



Metallurgical processes for the recovery and recycling of lanthanum from various resources—A review



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ABSTRACT

Various processes for the recovery of lanthanum from metallurgical and non-metallurgical resources are reviewed. Lanthanum is present as a major constituent of monazite, fluorocarbonate ores, bauxite and associated residues, tailings and waste liquors of rare earths, aluminium, etc. Phosphate rock, optical glasses and Ni-MH batteries serve as the most recent and high concentration secondary resources of lanthanum. This review discusses the various methods of hydro-/pyro-metallurgical origin, leaching, solvent extraction, precipitation and other pre-treatment options for lanthanum recovery. A separate section on recovery of lanthanum by selective biosorption from various model and actual solutions is also included. The need to develop new extraction systems with high lanthanum selectivity in the areas of leaching and solvent extraction are probable improvement trend for the future.

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1. Introduction

Lanthanum is light group rare earth element with atomic number 57 and atomic weight 138.9055 was discovered by Sweden chemist Carl Gustav Mosander in 1839 (Gupta and Krishnamurthy, 2004). The chemistry of lanthanum culpably involved its formation of the ionic +3 oxidation state and its higher basicity compared among 17 rare earth elements which determines the extent to which cation hydrolyzes in aqueous solution and relative stability of salts and by exploiting this chemical feature, it can be separated by a variety of techniques like solvent extraction, ion exchange, fractional precipitation methods (Yu, 1995; Chi and Xu, 1999; Vierheilig, 2014; Xie et al., 2014a, 2014b; Weeden et al., 2015). There are no ores which contain only lanthanum as the metal component, it is found in minerals that include all the lanthanoid elements. It mainly occurs in monazite (Ce, La, Y, Th)PO₄ and bastnäsite (La, Ce)FCO₃ ores in appreciable quantities (Clark, 1984; Lottermoser, 1992; Chi and Tian, 2007). Whilst lanthanum found in traces in the ores of aluminium, iron, zinc and gold (Maksimovic and Panto, 1991; Frietsch and Perdahl, 1995; Xu et al., 2006; Li et al., 2009; Kuzmin et al., 2012; Wang et al., 2013; Ismail et al., 2014; Jaireth et al., 2014). Monazite deposits in India, Malaysia, Australia, Brazil, China, South Africa, Sri Lanka, Thailand, and the United States, constitutes the second largest segment while bastenite deposits constitutes larger portion of world's rare earth resources, occurs mainly in China and

Australia (Clark, 1984; Wood, 1990a, 1990b; Chi et al., 2000a, 2000b; Noumav, 2008). In India, Monazite is found to be association with heavy minerals like ilmenite, rutile, zircon etc. in concentrations of 0.4–4.3% of total heavies in the beach and inland placer deposits of Andhra Pradesh, Bihar, Kerala, Odisha, Tamil Nadu, West Bengal. Although Bastnäsite has been reported in association with carbonatite from Purulia distt, West Bengal and monazite has been reported from carbonatite occurrence of Sung Valley (Meghalaya), Sevattur (Tamilnadu) and Samchampi (Assam), by geological survey of India (Gupta and Krishnamurthy, 2004).

Lanthanum is the 28th most abundant element within in the earth crust (~32 ppm), although exists in soil (~26 ppm, ranging from 1 to 120 ppm), seawater (~5 ppt) and nil in atmosphere (Henderson, 1984). Lanthanum content in unoxidized ores ranges from 20 to 100 ppm and averages 30 ppm, its direct variation in content with potassium and aluminium suggest that the lanthanum is concentrated in clay intensively exists in concentrates of carbocaceous materials from host rocks and unoxidized ores ranged from 160 to 222 ppm (Radtke, 1936; Gupta and Krishnamurthy, 2004). The main ores from which it is extracted today are monazite and bastnäsite, where lanthanum makes up 25% and 38% of the minerals, respectively (Gupta and Krishnamurthy, 2004). Allanite and cerite also contain lanthanum, but these ores are not mined for their lanthanum content in particular (Table 1). The main mining regions of the aforementioned ores making up the world production of lanthanum exceeding over 12,500 mt/year. The reserves of this element are thought to be in the region of 6 mt. Despite being one of the rare earths, lanthanum is probably one of the least rare, occurring in a tonnage similar to that of lead and tin combined

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Table 1
Prevalent mineral resources of lanthanum with % La content and occurrence.

Mineral	Chemical formula	% La content	Occurrence	References
Monazite	(La, Ce, Nd, Th)·PO ₄	25	India, China, Korea, U.S.A., Malaysia, Australia, Brazil, South Africa, Sri Lanka, Thailand	Maksimovic and Panto (1991); Frietsch and Perdahl (1995); Xu et al. (2006); Li et al. (2009); Kuzmin et al. (2012); Wang et al. (2013); Ismail et al. (2014); Jaireth et al. (2014)
Bastnäsite – La	(La, Ce)·CO ₃ F	35	China, Australia	Clark (1984); Wood (1990a, 1990b); Chi et al. (2000a, 2000b)
Fergusonite – Ce	(Ce, La, Nd)·NbO ₄ /Ce _{0.6} La _{0.3} Y _{0.1} NbO ₄ ·0.3(H ₂ O)	15–24	Northern China, Ukraine	Gupta and Krishnamurthy (2004)
Gadolinite – Ce	(Ce, La, Nd, Y) ₂ ·Fe ²⁺ ·Be ₂ Si ₂ O ₁₀	10.9–14	Norway	Gupta and Krishnamurthy (2004)
Loparite	(Ce, La, Na, Ca, Sr)·(Ti, Nb)·O ₃	11.89–13.84	U.S.A., China, Russia	Chakhmouradian et al. (1999); Rofer and Kaasik (2000); Gupta and Krishnamurthy (2004)
Parisite-Ce	Ca(Ce, La) ₂ (CO ₃) ₃ F ₂	23.27	Russia, Brazil	Zaitsev et al. (1998); Manfredi et al. (2013); Gupta and Krishnamurthy (2004)
Yttracrite	(Ca, Ce, La, Y)·F ₃ ·xH ₂ O	8.5–11.5	U.S.A.	Gupta and Krishnamurthy (2004); Sverdrup (1968)
Allanite	(Ca; La) ₂ (Al, Fe ²⁺ , Fe ³⁺) ₃ (SiO ₄)(Si ₂ O ₇)O(OH)	0.075–7.17	India, U.S.A.	Abhilash et al. (2014); Jordens et al. (2014)
Stavelotite-(La)	La ₃ Mn ²⁺ ₃ Cu(Mn ³⁺ , Fe ³⁺ , Mn ⁴⁺) ₂₆ [Si ₂ O ₇] ₆ O ₃₀	1–2	Belgium	Bernhardt et al. (2005)
Rhabdophane	(Ce, La)PO ₄	1.8–2.0	USA	Clark (1984)
Hornblende	(La, Ce, Ca, Na, K) ²⁻ ₃ (Mg, Fe ²⁺ , Fe ³⁺ , Ti, Al) ₅ (Si, Al) ₈ O ₂₂ (OH, F) ₂	5	USA	Aide and Aide (2012)

(Golev et al., 2014; Goonan, 2011a, 2011b; Massari and Ruberti, 2013). Lanthanum metal itself is produced by reacting lanthanum fluoride and calcium metal, however lanthanum is only commercial in its other forms such as, lanthanum oxide, carbonate, chloride, fluoride, nitrate and sulphate (Gschneidner, 1980; Gschneidner et al., 2002). But, the high price of lanthanum twinned with absence of reliable sources and long term production has limited its commercial applications. Lanthanum is a rare and expensive metal because of its scarce distribution and difficulties associated with its extraction (Stwertka, 2012; Emsley, 2011; Gray, 2012; Binnemans et al., 2013).

There are no such commercial uses for pure lanthanum metal, but there are for its alloys, although its compounds have been found wider applications in automobile catalytic convertors, ceramics, fluid catalytic cracking, glass additives, battery alloys, phosphors, glass polishing industries. The properties of lanthanum led to exploitation in many areas like Ni-MH batteries, high strength and conductive copper alloys, potentiometric chlorine gas sensors, high temperature lanthanum zirconium coatings, lanthanum carbonate as a medication, petroleum cracking catalysts, electron dense tracer in molecular biology, LaB₆ in electron microscopes, La–Ba radiometric dating to estimate age of rocks and ores, in carbon arc lamps, additive in liquid phase sintering, optical glasses (~40% usage of lanthanum oxide), lanthanum hexa-aluminate as thermal barrier coating for gas turbine application, lanthanum-chromite based materials for solid oxide fuel cell interconnects, as alloying element in many Bi–Te, Bi–Se–Te, Mg alloys, in the zeolite catalysts used in petroleum refining, in hybrid cars (Hendrick, 1985; Nakai et al., 1988; Chau and Lu, 1995; Gadow and Lischka, 2002; Kim and Shim, 2003; Fergus, 2004; Gupta and Krishnamurthy, 2004; Ji et al., 2005; Shijie et al., 2006; Tliha et al., 2007; Emsley, 2011; Dziubaniuk et al., 2012; Dejiu et al., 2013; Di Girolamo et al., 2014). Lanthanum hydroxide is used for adsorption of phosphates from waste water under the influence of chlorides, sulphates, nitrates and hydrogen carbonate and even lanthanum phosphates monoliths act as efficient perchlorate adsorbents (Sankar et al., 2014; Xie et al., 2014a, 2014b). The applications of lanthanum are increasing because of its significant chemical properties over other metals.

The recovery of lanthanum from its ores in substantial quantity usually involves pyrometallurgical, hydrometallurgical process or combined pyro–hydro metallurgical process. However, the energy consumption is higher in pyrometallurgical processes. Lanthanum found ameliorated in slags, residues, tailings and primarily produced as a by product during processing of various ores. Currently, hydrometallurgical processes, which mainly involve leaching, solvent extraction

and precipitation are most commonly used for lanthanum recovery. In this review, the metallurgical processes involving the intricate ways to recover lanthanum from various resources, depending on the different types and amounts of impurities are reviewed in consideration of selecting processes to recover lanthanum and incorporating the process for the production of the metal in bulk quantity. The selectivity of extraction can be improved using biosorption as a technique comparative to solvent extraction and co precipitation, which also reviewed in present work.

2. Metallurgical process for the recovery and recycling of lanthanum from various sources

2.1. Monazite

Monazite is a phosphate mineral which constitutes 10–40% La along with noteworthy elements like Ce, Sm, Pr and Nd, 4–12 wt.% thorium and a variable amount of uranium. Monazite is found throughout the world in placer deposits, beach sands, and is also a component of the Bayan Obo deposit (Thompson, 1980). In India, the estimate of monazite is 10.21 million ton (mt) in 2005 which has gone up to 11.93 mt in 2013. Monazite having high chemical and thermal stability, when attacked by strong hot acid led to transformation of phosphate ion to di-hydrogen phosphate salt and phosphoric acid, bequeathing the metal ions as water soluble salts which spifficate the phosphate matrix present in the mineral (Jiang et al., 2005). Metallurgical processing of monazite results in pulverization of rare earth complexes due to formation of phosphides. So, the separation of individual metal led to an essential requirement to remove phosphorus from the ore, which decompose at higher temperature due to their high thermal stability. Decomposition of monazite studied with the influence of 10% mixture of NaCl–CaCl₂ was found to be 79% at roasting temperature of 750 °C in 1 h; whereas charred coal at 1400 °C in 2 h led to 98% dephosphorization efficiency, even CaO in CaO–CaCl–CaCl₂ has ability to decompose monazite in temperature range varying from 610 to 700 °C (Shuchen et al., 2007; Wenyan et al., 2006).

Lanthanum recovery from monazite was investigated using Na₂CO₃ at temperature of 900 °C in both reducing and sulphidizing environment, in which extraction was made from water leached residue that selectively leaches the oxysulphide phase formed with 3.5 wt.% hydrochloric acid (Merritt, 1990). Chi et al., 2000a, 2000b reported a novel method for the recovery of RE containing lanthanum from weathered black earth slime using SO₂ leaching followed by ammonium chloride

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