Supplementary information

Lead soaps in paintings: friends or foes?

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List of scientific publications considered in the study

Table S1 lists the 180 publications considered for the study. Many of them were identified by a search "metal/lead soap/carboxylate" and "paint*" under SCOPUS.

The two right columns indicate the publications used to calculate data presented in Fig. 1 and Fig. 2. Some publications were not included in none of these figures, for example because they did not directly refer to the analysis of paintings (synthesis and characterization of reference carboxylates).

Publications	Fig.1	Fig.2
Aceto, M., Agostino, A., Boccaleri, E., Crivello, F. & Cerutti Garlanda, A. 2010. Identification of copper carboxylates as degradation residues on an ancient manuscript. Journal of Raman spectroscopy, 41(11):1434-40.	1	0
Adeosun, S. O. 1978. Thermal behaviour of the system lead (II) dodecanoate/lead acetate. Thermochimica Acta, 25(3):333-39.	0	0
Adeosun, S. O. & Akanni, M. S. 1978. A differential thermal analysis study of the system lead (II) dodecanoate/dodecanoic acid. Thermochimica Acta, 27(1):133-40.	0	0
Adriaens, A. & Dowsett, M. G. 2006. Applications of SIMS to cultural heritage studies. Applied Surface Science, 252(19):7096-101.	0	0
Amadori, M., Barcelli, S., Casoli, A., Mazzeo, R. & Prati, S. 2013. A scientific approach to the characterization of the painting materials of Fra Mattia della Robbia polychrome terracotta altarpiece. Applied Physics A, 113(4):1055-64.	1	1
Baeten, J., Romanus, K., Degryse, P., De Clercq, W., Poelman, H., Verbeke, K., Luypaerts, A., Walton, M., Jacobs, P., De Vos, D. & Waelkens, M. 2010. Application of a multi-analytical toolset to a 16th century ointment: Identification as lead plaster mixed with beeswax. Microchemical Journal, 95(2):227-34.	0	0
Balköse, D., Egbuchunam, T. O. & Okieimen, F. E. 2010. Thermal behaviour of metal soaps from biodegradable rubber seed oil. Journal of thermal analysis and calorimetry, 101(2):795-99.	0	0
Bayliss, S., van den Berg, K. J., Burnstock, A., de Groot, S., van Keulen, H. & Sawicka, A. 2016. An investigation into the separation and migration of oil in paintings by Erik Oldenhof. Microchemical Journal, 124(974-82.	1	0
Bearman, G., Doehne, E., Kronkright, D. & Manfredi, M. 2014. RTI Surface Normal Calibration with a 3D Printed Spatial Target: Turning Images into Data.	1	0
Beauchemin, S., MacLean, L. W. & Rasmussen, P. 2011. Lead speciation in indoor dust: a case study to assess old paint contribution in a Canadian urban house. Environmental Geochemistry and Health, 33(4):343-52.	1	1
Bergeon, S. & Faillant-Dumas, L. 1980. Restauration des peintures. Catalogue de l'exposition, paris, 1980. Éditions de la Réunion des musées nationaux.	0	0
Bonaduce, I., Carlyle, L., Colombini, M. P., Duce, C., Ferrari, C., Ribechini, E., Selleri, P. & Tine, M. R. 2012. New insights into the ageing of linseed oil paint binder: a qualitative and quantitative analytical study. Plos One, 7(11):e49333.	1	0
Bonaduce, I., Carlyle, L., Colombini, M. P., Duce, C., Ferrari, C., Ribechini, E., Selleri, P. & Tiné, M. R. 2012. A multi-analytical approach to studying binding media in oil paintings. Journal of thermal analysis and calorimetry, 107(3):1055-66.	1	0
Boon, J. J., van der Weerd, J., Keune, K., Noble, P. & Wadum, J. 'Mechanical and chemical changes in Old Master paintings: dissolution, metal soup formation and remineralization processes in lead pigmented ground/intermediate paint layers of 17th century paintings', in ICOM Committee for Conservation, ICOM-CC: 13th Triennial Meeting, Rio de Janeiro, 22-27 September 2002: preprints: ICOM-CC; James & James, 2002), pp. 401-06.	1	0

Boon, J. J., Gore, E., Keune, K. & Burnstock, A. 2004. Image analytical studies of lead soap aggregates and their relationship to lead and tin in 15th century lead tin yellow paints from the Sherborne Triptych. Infrared and Raman Users Group (IRUG) meeting, 29 March-1 April:66-74.	1	0
Boon, J., Keune, K. & Zucker, J. 2005. Imaging Analytical Studies of Lead Soaps Aggregating in Preprimed Canvas Used by the Hudson River Painter FE Church. Microscopy and Microanalysis, 11(S02):444-45.	1	0
Boon, J., Hoogland, F. & Keune, K. 'Chemical processes in aged oil paints affecting metal soap migration and aggregation', in Annual Meeting in Providence, Rhode Island, June, 2006), pp. 16-19.	1	1
Boon, J. & Van der Horst, J. 2008. Remarkably improved spatial resolution in SEM images of paint cross-sections after argon ion polishing. Preparation for painting The artist's choice and its consequences edited by Joyce H. Townsend, Tiarna Doherty, Gunnar Heydenreich and Jacqueline Ridge.	1	0
Boon, J. J., Ferreira, E. S. B., Van Der Horst, J., Stampanoni, M. & Marone, F. 'X-ray tomographic microscopy compared to ion polished paint cross sections of 19th century paints with and without metal soap aggregates ', in TECHNART 2009 Conference, ed. By Abstracts, B. o., (Athens, 2009), pp. 39.	1	0
Boon, J., Oberthaler, E., Ferreira, E., Marone, F. & Stampanoni, M. 2010. Xray Tomographic Microscopic Studies a Resin Embedded Paint Sample from a Mechanical Failing Area in the Floor Tiles of The Art of Painting by Johannes Vermeer (1632-1675). Microscopy and Microanalysis, 16(SupplementS2):1872-73.	1	1
Boon, J. J. & Hoogland, F. G. 2014. Investigating fluidizing dripping pink commercial paint on Van Hemert's Seven-series works from 1990–1995. Issues in Contemporary Oil Paint. Springer. pp. 227-46.	1	0
Bronken, I. A. T. & Boon, J. J. 2014. Hard dry paint, softening tacky paint, and exuding drips on composition (1952) by Jean-Paul Riopelle. Issues in Contemporary Oil Paint. Springer. pp. 247-62.	1	0
Burnstock, A., van den Berg, K. J., de Groot, S. & Wijnberg, L. 'An Investigation of Water-Sensitive Oil Paints', in Modern Paints Uncovered: Proceedings from the Modern Paints Uncovered Symposium: Getty Publications, 2007), pp. 177.	1	0
Carlyle, L. 1999. Paint driers discussed in 19th-century British oil painting manuals. Journal of the American Institute for Conservation, 38(1):69-82.	1	0
Carlyle, L. 2006. Historically accurate reconstructions of oil painters' materials: an overview of the Hart project 00-005. Reporting Highlights of the De Mayerne Programme,(Jaap J. Boon and Esters SB Ferrera), Den Haag:63-76.	1	0
Carlyle, L., Boon, J. J., Haswell, R. & Stols-Witlox, M. 2008. Historically accurate ground reconstructions for oil paintings. Preparation for Painting:110-22.	1	0
Casadio, F. & Rose, V. 2013. High-resolution fluorescence mapping of impurities in historical zinc oxide pigments: hard X-ray nanoprobe applications to the paints of Pablo Picasso. Applied Physics A, 111(1):1-8.	1	1
Casadio, F., Miliani, C., Rosi, F., Romani, A., Anselmi, C., Brunetti, B., Sgamellotti, A., Andral, J. L. & Gautier, G. 2013. Scientific investigation of an important corpus of Picasso paintings in Antibes: New insights into technique, condition, and chronological sequence. Journal of the American Institute for Conservation, 52(3):184-204.	1	1
Catalano, J., Murphy, A., Yao, Y., Alkan, F., Zumbulyadis, N., Centeno, S. A. & Dybowski, C. 2014. 207Pb and 119Sn Solid-State NMR and Relativistic Density Functional Theory Studies of the Historic Pigment Lead–Tin Yellow Type I and Its Reactivity in Oil Paintings. The Journal of Physical Chemistry A, 118(36):7952-58.	1	0
Catalano, J., Yao, Y., Murphy, A., Zumbulyadis, N., Centeno, S. A. & Dybowski, C. 2014. Nuclear Magnetic Resonance Spectra and 207Pb Chemical-Shift Tensors of Lead Carboxylates Relevant to Soap Formation in Oil Paintings. Applied Spectroscopy, 68(3):280-86.	1	0
Catalano, J., Murphy, A., Yao, Y., Yap, G. P. A., Zumbulyadis, N., Centeno, S. A. & Dybowski, C. 2015. Coordination geometry of lead carboxylates - spectroscopic and crystallographic evidence. Dalton Transactions, 44(5):2340-47.	1	0
Centeno, S. A. & Mahon, D. 2009. The chemistry of aging in oil paintings: metal soaps and visual changes. The Metropolitan Museum of Art Bulletin:12-19.	1	1
Chevreul, M. E. & France, I. n. d. 1850. Recherches expérimentales sur la peinture à l'huile. F. Didot frères.	1	0
Chiriu, D., Ricci, P. C., Carbonaro, C. M., Nadali, D., Polcaro, A. & Mocci, F. 2016. Drying oil detected in mid-third Millennium B.C. Mesopotamian clay artifacts: Raman spectroscopy and DFT simulation study. Microchemical Journal, 124(386-95.	1	0
Clarke, M. & Boon, J. J. 'A multidisciplinary NWO PRIORITEIT project on Molecular Aspects of Ageing in Painted Works of Art. Final report and highlights 1995-2002', (Amsterdam: © FOM Institute AMOLF, 2003).	1	1
Corbeil, M.-C. & Robinet, L. 2002. X-ray powder diffraction data for selected metal soaps. Powder Diffraction, 17(1):52-60.	1	0
Corkery, R. & Hyde, S. 1996. On the swelling of amphiphiles in water. Langmuir, 12(23):5528-29.	0	0
Corkery, R. 1997. Langmuir-Blodgett (LB) multilayer films. Langmuir, 13(14):3591-94.	0	0
Corkery, R. 1998. Artificial biomineralisation and metallic soaps.	0	0
Corkery, R. W. 2004. A variation on Luzzati's soap phases. Room temperature thermotropic liquid crystals. Physical Chemistry Chemical Physics, 6(7):1534-46.	1	0
Corkery, R. W. 2008. Metal organic framework (MOF) liquid crystals. 1D, 2D and 3D ionic coordination polymer structures in the thermotropic mesophases of metal soaps, including alkaline earth, transition metal and lanthanide soaps. Current Opinion in Colloid & Interface Science, 13(4):288-302.	0	0
Correia, A. M., Clark, R. J. H., Ribeiro, M. I. M. & Duarte, M. L. T. S. 2007. Pigment study by Raman microscopy of 23 paintings by the Portuguese artist Henrique Pousão (1859–1884). Journal of Raman spectroscopy, 38(11):1390-405.	0	0
Cotte, M., Checroun, E., Susini, J., Dumas, P., Tchoreloff, P., Besnard, M. & Walter, P. 2006. Kinetics of oil saponification by lead salts in ancient preparations of pharmaceutical lead plasters and painting lead mediums. Talanta, 70(5):1136-42.	1	0
Cotte, M., Checroun, E., Susini, J. & Walter, P. 2007. Micro-analytical study of interactions between oil and lead compounds in paintings. Applied Physics a-Materials Science & Processing, 89(4):841-48.	1	1
Cotte, M., Susini, J., Solé, V. A., Taniguchi, Y., Chillida, J., Checroun, E. & Walter, P. 2008. Applications of synchrotron-based micro-imaging techniques to the chemical analysis of ancient paintings. Journal of Analytical Atomic Spectrometry, 23(820-28.	1	1
Daniels, V. 2007. Analyses of Copper- and Beeswax-Containing Green Paint on Egyptian Antiquities. Studies in Conservation, 52(1):13-18.	1	0

De Groot, S., van Keulen, H., Burnstock, A. & Van den Berg, J. D. J. 'FTIR analysis of five paintings by Pierre-Auguste Renoir', in IRUG6 Conference, (Firenze, Italy, 2004), pp. 143-48.	1	1
de Rocha Marques, R. 2014. Analysis and Treatment of a Nineteenth Century Oil Painting, Faculdade de Ciências e Tecnologia, Universidade Nova de Lisboa.	1	0
de Viguerie, L., Payard, P. A., Portero, E., Walter, P. & Cotte, M. 2016. The drying of linseed oil investigated by Fourier transform infrared spectroscopy: Historical recipes and influence of lead compounds. Progress in Organic Coatings, 93(46-60.	1	0
Devesa, J. M. 2012. Treatment of a nineteenth century male portrait in oil including the characterisation of materials, technique and a study of the lead soap aggregation in the paint composite.	1	1
Doherty, B., Daveri, A., Clementi, C., Romani, A., Bioletti, S., Brunetti, B., Sgamellotti, A. & Miliani, C. 2013. The Book of Kells: A non-invasive MOLAB investigation by complementary spectroscopic techniques. Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, 115(0):330-36.	0	0
Dubus, M., Aucouturier, M., Colinart, S., Dran, J.-C., Gunn, M., May, R., Moignard, B., Salomon, J., Walter, P., Colson, I., Laurent, A.-M. & Leroy, M. 2002. Evaluation de la corrosion du plomb aux archives nationales de France. PUBLISHED IN THE 13TH TRIENNIAL MEETING RIO DE JANEIRO PREPRINTS ICOM COMMITTEE FOR CONSERVATION, II(851-59.	0	0
Duffy, M., Martins, A. & Boon, J. J. 2014. Metal Soaps and Visual Changes in a Painting by René Magritte–The Menaced Assassin, 1927. Issues in Contemporary Oil Paint. Springer. pp. 197-203.	1	0
Duval, A. R. 1992. Les préparations colorées des tableaux de l'ecole française des 17 et 18 siècles. Studies in Conservation, 37(239-58).	0	0
Eissa, S., Lampakis, D., Karapanagiotis, I., Panayiotou, C., Afifi, H. A. M. & Abd-El Hady, M. 2015. Investigation of painted stucco in historic buildings of Delta, Egypt. Archaeological and Anthropological Sciences:1-10.	1	0
Erhardt, D., Tumosa, C. S. & Mecklenburg, M. F. 2005. Long-term chemical and physical processes in oil paint films. Studies in Conservation, 50(2):143-50.	0	0
Essien, E. A., Umoren, S. A., Essien, E. E. & Udoh, A. P. 2012. Preparation and evaluation of cucumeropsis mannii naud. Seed oil metallic soaps as driers in gloss paint. Journal of Materials and Environmental Science, 3(3):477-84.	1	0
Favaro, M., Bianchin, S., Vigato, P. A. & Vervat, M. 2010. The palette of the Macchia Italian artist Giovanni Fattori in the second half of the xixth century. Journal of Cultural Heritage, 11(3):265-78.	1	1
Faubel, W., Simon, R., Heissler, S., Friedrich, F., Weidler, P. G., Becker, H. & Schmidt, W. 2011. Protrusions in a painting by Max Beckmann examined with confocal [small mu]-XRF. Journal of Analytical Atomic Spectrometry.	1	0
Ferreira, E. S., Morrison, R., Keune, K. & Boon, J. J. 2006. Chemical Characterisation of thin intermediate layers: Case study of a sample from the 15th century painting, The Descent from the Cross by Rogier van der Weyden. Reporting Highlights of the De Mayerne Programme:53.	1	1
Ferreira, E. S., Morrison, R. & Boon, J. J. 2008. Imaging Chemical Characterisation of Preparatory Layers in Fifteenth and Sixteenth-century Northern European Panel Paintings. Preparation for Paintings: The Artist's Choice and Its Consequences:50-58.	1	0
Ferreira, E. S. B., Boon, J. J., Van Der Horst, J., Scherrer, N. C., Marone, F. & Stampanoni, M. '3D synchrotron x-ray microtomography of paint samples', in Proceedings of SPIE - The International Society for Optical Engineering, 2009).	1	1
Ferreira, E. S. B., Boon, J. J., Stampanoni, M. & Marone, F. 'Study of the mechanism of formation of calcium soaps in an early 20th century easel painting with correlative 2D and 3D microscopy', in ICOMCC, (Lisboa, 2011).	1	0
Ferreira, E., Gros, D., Wyss, K., Scherrer, N., Zumbühl, S. & Marone, F. 2015. Faded shine…. The degradation of brass powder in two nineteenth century paintings. Heritage Science, 3(1):1-11.	1	1
Fiore, D., Maier, M., Parera, S. D., Orquera, L. & Piana, E. 2008. Chemical analyses of the earliest pigment residues from the uttermost part of the planet (Beagle Channel region, Tierra del Fuego, Southern South America). Journal of Archaeological Science, 35(12):3047-56.	1	0
Fischer, A. & Hanan, S. 1965. Driers based on tall oil—A review. Journal of the American Oil Chemists' Society, 42(2):109-11.	1	0
Gautier, G., Bezur, A., Muir, K., Casadio, F. & Fiedler, I. 2009. Chemical Fingerprinting of Ready-Mixed House Paints of Relevance to Artistic Production in the First Half of the Twentieth Century. Part I: Inorganic and Organic Pigments. Applied Spectroscopy, 63(6):597-603.	1	0
Gramtorp, D., Botfeldt, K., Glastrup, J. & Simonsen, K. P. 2015. Investigation and conservation of Anne Marie Carl-Nielsen's wax models. Studies in Conservation, 60(2):97-106.	1	0
Heeren, R., Boon, J. J., Noble, P. & Wadum, J. 'Integrating imaging FTIR and secondary ion mass spectrometry for the analysis of embedded paint cross-sections', in Triennial meeting (12th), Lyon, 29 August-3 September 1999: preprints. Vol. 1: James & James, 1999), pp. 228-33.	1	0
Helou-de La Grandiere, P., Le Hô, A.-S. & Mirambet, F. 2008. Delaminating paint films at the end of 1950s: A case study on Pierre Soulages. Preparationfor Paintings: The Artist's Choice and Its Consequence:156-62.	1	1
Helwig, K., Poulin, J., Corbeil, M.-C., Moffatt, E. & Duguay, D. 2014. Conservation Issues in Several Twentieth-Century Canadian Oil Paintings: The Role of Zinc Carboxylate Reaction Products. Issues in Contemporary Oil Paint. Springer. pp. 167-84.	1	0
Herm, C. 'Infrared spectroscopic study of some more metal soaps', in IRUG6 Conference, (Firenze, 2004), pp. 267-72.	1	0
Hermans, J. J., Keune, K., van Loon, A., Corkery, R. W. & Iedema, P. D. 2014. The molecular structure of three types of long-chain zinc(II) alkanoates for the study of oil paint degradation. Polyhedron, 81(0):335-40.	1	0
Hermans, J. J., Keune, K., van Loon, A. & Iedema, P. D. 2015. An infrared spectroscopic study of the nature of zinc carboxylates in oil paintings. Journal of Analytical Atomic Spectrometry.	1	0
Higgitt, C., Spring, M. & Saunders, D. 2003. Pigment-medium interactions in oil paint films containing red lead or lead-tin yellow. National Gallery Technical Bulletin, 24(75-95.	1	1
Higgitt, C. & Plater, M. J. 2004. Old masters in the spotlight. Chemistry World, 1(4).	1	1
Higgitt, C. & White, R. 2005. Analyses of paint media : New studies of italian paintings of the fifteenth and sixteenth centuries. National Gallery Technical Bulletin, 26(88-104.	1	0
Holubec, I. M. 2008. The whole world is angelikamad': preparatory layers on canvas used by Angelika Kauffmann. Preparation for Painting: The Artist's Choice and Its Consequences:100.	0	0

Huijbregts, Z., van Schijndel, A., Schellen, H., Keune, K. & Hommes, M. E. 'Computational Modelling of the Impact of Solar Irradiance on Chemical Degradation of Painted Wall Hangings in an Historic Interior': Proceedings of the 2014 COMSOL Conference in Cambridge, 2014).	1	0
Ishioka, T., Maeda, K., Watanabe, I., Kawauchi, S. & Harada, M. 2000. Infrared and XAFS study on structure and transition behavior of zinc stearate. Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, 56(9):1731-37.	0	0
Ishioka, T., Yan, W., Strauss, H. L. & Snyder, R. G. 2003. Normal mode analyses of methyl palmitate all-trans and disordered forms in wagging progressive region. Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, 59(4):671-80.	0	0
Izzo, F. C., Balliana, E., Pinton, F. & Zendri, E. 2014. A preliminary study of the composition of commercial oil, acrylic and vinyl paints and their behaviour after accelerated ageing conditions. Conservation Science in Cultural Heritage, 14(1):353-69.	1	0
Izzo, F. C., Ferriani, B., Van den Berg, K. J., Van Keulen, H. & Zendri, E. 2014. 20th century artists' oil paints: the case of the Olii by Lucio Fontana. Journal of Cultural Heritage, 15(5):557-63.	1	1
Izzo, F. C., van den Berg, K. J., van Keulen, H., Ferriani, B. & Zendri, E. 2014. Modern oil paints–formulations, organic additives and degradation: some case studies. Issues in Contemporary Oil Paint. Springer. pp. 75-104.	1	0
Janssens, K., Dik, J., Cotte, M. & Susini, J. 2010. Photon-Based Techniques for Nondestructive Subsurface Analysis of Painted Cultural Heritage Artifacts. Accounts of Chemical Research, 43(6):814-25.	1	0
Jacobsen, A. & Gardner, W. H. 1941. Zinc Soaps in Paints. Zinc Oleates. Industrial & Engineering Chemistry, 33(10):1254-56.	1	0
Jogarao, A. 1948. Evaluation of red lead pigments. Pt. II. J. Sci. Indust. Res., India B, 7(138-44).	1	0
Kastens, M. L. & Hansen, F. R. 1949. Drier soap manufacture. Industrial & Engineering Chemistry, 41(10):2080-90.	1	0
Katsibiri, O. & Howe, R. F. 2010. Characterisation of the transparent surface coatings on post-Byzantine icons using microscopic, mass spectrometric and spectroscopic techniques. Microchemical Journal, 94(1):14-23.	1	1
Kaszowska, Z., Malek, K., Pańczyk, M. & Mikołajska, A. 2013. A joint application of ATR-FTIR and SEM imaging with high spatial resolution: Identification and distribution of painting materials and their degradation products in paint cross sections. Vibrational Spectroscopy, 65(0):1-11.	1	0
Keune, K. & Boon, J. J. 2004. Imaging Secondary Ion Mass Spectrometry of a Paint Cross Section Taken from an Early Netherlandish Painting by Rogier van der Weyden. Analytical Chemistry, 76(1374-85.	1	0
Keune, K. 2005. Binding medium, pigments and metal soaps characterised and localised in paint cross-sections.	1	1
Keune, K., Ferreira, E. S. & Boon, J. J. 2005. Characterization and localization of the oil-binding medium in paint cross-sections using imaging secondary ion mass spectrometry. ICOM Committee for Conservation, preprints, 2(796-802.	1	0
Keune, K. & Boon, J. J. 2007. Analytical imaging studies of cross-sections of paintings affected by lead soap aggregate formation. Studies in Conservation, 52(3):161-76.	1	0
Keune, K., Hoogland, F., Boon, J. J., Peggie, D. & Higgitt, C. 2009. Evaluation of the "added value" of SIMS: A mass spectrometric and spectroscopic study of an unusual Naples yellow oil paint reconstruction. International Journal of Mass Spectrometry, 284(1–3):22-34.	1	0
Keune, K., van Loon, A. & Boon, J. J. 2011. SEM backscattered-electron images of paint cross sections as information source for the presence of the lead white pigment and lead-related degradation and migration phenomena in oil paintings. Microscopy and Microanalysis, 17(05):696-701.	1	1
Keune, K., Boon, J. J., Boitelle, R. & Shimadzu, Y. 2013. Degradation of Emerald green in oil paint and its contribution to the rapid change in colour of the Descente des vaches (1834–1835) painted by Théodore Rousseau. Studies in Conservation, 58(3):199-210.	1	1
Keune, K. & Boevé-Jones, G. 2014. Its surreal: zinc-oxide degradation and misperceptions in Salvador Dalí's Couple with Clouds in their Heads, 1936. Issues in Contemporary Oil Paint. Springer. pp. 283-94.	1	0
Kneepkens, I. 2012. Understanding historical recipes for the modification of linseed oil.	0	0
Kokkori, M., Casadio, F., Sutherland, K. & Vermeulen, M. 2014. Charting the development of oil-based enamel paints through the correlation of historical paint technology manuals with scientific analysis. Issues in Contemporary Oil Paint. Springer. pp. 117-25.	1	0
Lacouture, F., Francois, M., Didierjean, C., Rivera, J.-P., Rocca, E. & Steinmetz, J. 2001. Anhydrous lead (II) heptanoate. Acta Crystallographica Section C: Crystal Structure Communications, 57(5):530-31.	0	0
Legrand, S., Alfeld, M., Vanmeert, F., De Nolf, W. & Janssens, K. 2014. Macroscopic Fourier transform infrared scanning in reflection mode (MA-rFTIR), a new tool for chemical imaging of cultural heritage artefacts in the mid-infrared range. Analyst, 139(10):2489-98.	1	0
Leone, B., Burnstock, A., Jones, C., Hallebeek, P., Boon, J. J. & Keune, K. 'The deterioration of cadmium sulphide yellow artists' pigments '.	1	0
Liang, J. & Scott, D. A. 2014. Green-copper containing waxy paint on two Egyptian polychrome artifacts: A technical study. Studies in Conservation, 59(6):391-403.	1	0
Manguta, C. 2010. The XVIIth Century's thixotropic painting medium: Rubens, reconstruction following Van Dyck's recipes.	0	0
Maor, Y. 2008. Delamination of oil paints from acrylic grounds.	1	0
Martin, E., Duval, A. R. & Laveissiere, S. 1998. La matière comme indice chronologique: Georges de La Tour Le Tricheur à l'as de carreau.	1	0
Martin, E. 2008. Grounds on canvas 1600–1640 in various European artistic centres. Preparation for painting, the artist's choice and its consequences. London: Archetype Publications:59-67.	1	1
Mass, J. L., Woll, A. R., Ocon, N., Bisulca, C., Wazny, T., Griggs, C. B. & Cushman, M. 'Collaboration or appropriation? Examining a 17th c. panel by David Teniers the Younger and Jan Brueghel the Younger using confocal X-ray fluorescence microscopy', in MRS Proceedings: Cambridge Univ Press, 2007), pp. 1047-Y07-01.	1	1
Matura, R. 1965. Divalent metal salts of long chain fatty acids. J. Chem. Soc. Jpn, 86(560-72.	0	0
Mazzeo, R., Joseph, E., Prati, S. & Millemaggi, A. 2007. Attenuated Total Reflection-Fourier transform infrared microspectroscopic mapping for the characterisation of paint cross-sections. Analytica Chimica Acta, 599(1):107-17.	1	0
Mazzeo, R., Prati, S., Quaranta, M., Joseph, E., Kendix, E. & Galeotti, M. 2008. Attenuated total reflection micro FTIR characterisation of pigment-binder interaction in reconstructed paint films. Analytical and Bioanalytical Chemistry:1-12.	1	0

Meilunas, R. J., Bentsen, J. G. & Steinberg, A. 1990. analysis of aged paint binders by FTIR spectroscopy. Studies in Conservation, 35(33-51).	1	1
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List of historical recipes describing preparation of a lead-oil medium

Before 11th Century

Greek sources? Mappae clavicula, early 9th Century (earliest known copy) (Nadolny, 2008) p. 2

Quot1- *Gilding recipe.*Covering: Raw hide. Ground: **lead white** or "whatever other colour the ground may be", Binder not specified. It is unclear whether the stretched hide should be interpreted as a covering over a wooden panel or simply as a type of leather object.

Manuscript of Eraclius (Merrifield, 1999),p. 228 and 230

Quot2- XXIV. [268] *How wood is to be prepared before painting on it.* –[…] Grind dry **white –lead** upon a stone, but not so finely as if you were going to paint with it. Then melt wax over the fire in a vase, add tiles ground fine. Then mix it with the white-lead which you had ground, stirring it frequently with a small stick, and so let it cool. Then heat an iron, and with it melt the wax into the little fissures [of the wood], until they are level, and then scrape off the rough parts with a knife. And if you hesitate about mixing white-lead with the wax, know that the more you mix with it, the harder it will be. And when you made it smooth, as I was saying, mix plenty of **white-lead** very finely ground, with linseed-oil, and lay an excessively thin coat of it wherever you intend to paint with a brush of ass's hair adapted for that purpose. […]

Quot3- XXIX [260] *How oil is prepared for tempering colours*.- Put a moderate quantity of lime into oil and heat it, continually scumming it; add **ceruse** to it according to the quantity of oil, and put it in the sun for a month or more, stirring it frequently. And know that the longer it remains in the sun, the better it will be. Then strain and keep it, and distemper the colours with it.

15th Century

Bolognese manuscript (Merrifield, 1999) p.488

Quot4- 207. *To make liquid varnish in another manner*. - Take 1 lb. of linseed oil, and put it into a new glazed jar, and then take 1/2 a quarter [of an ounce?] of roche alum in powder, and an equal quantity of **minium** or vermilium ground fine, and 1/2 oz. of incense also ground fine. Mix all these ingredients together and put them into the oil to boil, stirring it with a stick; and when the oil is boiling, as it is likely to run over, have another glazed jar ready, and put it by that which contains the oil, so as to catch the oil that runs over, in order that it may not run on the ground, and in this manner make it boil up 3 or 4 times, and each time pour back what has run over, on that which is boiling in the jar. Having done this, set fire to the oil on the right hand side with a lighted straw, and let the oil burn on the upper part, but so that the jar may not burn on the inside, on account of the too great heat, for otherwise the oil would smell unpleasantly. When you light the oil with the straw, remove the jar from the fire and let it burn while you can say three paternosters, then extinguish the oil with a wooden cover, putting it upon the jar, and when it is extinguished, remove the cover in order to let the vapour escape. Then put it back over the fire; do this 3 times, and it is done.

Cennini, *Libro dell'arte ,* 1437 (Merrifield, 1999) p. ccxxxxix

Quot5- The mordant of Cennini consisted of linseed oil, vernice (dry sandarac), and **white lead**.

Liber diversarum arcium(14th-15th C.)(Nadolny, 2008)

Quot6- *Ground recipe for wood*: […] Priming for painting on oil: **lead (white)** and oil (albo ad oleum).

The formula for the first lead medium, Antonello da Messina, as proposed by Maroger (Maroger, 1948)

Quot7- Litharge or **White Lead**. one part (measured by weight) Linseed or Walnut Oil. three to four parts (measured by weight). The **lead of litharge** should be ground with a little of the oil in the same manner that colors are ground. This will not take the whole amount of the oil, so when the **lead** issufficiently pulverized, the rest should be added and the mixture cooked over a low fire, care being taken to interpose between receptacle and the source of the heat several asbestos mats, or even a metal screen, so that the heat will be evenly distributed. This mixture must be stirred constantly with a spatula.

*The formula for the second lead medium, the probable technique of Leonardo da Vinci*as proposed by Maroger (Maroger, 1948)

Quot8- Litharge I part; Raw Linseed or Walnut Oil. 3 to 4 parts; Water 3 to 4 parts. After weighing, the **litharge** and oil are mixed thorougly by grinding the **litharge** first with a little o the oil, then adding the rest. This mixture is placed over a low fire and allowed to siromer. (Asbestos mats should be used under the vessel to distribute the heat evenly and prevent burning.) It is necessary to stir the mixture often with a spatula. As soon as the water begins to boil, a strong yellowish spume will appear on the surface. It will increase rapidly in volume and after a certain time-about half an hour-according to the quantity with which one is working-this spume will become whitish. The operation can be considered terminated when the bubbles no longer have small particles of litharge attached to their periphery. It is advisable to replace the water as fast as it evaporates, but this must be done very cautiously because the introduction of too rouch water at one time retards the operation. When all the spume has become a yellowish white, it should be tested by taking a small quantity with the palette knife and spreading it out on the plaque that is used for grinding colors. The mixture should appear homogeneous and should contain no more particles of unassiroilated litharge. When it cools, it should have the aspect of congealed grease, but it will be lighter in color. When it meets these conditions, it may be removed from the fire and allowed to cool. It should be washed thoroughly to eliminate all the glycerin and other extraneous matters which will remain in solution in the water. Its color will depend largely upon the oil used, but by making the paste in this way, it should be lighter-a clear yellow-the color almost of fresh butter.

16th Century

The formula for the lead and wax mediums of the Venetians, Giorgione, Titian, Tintoretto,as proposed by Maroger (Maroger, 1948)

Quot9- THE BLACK OIL, which seems to have been the invention of Giorgione, was the basis of the technique of the Italians of the High Renaissance, and of their successors down to the early part of the nineteenth century..

It can be made with **white lead** (basic carbonate), or with **calcined white lead** (**massicot or protoxide**) or with **litharge** (oxide).'The procedure is this (all measurements are given by weight). Place in a bowl: I or 2 parts (or 3-10 Grammes) of lead or litharge 20 parts (or I00 grammes) of raw linseed or walnut oil. These must be ground and mixed in the same way as for Antonello's medium. Place the receptacle containing the mixture over a low fire (protected by the asbestos mats, mentioned in previous formulas) and stir with a spatula, from time to time, during the cooking.

If white lead is used, it will be seen in following the operation with a thermometer that the oil will begin to foam at about I00 degrees centigrade. This foam is often very abundant and this is due to the presence of water in the lead (which is hydrated). This phenomenon does not occur with litharge.

Between 180 and 200 degrees centigrade, the oil begins to smoke and to become brownish in color. This is the sign of the beginning of its combination with the lead. The litharge, more than the white lead, has a tendency to agglutinate at about I50 degrees, and the spatula will stick in it at the bottom of the vessel. Towards 2I0 degrees, however, this deposit softens and it will finally mix with the oil at about 230 degrees. Towards 250 degrees, the oil takes on the brown and transparent color of coffee and loses the muddy look it had earlier in the process..

If there is no means of gaging the heat, as with a thermometer, it is important to see that the oil smokes only very slightly. Too long and too hot a cooking (as high as 280 degrees) will lend to a sort of rubberiness in the oil and the product will be unfit for use in painting.

S. German, *Liber illuministrarum*(1512)(Nadolny, 2008)

Quot10- *Ground recipe for walls or wood to be painted in oils*. First "moisten" the walls and wood with oil that has been ground with **red lead**, so that it driers better.

Tegernsee manuscript (Kneepkens, 2012)

Quot11- Minium (red lead oxide) is specifically named as a dryer, albeit for a paint that was used on walls and wood (presumably wood for building, not panels).

Vasari,*Vite*, 1550 (Stols-Witlox*, et al.*, 2008) p. 79

Quot12- *Imprimatura*: […] a composition must be made of pigments with siccative qualities such as **lead white**, **lead-tin yellow**, and "earth from bells" [terra da campane], all thoroughly mixed together and of a single tint, and when the size is dry this must be plastered over the panel and then beaten with the palm of the hand, so that the mixture becomes evenly united and spread all over, and this is what many call the " imprimatura".

17th Century

Pacheco 1564–1654(Merrifield, 1999) p. ccxli

Quot13- Others added linseed oil which has been suffered to fatten by mixing with it **red lead** in powder.
Dryer for carmine: either ground glass or **litharge** in powder, or a little of the fat oil (with **minium**)

Pierre Lebrun, *Brussels manuscript, "Recueil des essays des merveilles de la peinture »*, 1635 (Merrifield, 1999) p. ccxxxviii and p. 816-818

Quot14- 23. *Drying oil* (huile siccative) is made by putting some nut-oil into a pipkin, into which is put a rage containing umber and **minium**, which is suspended to the handle of the pipkin, and then boiled.

Quot15- 24. *Fat oil*(huile grasse) is made by putting a bag of **litharge** into a pipkin with oil, and boiling it.

Quot16- 25. Or the **litharge** may be ground on the porphyry with oil made into a little ball and dried. When it is wanted for use it is boiled until the litharge is dissolved and when cold the oil becomes as clear as rock-water.

Quot17- 26. This oil is very good for drying the colours which do not dry, such as common lake, fine lake, **white lead**, black, ochre, [?], and other similar, which dry slowly.

Baldinucci(Merrifield, 1999) p. ccxxxxix

Quot18- *Olio cotto* was prepared by boiling linseed or nut-oil, either alone, or with **litharge** or glass, finely ground with water. It is stated by this author to have been used to temper those colours which are slow in drying, such as lake, terra nera, bone and other blacks, because both **litharge** and ground glass have the property of making them dry quickly. Oil, boiled without either of these ingredients, as used to accelerate the drying of those colours, which dry well of themselves, such as **white lead**, **minium**, terra verde, umber, cinnabar, smalti, and others; but if used with white lead it would become yellow. "*Pure boiled oil*", continues Baldinucci,"when it is prepared with very clear oil, is also used by painters instead of varnish in the darkest shades, and where the colours have sunk in. And remember, that raw nut and linseed oil are by nature drying, but they do not dry so soon as when boiled, and especially as when mixed with ground glass and litharge."

Paduan Manuscript (Merrifield, 1999) p. ccxxxviii and p. 692

Quot19- 96. *Mode of gilding and painting on glass*. - First grind the colours with boiled oil, that is, oil prepared in this manner. Take half a pound of **litharge of gold** and one pound of nut oil; grind the **litharge** and put the whole into a varnished pipkin, and boil the oil until it is reduced to two-thirds of its previous quantity. Preserve it for painting on glass.

To make the mordant for gilding on glass, take equal quantities of **white lead**, terra gialla, and **minium**; grind the whole together with nut oil, and mix what is ground in a shell or vase with oil boiled as above, boil the whole together a little, and then use it.

Volpatto Manuscript (Merrifield, 1999) p. ccxxxviii and p. 740

Quot20- S. *How is boiled oil prepared?* (olio coto)

F. The linseed oil is put into a clean pipkin or saucepan with some **litharge of gold**, which is tied up in a rag, and fixed to a small piece of wood, which being laid across the pipkin or saucepan, suspends the rag so that it does not touch the bottom, because, if it should touch, it would burn, and the oil would become black, and when the oil boiled it would rise to the brim of the pipkin and flow over, but when the litharge is suspended these effects are not

so easily produced. If the oil is boiled very much it will be more drying, and so whether you use much or little litharge; you may also boil with it a little umber, this will have the same effect, except that the oil will not be so light coloured. [...]

J. Smith, *The art of painting in Oyl*, 1687 (White & Kirby, 1994)

Quot21- John Smith obtained a pale, clear oil by adding two ounces of **litharge** to a quart of linseed oil and leaving it in the sun for a month, stirring it twice a week.

Quot22- The other method of bodying the oil was to heat it, usually with the addition of **lead salts** or other driers. John Smith, for example, recommended heating the oil with the finely powdered **litharge** for one hour until it was "almost of the thickness of Treacle".

Quot23- CHAP. VII. *How to make Gold size to lay Gold on when you guild*.: Gold size is made of fat Oyl, and Yellow Oaker; the Oyl is no other than Linseed Oyl thus ordered; Take what quantity of Linseed Oyl you judge will serve your turn, put it in a Brazen or other Vessel that will endure the fire; when it is in the Vessel, put to it a certain quantity of **Red-Lead****; the more you put in, the better will your Oyl be (provided you put not in so much as to hinder its boyling) for this Red-Lead adds a drying quality to the Oyl, which otherwise being thus ordered, would not dry in any time: when the Oyl and Lead are thus mingled together, let them gently boyl over a fire of coals without flame a pretty while; when it's boyled enough, (which you may know by taking a little of it, and let it cool, and if it roape like thin Treacle, then it is enough) then with a lighted paper set it on fire, (which fireing will burn away much of the greasiness of it) which let burn about a minute or two, or more or less, according as your quantity of Oyl is; and then let it be extinguished (by clapping a Cloath over it) afterwards let it stand to cool and settle; and when all the Lead be sunk to the bottom, and the Oyl be clear, then pour it off, and reserve it in a Bladder for use.

** Red-lead: translated by White as litharge. Red-Lead, Is a Sandy colour, not to be ground very fine on a stone: The onely way to make it fine is by washing (which shall be shewed afterwards): This Colour is an exceeding great dryer and binder, for which purpose it's many times mixed with other Colours (such as will bear it) to make them dry speedily: 'Tis a Colour that resists the weather as well as any colour whatsoever, if it have the same advantage in working. It's of it self an Orange colour, and is the onely Colour used in making of drying and fat Oyls.

Edward Norgates, 1650, quoted by De Mayerne

Quot24- Recommend that **lead white** should be mixed with the oil, together with breadcrumbs and sawdust (...) stirring the ingredients together five of six times a day for the four or five days that the oil was to be left in the sun.

Father Lana (cited in Volpato manuscript), 1670 (Merrifield, 1999) p. 746

Quot25- The directions as to "oiling out" are precise. Lana recommends for this purpose "boiled linseed oil, that is to say, linseed oil to which have been added two ounces of **litharge** for each pound of oil, warmed until it begins to boil". "This application", he says, "is not injurious to the picture, as some have imagined; and the advantage is that it dries quickly, for raw oil is a long time in drying."

Quot26- According to P. Lana, "the priming consists in covering the picture with some colour, which is usually umber finely ground with a little **white lead** and terra rossa, in linseed oil [...].

De Piles, *L'Idée du Peintre Parfait*, 1677 (Merrifield, 1999) p. ccxxxxix

Quot27- *Drying oil*: nut-oil boiled with **litharge** and sandarac; identical with the old "vernice liquida"

Willem Beurs,*De groote Waereld in het Kleen Geschildert,* 1692 (Noble*, et al.*, 2008) p.76

Quot28- One first lays on the panel a ground of weak glue mixed with white chalk to cover the grain of the wood; after this one must scrape it off and make the panel smooth and even, taking care that the grain remains filled. After this one grinds umber and **lead white** very thickly with oil and applies it to the panel first with a knife, after which one smoothes it with the hand [...]

18th Century

Fra Fortunato, 1711 (Merrifield, 1999) p. ccxxxxix

Quot29- The recipe for "*olio cotto*" given by Fra Fortunato , differs from these recipes [Lebrun, Paduan] in directing the addition of water, which is to be boiled with the litharge and oil, which he says cause the oil to become as clear (colourless) as water itself.

Dossie, R.,*The handmaid to the arts*, 1758. vol. I. London: J. Nourse. (Carlyle, 1999)

Quot30- *Drying Oil Recipes*: describes the preparation of drying oil (mainly intended for "coarser work") as involving boiling or simmering the oil in conjunction with a variety of driers used at once, including **litharge**, **white lead**, **red lead**, **lead acetate**, and sulfate of zinc.

Williams, W.1787,*An essay on the mechanic of oil colour*. Bath, England: W. Williams. (Carlyle, 1999)

Quot31- *Raw oil clarification*: recommends adding 1 part **litharge** to 2 parts oil and shaking them together over several days. This would cause "a tallow-like grease" to be thrown down with the litharge.

Palomino (Merrifield, 1999) p. ccxli

Quot32- Describes a drying oil for blues and whites composed of ground glass, **litharge**, **white lead**, and **red lead**, of each one ounce, and half a pound of oil boiled for a short time together in a water bath.

19th Century

Chilone (old Venetian painter, died 1834 or 1836)(Merrifield, 1999) p. ccxl

Quot33- Canal [1703-1767] and Canaletto [1697-1768] used oil boiled on **litharge**, which they recommended [Chilone] to use also, and that they frequently spread it over the whole picture.

Field, G., *Chromatography; or, A treatise on colours and pigments, and of their powers in painting*. 1835., London: Charles Tilt. (Carlyle, 1999)

Quot34- *Raw oil clarification*:Same method [as Williams 1787, quot31] but calls for 1 part **litharge** to 8 parts oil

Quot35- Japanner's gold size was made by boiling oil with a number of driers in combination such as **litharge**, **red lead**, umber, zinc sulfate, and manganese.

Roberson Archive, Hamilton Kerr Institute, Cambridge University, Whittlesford, Cambridge, England. Recipe book HKI MS 788–1993.(Carlyle, 1999)

Quot36- *Raw oil clarification*:Another recipe for preparing oil, entitled "Liebig's," was found in one of Roberson's recipe books and involved combinations of driers such as lead acetate and **litharge** with water.

Sully, T. *Hints to young painters, and the process of portrait painting as practiced by the late Thomas Sully*. 1873, Philadelphia: J. M. Stoddard & Co. (Carlyle, 1999)

Quot37- *Drying Oil Recipes*: One of the preferred methods was to use **metallic lead**, either by agitating the oil with lead shot or by "grinding" the oil with a leaden pestle in a lead-lined mortar. Prolonged storage in a **leaden vessel** was also recommended. **Litharge** (and very occasionally **white lead**) was sometimes substituted for **metallic lead** and used in a similar manner. For example, Rembrandt Peale's method was to add 2 tablespoons of **litharge** to and 8-ounce phial which was then filled with linseed oil. It was to be kept exposed to the sun or near a fire for a few days and shanken frequently.

Winsor and Newton Ground preparation, 1871 (Carlyle*, et al.*, 2008)

Quot38- The study of the Winsor and Newton archive revealed one comprehensive historic commercial recipe for ground preparation, dated from 1871. It is presented in details and reproduced in (Carlyle*, et al.*, 2008). The recipe consists in three ground layers.

The 1st Colour is made of "a putty of chalk mixed with an oil preparation containing linseed oil, "varnish bottoms" (including lead-treated drying-oil), and patent dryer". The 2nd and 3rd ground layers are made of this 1st putty, mixed with **lead white** in oil and a patent dryer, in different proportions. The 1st Colour oil, and in particular the "varnish bottoms" it is made of, were reproduced based on other recipes from the archive, in particular the "Extra Boiled Oil" recipe, dated from 26 April 1850 and described as "Much liked for canvass works". It is produced by heating oil to 165°C, adding dryers (**litharge** first then **red lead**, then umber), heating gradually to a top temperature of 243°C, allow to cool to 165°C, and leaving standing one day before drawing off. Several recipes for "Patent dryer" are mentioned in the archive. The one chosen for the reproduction was the one labeled "Extra ground for canvas use". It is composed of **sugar of lead** (lead acetate trihydrate), barium sulphate, whiting [chalk], **powdered litharge**, zinc sulphate and linseed oil.

Mitsuda-e 密陀絵: Lit. Litharge painting. A modern term for a type of ancient oil painting that uses lead oxide, or *mitsudasou 密陀僧, as a desiccant. The medium consists of powdered pigments added to a base of perilla oil and a small amount of lead-oxide that has been heated with the oil. The technique, which may have originated in Persia, was used in China at least from the Han dynasty. Transmitted from China to Japan in the 7c, *mitsuda-e* was used on wood and leather. Its use was prevalent in the Nara period because it easily produced the color white, this not being possible in lacquer painting *urushi-e 漆絵. The technique is also called *yuga* 油画, which some scholars include, along with *yushoku* 油色 (paintings coated with perilla oil), as a type of *mitsuda-e*. Examples of *yuga* include the Tachibana Shrine *Tachibana fujin no zushi* 橘夫人厨子 (early 8c) and according to some scholars, the Beetle wing Shrine *Tamamushi no zushi 玉虫厨子 (mid-7c), both in Houryuuji 法隆寺, Nara. The craftsmen of the *Tamamushi no zushi* seem to have used the technique in combination with lacquer painting. Examples of *yushoku* include a *biwa* 琵琶 in the *Shousouin 正倉院, Nara (see * kanpachiga 捍撥画). Although examples of *yushoku* also exist from the Kamakura and the Momoyama periods, there are very few examples of *yuga* dating after the Heian period. During the Momoyama and early Edo periods, however, *yuga* was used extensively in the decoration of lacquerware. *Mitsuda-e* lacquerware are presently produced in such places as Jouhana 城端 in Toyama prefecture and Okinawa.[1]

Recipe for «simple plaster » (Diderot & d'Alembert, 1755), translated in English by the authors

The wonderful aspect, or better the simplicity of this process relies in this: oil and litharge are treated as in a water-bath, and this with water which makes the bath present in the same vessel as the materials it heats; it is indeed useless to put it in a separate vessel, because it does not have any chemical actions on these materials. Yet it is important to expose these materials just at the level of heat, because part of oil could be burnt if the temperature was higher, leading to formation of coal, and calcined white lead [litharge] could be reduced, or at least blackened: both effects would affect the plaster elegance, assuming that elegance would not depend on blackness; because rules are here rather weird & rather arbitrary. A plaster belonging to the class we are talking about here would fail if lead was burnt; the black plaster or of burnt ceruse, & the "onguent de la mere" (which is a plaster), would conversely fail if they were not burnt. […] I assume that my readers do not ignore that oil does not boil at the degree of boiling water, & and that each time two immiscible liquids are mixed together, whatever the proportion, and exposed to fire, the heat in the entire mass can never go higher than the highest degree reachable by the most volatile liquid, or by the one of the two of which the extreme degree of heat is the lowest, caeteris paribus ; that, consequently in the present case, oil can reach only the degree of heat of boiling water. Second, it is better to apply boiling water immediately, than to insert a vessel between this liquid and the materials to be mixed; because, in addition to the fact that this method is easier & faster, it acts also thanks to the fact the water boiling shakes the plaster mass in all its parts, & contributes very efficiently to the movement which is done by brewing; movement which speeds up all the dissolutions.

[1] From JAANUS web page, April 2016: http://www.aisf.or.jp/~jaanus/deta/m/mitsudae.htm

Le merveilleux, ou plutôt le beau simple de cette opération, consiste en ceci : on traite proprement l'huile & la litharge au bain-marie, & cela, quoique l'eau qui fait le bain soit contenue dans le même vaisseau que les matières qu'elle échauffe ; & il est inutile en effet de la placer dans un vaisseau séparé, parce qu'elle n'a aucune action chimique sur ces matières. Or il est utile de ne les exposer, ces matières, qu'à ce degré de chaleur, parce qu'une partie de l'huile pourroit être brûlée à un degré de feu supérieur, & fournir par conséquent du charbon, & la chaux de plomb être réduite, ou du moins noircie : l'un & l'autre inconvénient ôteroit à l'élégance de l'emplâtre, supposé toutefois que l'élégance ne dépendît pas de la noirceur ; car les lois sont ici fort bisarres & fort arbitraires. Un emplâtre de la classe de ceux dont nous parlons ici seroit manqué, si on brûloit le plomb ; l'emplâtre noir ou de céruse brûlée, & l'onguent de la mère (qui est un emplâtre), seroient manqués au contraire, si on ne le brûloit pas. […] Je suppose que mes lecteurs n'ignorent pas que l'huile ne bout point au degré de l'eau bouillante, & que toutes les fois que deux liquides immiscibles se trouvent confondus en quelque proportion que ce soit, & exposés au feu, la chaleur ne peut jamais s'élever dans la masse entière au-dessus du plus haut degré dont est susceptible le liquide le plus volatil, ou celui des deux dont le degré de chaleur extrême est le plus foible, caeteris paribus ; que par conséquent dans le cas dont il s'agit, l'huile ne peut contracter que le degré de chaleur de l'eau bouillante. Secondement, il vaut mieux appliquer l'eau bouillante immédiatement, que d'interposer un vaisseau entre ce liquide & les corps à unir ; parce qu'outre que cette méthode est plus commode & plus courte, elle sert encore, en ce que le bouillonnement de l'eau agite la masse de l'emplâtre dans toutes ses parties, & concourt très-efficacement au mouvement qu'on se propose d'exciter en brassant ; mouvement qui hâte toutes les dissolutions.

Description of the samples

"Huile de litharge"

"Huile de litharge"(cf. Fig. 4b and spectrum Fig. 5b) was prepared following Folio 28v, De Mayerne:home-made walnut oil, water and PbO (VWR, Fontenay S/Bois, France) are mixed in the mass proportions 4 : 4 : 1. PbO is first mixed with oil for 40 min at 60 ◦C. Then, boiled water is added and temperature is increased to 100 °C for 2 h under constant stirring. The precise description and analysis of this reproduction can be found in (Cotte*, et al.*, 2006).

Mock-up painting sample

The mock-up painting was prepared using a similar "huile de litharge", but prepared with a poorly grinded PbO. It was then mixed with ochre. This mixture was applied on a canvas and partially covered with a brown colored layer made of the same lead-medium mixed with azurite, vermilion and raw umber (Cotte*, et al.*, 2007). A single layer of lead-medium mixed with ochre was also applied on a Melinex polymer foil. The protrusion studied here was sampled from this "Melinex mock-up" after 9 years of conservation at room conditions (in the dark) and prepared as a thin (4µm) section with a microtome using a non-embedding protocol (Pouyet*, et al.*, 2014). For µFTIR analyses, the sample was put horizontal on a 0.4mm BaF_2 window. For µXRD analyses, the same sample was mounted vertically, sandwiched between two ultralene foils (4 µm thick, from Spex Sample Prep).

"Walnut oil – litharge" system

The sample shown in Fig. 5c-e and Fig.6 was prepared by depositing a droplet of home-made walnut oil on a 0.4mm BaF_2 window. PbO powder was gently deposited on the droplet. The sample was left in room conditions and analyzed at t = 0and t = 1 month.

Description of the instruments

Bulk ATR/FTIR measurement

FTIR spectrum of "huile de litharge", at the end of its preparation,was obtained using an Impact 420 (Nicolet) with an ATR accessory in diamond. Acquisitions were performed at room temperature. The spectrum is the result of an accumulation of50 scans with a spectral resolution of 8 cm^{-1}(Cotte*, et al.*, 2006).

Micro FTIR measurements

µFTIR analyses were carried out at the ID21 beamline, ESRF (Susini*, et al.*, 2007). Measurements were performed in transmission mode using the Globar source for low resolution (20×20µm²) and the synchrotron light for high resolution (7×7µm²) mapping. For a detailed description please cf. (Cotte*, et al.*, 2007). FTIR maps were processed using the PyMca software (Sole*, et al.*, 2007).

Micro XRD measurements

µXRD maps were acquired at two different beamlines, ID13 and ID21, both giving similar results. In both cases µXRD maps were acquired in transmission with a monochromatic beam by raster scanning the protrusion thin section in air. Data were analyzed using the PyFAI (Ashiotis*, et al.*, 2015)and the XRDUA (De Nolf*, et al.*, 2014) software packages.

At ID13, measurements were carried at the "microbranch" second experimental hutch). The energy of the incident beam was 13.6keV; the X-ray beam was focused to 2 (ver.) × 3 (hor.) µm² using

compound refractive lenses (56 Be-lenses with 200 μm radius of curvature at the apex) which are mounted in a transfocator. The XRD patterns were collected using a MAXIPIX 2x2 detector (516×516 pixels of 55μm, exposure time 0.03s). The XRD map has been recorded via raster-scanning with stepper motor based y-z scanning stages (raster 2x2μm^2,601x151 sampling points, positioning resolution 0.5 μm).

At ID21, measurements were carried out at the newly developed μXRD/μXRF side-branch end-station. The energy of the incident beam was 8.5keV; the X-ray beam was focused to 0.7 (ver.) × 1.7(hor.)μm^2 using a Kirkpatrick-Baez mirror system and XRD patterns were collected using a FReLoN camera (2048x2048 pixels of 51μm, exposure time 4s).

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