are produced. In both cases, no significant improvement was observed in comparison to the reactions that occurred under dry conditions.

Finally, we tested a mixture of sodium carbonate and vegetable oil and found that even with mild heating, the oil easily burns,

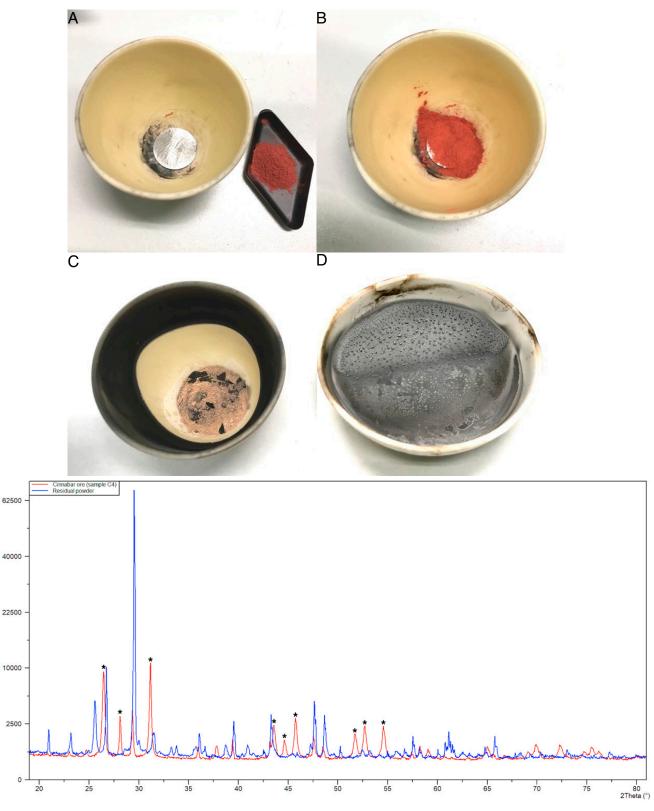


Fig. 2. *Top*, reaction vessel with iron plate and cinnabar powder before the reaction (A and B) and after 10 min of heating using a Bunsen burner (C)—the mercury condensed on the lid (D). *Bottom*, comparison between the XRPD of the cinnabar ore (sample C4, red solid line) and the residual powder obtained from the reaction of the cinnabar ore (sample C4) and iron plate (blue solid line); the reaction mixture was heated with a Bunsen burner. The asterisks in the red XRPD diffractogram indicate the peaks assigned to cinnabar.

and its decomposition prevents the extraction of mercury. If the temperature is reduced, decomposition is avoided, yet the extraction does not proceed. To summarize, like iron in the second method described above, natron serves as a reducing agent in the

reaction when this is carried out within a closed apparatus. The use of such equipment, always emphasized in ancient alchemical texts, contrasts with the common assumption that mercury was produced by simply roasting cinnabar in the presence of oxygen.

Intensity (counts



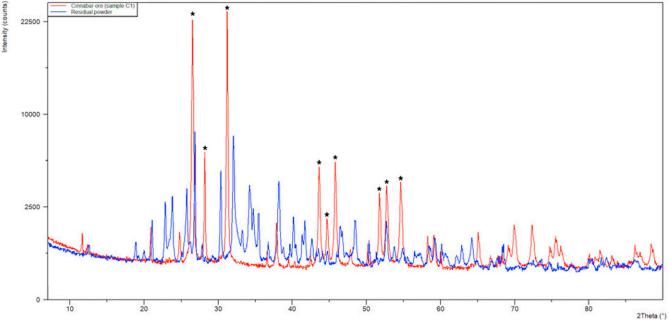


Fig. 3. *Top*, residual powders obtained after 48 h of heating at 300 °C—a mixture of cinnabar ores and sodium carbonate (*Left*) and the cinnabar ore (*Right*). *Bottom*, comparison between the XRPD of the cinnabar ore (sample C1, red solid line) and the residual powder obtained from the reaction of the cinnabar ore (sample C1) with sodium carbonate (blue solid line); the reaction mixture was then heated with a mantle (at 300 °C) for 48 h. The asterisks in the red XRPD diffractogram represent the peaks assigned to the cinnabar.

Discussion

This combined textual and experimental investigation of ancient sources rests on a basic assumption: the elusive and imaginative language of ancient alchemical sources ultimately captures a concrete material dimension and describes different attempts to manipulate a varied range of substances. Needless to say, the paradigms for conceptualizing and making use of these substances have changed dramatically since antiquity. Mercury, for instance, entered into many alchemical procedures meant to transform base metals into gold, an ambition that has long been abandoned in chemical laboratories. The efforts of the ancients to make gold, however, relied on and fostered experimental practices, such as the set of extraction techniques explored in this paper. These techniques exploited chemical properties of matter, which have remained unchanged over time and can be tested in modern laboratories. The goal of replication may be described as an attempt to translate the technical lore of ancient alchemy into the modern language of chemistry. Through replication, we try to identify ingredients, understand reactions, and measure variables about which ancient sources remain silent (e.g., time and temperature). The laboratory experience allowed us to verify different working hypotheses, and replication has shown that only a portion of what is possible in theory happens in practice. For instance, we discovered that iron—unlike copper, tin, and lead—does not work in cold extractions.

Moreover, it was demonstrated how crucial procedural information can often be found in the apparently minor textual details of ancient recipes. In cold extraction, copper was initially provided by the instruments (pestle and mortar) and later developed into a steady addition in the form of copper scrapings. Similarly, an iron shell turned out to be no mere sample holder but the source of the fundamental ingredient that enabled a reaction to take place. The experiments in a closed vessel have demonstrated that the role of oxygen in the hot extraction is definitely more marginal than what has been assumed so far. All these crucial details easily pass unnoticed, or can be even misinterpreted, when recipes are considered only in their textual dimension, stripped of their procedural value.

We demonstrated that, like iron, natron oil too was added to cinnabar, as it improved the extraction yield. The addition of these ingredients may have responded to practical needs. Indeed, they let cinnabar fully convert into mercury in closed vessels, which were introduced to limit the exposure to toxic fumes and collect the mercury drops more easily. The problematic identification of some substances and their dubious experimental role led us to wonder whether the choices of ingredients were not only based on technical factors but also influenced by cultural aspects. We wonder, for instance, why quicklime-which was well known by the ancients-is never mentioned in recipes describing hot extraction techniques. This mineral is now known to act as a reducing agent in mercury extraction by means of a reaction that does not require oxygen (22). In other words, it works as well as natron (SI Appendix, Supplementary Information *Text*). In this respect, the choice of the latter may have not only responded to practical needs. Natron, in fact, had a profound religious value in Graeco-Roman Egypt. The walls of Graeco-Egyptian temples feature recipes for the production of natron balls, which were used to cleanse the metallic statues of gods and to transform corpses into mummies as perfected bodies for the afterlife (40). This ritual background could have affected the choices of alchemists in their "experiments", which were intended to purify, transform, and enhance certain substances. In our case, cinnabar was purified and perfected into mercury by adding natron oil. Cultural considerations along with technical observations, thus, may have served as driving forces in ancient alchemical practices. These elements were at the core of the investigation reported in this study that began by exploring what ancient alchemists did and evolved into an analysis of how and why they operated as they did.

Conclusion

To sum up, this study explores the avenue opened by applying a chemical reading to ancient texts on the extraction of mercury. Modern replications and tests are based on a rigorous textual examination of these sources, many of which belong to the Graeco-Roman alchemical literature and have been often overlooked or simply ignored in previous studies. This interdisciplinary approach disclosed the chemical reality of ancient recipes by identifying the reaction products and formalizing the chemical reactions. These results deepen our understanding of ancient alchemy and broaden its spectrum of chemical practices. In this way, chemistry can regain a centuries-long history that has fallen into oblivion.

Ancient alchemists used several metals whose respective roles in cold extraction have been clearly defined in the course of

- L. M. Principe, The Secrets of Alchemy (University of Chicago Press, Chicago, 2013).
- H. Fors, L. M. Principe, H. O. Sibum, From the library to the laboratory and back again: Experiment as a tool for historians of science. Ambix 63, 85-97 (2016).
- M. Berthelot, R. Duval, La Chimie au Moyen-Âge, Vol. 2. L'Alchimie Syriaque (Imprimerie Nationale, Paris, 1893), p. 245.
- 5. D. E. Eichholz, Theophrastus, De Lapidibus. Edited with Introduction, Translation and Commentary (Clarendon Press, Oxford, 1965), p. 81.
- K. C. Bailey, The Elder Pliny's Chapters on Chemical Subjects, Part II. Edited, with Translation and 6. Notes (Edward Arnold & Co., London, 1932), p. 223.

our laboratory experiences. In the formation of amalgams (and mercury in different degrees of purity), we recognized the observational reality behind a key alchemical notion: mercury is a common constituent of all metals. By replicating the hot extraction procedures, we realized that the details of ancient recipes were often overlooked and downplayed by assuming that only atmospheric oxygen reduced cinnabar. On the contrary, these details point to a variety of substances (iron and natron oil) that are fundamental to the reaction, especially when performed in closed vessels. Ancient alchemical techniques exploited the chemical properties of substances, which can be explored in modern laboratories, and, at the same time, remain anchored in the cultural and historical ideas and beliefs of ancient practitioners.

Materials and Methods

Cold extractions were carried out by grinding the powder in a ceramic or bronze mortar or using a ball mill. Hot extractions were performed in both alumina crucibles with a lid on and in a round-bottom flask. In hot extractions under vacuum, a water pump was used (10^{-3} mbar). The reaction vessel was heated with a Bunsen burner (butane) or heating mantle (300 °C) when gentle heating was required. When the powdered cinnabar (or mercury sulfide) inside the reaction vessel was heated, the extraction proceeded, and metallic mercury condensed on the lid or in the neck of the flask, from where it could be recovered.

Mercury sulfide and sodium carbonate were purchased from Carlo Erba. Acetic acid, iron and tin powder, and copper (in powder and in small pieces) were purchased from Sigma-Aldrich; the extraction of mercury with lead was performed with lead obtained from an airgun pellet.

Olive oil, flaxseed oil, and wine vinegar were bought at a supermarket, whereas the cinnabar ores were acquired from Monte Amiata National Park in Italy (the ore composition consisted of mercury sulfide, calcite, and quartz).

A Retsch MM200 ball mill was used for the solid-state reaction, and the mechanosyntheses were performed using 5-mL steel jars that contained single agate spheres ($\emptyset = 5 \text{ mm}$, 0.545 g) at 20 Hz; in some cases, drops of vinegar or acetic acid were added. The quantities of the reagents were adjusted to enable complete extraction.

For the purpose of identifying the individual phases, XRPD patterns were collected using a PANalytical X'Pert Pro Automated diffractometer equipped with an X'celerator detector conforming to Bragg-Brentano geometry; Cu-K α radiation ($\gamma = 1.5418$ Å) without a monochromator at 2 θ ranged between 7° and 90° (step size: 0.033°; time/step: 20 s; Soller slit: 0.04 rad; antiscatter slit: 1; divergence slit: 1/2; 40 mA*40 kV).

PDF2 release 2004 was used for phase identification, and PDF card numbers are reported in *SI Appendix*.

Data Availability. All study data are included in the article and/or SI Appendix.

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- Syriac Manuscript, Cambridge University Library, Mm. 6.29, fol. 60v
- 10. M. Berthelot, R. Duval, La Chimie au Moyen-Âge, Vol. 2. L'Alchimie Syriaque (Imprimerie Nationale, Paris, 1893), p. 47.
- 11. M. Berthelot, C. É. Ruelle, Collection des Anciens Alchimistes Grecs, 3 Vols (G. Steinheil, Paris, 1887-1888), vol. 2, p. 172.
- 12. Syriac Manuscript, Cambridge University Library, Mm. 6.29, fol. 59v.
- A. Janosi, La structure du sulfure cuivreux quadratique. Acta Crystallogr. 17, 311-312 (1964). 13.

W. R. Newman, L. M. Principe, Alchemy vs. chemistry: The etymological origins of a historiographic 1. mistake. Early Sci. Med. 3, 32-65 (1998). 2

^{7.} L. Takacs, Quicksilver from cinnabar: The first documented mechanochemical reaction? JOM 52,

^{12-13 (2000).} H. Rackham, Pliny, Natural History, with an English Translation in Ten Volumes. Volume IX. Libri 8 XXXIII-XXXV (Loeb, Cambridge, MA, 1961), pp. 92-93.

- 14. B. J. Wuensch, M. J. Buerger, The crystal structure of chalcocite, Cu2S. Mineral Soc Am, Spec Pap 1, 164-170 (1963).
- P. Baláž, Mechanochemistry in extractive metallurgy: The modern science with old routes. Acta 15 Metall. Slovaca 4, 23-28 (2001).
- M. K. Beyer, H. Clausen-Schaumann, Mechanochemistry: The mechanical activation of covalent 16 bonds. Chem. Rev. 105, 2921-2948 (2005).
- 17 P. Baláž et al., Hallmarks of mechanochemistry: From nanoparticles to technology. Chem. Soc. Rev. 42, 7571-7637 (2013).
- 18 D. Tan, F. García, Main group mechanochemistry: From curiosity to established protocols. Chem Soc. Rev. 48, 2274-2292 (2019).
- M. Berthelot, R. Duval, La Chimie au Moyen-Âge, Vol. 2. L'Alchimie Syriaque (Imprimerie Nationale, 19 Paris, 1893), p. 244.
- 20. M. Martelli, Alchemical lexica in Syriac: Planetary signs, code names and medicines. Asiatische Studien - Études Asiatiques 75, 485-512 (2021).
- 21
- Syriac Manuscript, Cambridge University Library, Mm. 6.29, fol. 59v20-60r. L. H. Duschak, C. N. Schuette, *The Metallurgy of Quicksilver* (Washington Government Printing 22. Office, 1925).
- 23 I. D. Rowland, M. J. Dewar, Vitruvius, Ten Books on Architecture (Cambridge University Press, Cambridge, 1999), **book 7**, chap. 8, p. 93.
- 24
- E. R. Caley, Mercury and its compounds in ancient times. J. Chem. Educ. 5, 419–424 (1928).
 F. W. Dickson, G. Tunell, The stability of cinnabar and metacinnabar. Am. Mineral. 44, 471–487 (1959). 25
- 26 L. Y. Beck, Pedanius Dioscorides of Anazarbus, De Materia Medica (Olms-Wiedemann, Hildesheim, Zürich, New York, 2011), p. 379.

- 27. The Leyden Papyrus, fol. 16.
- E. F. Bertraut, Structure de FeS stoechiometrique. Bull. Soc. Fr. Mineral. Cristallogr. 79, 276 (1956).
 J. F. Healey, Pliny the Elder on Science and Technology (OUP, Oxford, 1999), p. 129.
- A. Hauptmann, Archaeometallurgy-Materials Science Aspects (Springer Nature, 2021), p. 379. 30 M. Berthelot, C. É. Ruelle, Collection des Anciens Alchimistes Grecs, 3 Vols (G. Steinheil, Paris, 31.
- 1887-1888), **vol. 2**, p. 123.
- 32. M. Berthelot, C. É. Ruelle, Collection des Anciens Alchimistes Grecs, 3 Vols (G. Steinheil, Paris, 1887-1888), vol. 2, p. 38.
- 33. A. Colinet, Recettes Alchimiques (Par. Gr. 2419; Holkhamicus 109), Cosmas le Hiéromoine, Chrysopée (Les Belles Lettres, Paris, 2010), pp. liv-lviii.
- R. Halleux, Papyrus de Leyde, Papyrus de Stockholm, Recettes (Les Belles Lettres, Paris, 1981), 34. p. 223.
- I. Düring, Aristotle's Chemical Treatise. Meteorologica, Book IV, with Introduction and Commentary 35. (Elanders Boktryckeri Antiebolag, Göteborg, 1944), p. 10.
- M. Berthelot, C. É. Ruelle, Collection des Anciens Alchimistes Grecs, 3 Vols (G. Steinheil, Paris, 36. 1887-1888), **vol. 2**, p. 134.
- 37. P. Berthier, De L'Action des Alcalis et des terres alcalines sur quelques sulfures métalliques. Ann. Chim. Phys. 2, 154-181 (1826).
- 38. A. Peloso, Analisi Chimica Qualitativa Inorganica Vol. II (Edizioni Libreria Cortina Padova, 1991), pp. 266-269.
- M. Berthelot, C. É. Ruelle, Collection des Anciens Alchimistes Grecs, 3 Vols (G. Steinheil, Paris, 39 1887-1888), **vol. 2**, p. 367.
- S. Aufrère, L'Univers Minéral dans la Pensée égyptienne (IFAO, Cairo, 1991), vol. 2, pp. 606-639 40.