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Calcium Sulfo Aluminate (CSA) Cement from Coal Ash Utilized as Barriers for Radioactive Waste Disposal

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Abstract

In this paper, we reviewed briefly about the barrier materials for the radioactive waste disposal. The primary concept of the radioactive waste disposal is safety. The goal of the radioactive waste management is to assure that the environment is not adversely affected and also public. There are a wide variety of materials are available for the radioactive waste disposal or storage. Among those coal fly ash is one of the significant materials are used as a barrier material. Here we reported, the Calcium sulfo aluminate (CSA) from coal fly ash is effectively suitable for the radioactive waste disposal. This is one of the ways of utilization of waste and manufactured the valuable materials for future indeeds.

Key words : Radioactive waste, disposal, calcium sulfo aluminate (CSA), coal fly ash

1. Introduction

Currently, nuclear power is the major source for electricity generating technology, and it's sharing the 14% of the world's electricity, and 21% of the electricity in OECD countries. [1)] In 2015, 68.5% of global electricity production was from the nonrenewable fuel source. Among these, 16.0% of hydroelectric plants, 10.6% of nuclear plants, tide, solar, geothermal, wind, and other sources 4.9%, and biofuels, waste, etc. made up the remaining 2.2% (Fig.1a and b).[2,3)] After Fukushima Daiichi incident, the nuclear energy growth has been much lower.

By 2035, 70% of the global nuclear energy production is expected to rise, particularly China, India and Korea are leading growth. Regarding the amount of low nuclear capacity falls from 393 GW to 335 during the years from 2010 to 2035. International Energy Agency (IEA), thinking the falling scenario of nuclear capacity impacts on the energy security, diversion of the fuel sources, energy imports and exports and CO_2 emissions from power plants.

Currently, nuclear energy plays a significant role in the CO_2 emissions reduction from the energy sector nearly 1.3 ~ 2.6 gigatonnes (Gt) annually, considering an alternative source of natural gas or coal. Due to the nuclear power, more than 60 Gt CO_2 has been avoided since the 1960s (Fig. 2) [4)]. Globally, 13% of the emissions reduction required in the power sector with the participation in different countries such as Republic of Korea (24%), European Union (23%) and in China (13%). The well-known fact is nuclear energy clearly attributed a significant role in providing low-carbon electricity generation throughout the globe.

The amount of waste generated from nuclear power plants which are nuclear substances or residues of

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the burning of nuclear fuels in the reactor. It contains radioactive materials and leads to the acute radiation impacts on the public health. The nuclear waste has been segmented into three types such as low level radioactive waste (LLRW), intermediate level radioactive waste (ILRW) and high level radioactive waste (HLRW) (Table 1).

Based on the figure, nuclear waste management requires for safe radioactive disposal or storage. Before the disposal of nuclear waste, it should minimize the radiation level by keeping the cans in under water for suitable storage level. After this minimization of radiation then, final disposal of radioactive waste is done. Recently, Swedish national council published several reports on the nuclear waste.[5-9]. The IAEA estimates[10] that over 80% of all lower level radioactive waste and very low levels radioactive waste produced for disposal. For intermediate level radioactive waste, 20% is in disposal estimated by the agency, with the suitable balance in storage (Table-2).

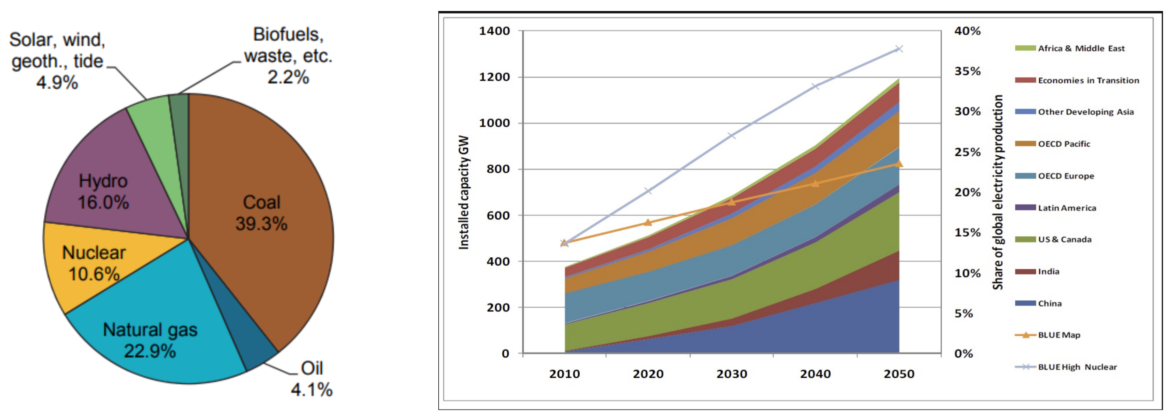

Fig. 1. (a) World gross electricity production by source, 2015 (b) IEA nuclear roadmap 2010 share of nuclear.(adopted from ref.2&3).

Table-1. Different categories of radioactive waste

Low Level Waste(LLW)	Intermediate Level Waste(ILW)	High Level Waste(LLW)
Sources: nuclear facilities, and is mainly paper, plastics and scrap metal items. A sub category of LLW is a very low level waste, building rubble, soil and steel items such as framework, pipework and reinforcement from the dismantling and demolition of nuclear reactors and other nuclear facilities and the clean up of nuclear sites. This comprises small volumes principally from hospitals and universities that can be safely disposed of with municipal, commercial or industrial waste (either directly or after incineration).	Sources: The major components of ILW are metal items such as nuclear fuel casing and nuclear reactor components, graphite from reactor components, graphite from reactor cores, and sludge's from the treatment of radioactive liquid effluents. Wastes exceeding the upper boundaries for LLW that do not generate sufficient heat for this to be taken into account in the design of waste storage or disposal facilities.	Sources: Initially HLW comprises nitric acid solutions containing the waste products of reprocessing spent nuclear fuels. Wastes in which the temperature may rise significantly as a result of their radioactivity, so this factor has to be taken into account in the design of waste storage or disposal facilities.

Table 2. Nuclear waste inventory (IAEA estimates, 2016)[10]

Solid radioactive waste in storage(m^3)	Solid radioactive waste in storage(m^3)	Proportion of waste type in disposal
VLLW 2,356,000	7,906,000	77%
LLW 3, 479,000	20,451,000	85%
ILW 460,000	107,000	19%
HLW 22,000	0	0

All kinds of hazardous waste need keen careful management and disposal, not only radioactive waste. As compared the waste generation from other electricity generation sectors, nuclear power plants produced lower. The average radioactive waste generated in the UK is about 4.9 million tonnes by 2125. The UK has the world's historical and oldest nuclear industries. The annual generation of waste classified as conventional waste (200 Mt) in the UK, among these waste, the hazardous waste is 4.3 million tonnes. **Lower level radioactive is around** 94 %**, 6% is intermediate level of radioactive waste**, and **below** 0.03% is **considered** as **high level radioactive waste**[11].

2. International Atomic Energy Agency (IAEA) Activities Regarding Radioactive Waste Disposal

The **International Atomic Energy Agency (**IAEA**)** activities are mainly focused on **security and** safety (to protect **the public** and environment from radiation **effect**), **verification &** safeguards (to **control** the further **unlimited** nuclear weapons), science **and** technology (to mobilize **the** nuclear technology **for sustainable society**). This includes radioactive waste processing and disposal technologies such as i. developing standard limits and policies; ii. providing evaluation and solutions; iii. Providing infrastructure; iv. Promoting and transferring technology networks; v. conducting forums for increasing transparency, sharing and learning lessons through workshops, communicating, meetings, and social media (Fig.3)[12].

2.1. Radioactive Waste management by U.S. DOE

The U.S. Department of Energy (DOE) focussed on assessment alternatives for the permanent disposal of spent nuclear fuel and high-level radioactive waste [13]. DOE-managed high-level radioactive waste and spent nuclear fuel from defense or DOE research and development activities.

2.2. Radioactive Waste Management in Korea

In South Korea, there are twenty three nuclear power plants are in under operation and five is in under construction and other four is in under contemplation. In Korea, the radioactive waste management facilty was established and low intermediate level waste facility is in under construction and started operation in 2014[14].

The Korean low intermediate level waste repository is in under construction KORAD which is reposnsible for radioactive waste management in Korea is planning to develop the 2nd stage of a radioactive disposal facility in Wolsong site. As of march 2013, the total radiaoctive waste was generated 132, 257 Bq/g. Among these, 68.5 5 from nuclear power plants and 31.5% from other facilities existed. Wolsong site is the radioactive waste disposal facility with engineered barriers. The Korea Institute of nuclear safety (KINS) considered a new radioactive waste classification system for the linkage between the category and radioactive waste disposal type based on the long term safe deposits.

2.3. Radioactive Waste Discharge Standards

National regulatory authorities (NRA) granted to release the lower contaminated effluents to the envi-

Safety Fundamentals	
General Safety Requirements	General Safety Requirements
Vol.1 Governmental and regulatory framework	1.Site evaluation for nuclear installations
Vol.2 Leadership and management for safety	2. Safety of nuclear plants 2.1. Design and construction 2.2. Commissioning and operation
Vol.3 Radiation protection and safety of radiation sources	3. Safety of research reactors
Vol.4 Safety assessment	4.Safety of nuclear fuel cycle facilities
Vol.5 Predisposal management of radioactive waste	5. Safety radioactive waste disposal facilities
Vol.6 Decommissioning and termination of activities	6. Safe transport of radioactive material
Vol.7 Emergency preparedness and response	

Fig. 3. IAEA established safety and security fundamentals (adopted and modified from reference 12, Michael I. Ojovan).

Table-3. The discharge limits set by IAEA for the years 1990-1994) (adopted from ref.15).

Type of Nuclear Power Station	Normalized discharges (Bq/(GW(e)a))						
	Gaseous					Liquid	
	H-3	C-14	Noble gases	I-131	Particulates	H-3	Other
PWR	2.3×10^{12}	2.2×10^{11}	2.7×10^{13}	3.0×10^{8}	2.0×10^{8}	2.2×10^{13}	1.9×10^{10}
BWR	9.4×10^{11}	5.1×10^{11}	3.5×10^{14}	8.0×10^{8}	1.8×10^{11}	9.4×10^{11}	4.3×10^{10}
GCR[2]	4.7×10^{12}	1.4×10^{12}	1.6×10^{15}	1.4×10^{9}	3.0×10^{8}	2.2×10^{14}	5.1×10^{11}
HWR	6.5×10^{14}	1.6×10^{12}	2.1×10^{15}	4.0×10^{8}	5.0×10^{7}	4.9×10^{14}	1.3×10^{11}
RBMK	2.6×10^{13}	1.3×10^{12}	1.7×10^{15}	7.0×10^{9}	1.4×10^{10}	1.1×10^{13}	5.0×10^{9}
FBR	4.9×10^{13}	1.2×10^{11}	3.8×10^{14}	3.0×10^{8}	1.2×10^{10}	1.8×10^{12}	4.9×10^{10}

ronment with the standard limits through the authorized liberate. The International Atomic Energy Agency (IAEA) provided a guide namely WS-G-2.3 and IAEA-TECDOC-1638 for the authorized procedure for the disposal of radioactive waste within the limits and also the IAEA set the ejected limits. In table 3, we reported the discharge limits set by IAEA in the year 1990-1994[15].

3. Cementaneous Materials for radioactive Waste Management

The geological disposal is the final long term safe storage of high level radioactive waste with well-equipped and engineered materials. In general, the geological disposal concerns the facility constructed plant with 300 ~500 meters depth for the disposal of high level radioactive waste[16].

Cementitious materials are used as elementary materials in the geological disposal. In general, these are used as for tunnel thickeners, construction materials, tunnel plugs, backfills, waste packages, sealants and fracture grouts. Low and intermediate level radioactive wastes contain the larger quantities of cementitious materials [17]. Among the several nuclear waste disposal materials, the cementitious materials are at most significant to immobilize intermediate level wastes. Intermediate level wastes may consist Neptunium-237 in the reasonable quantity, which is highly hazards to the environment due to its long half-life period nearly 2.14 · 106 years [18]. The relation between the cementitious materials and re-

Table-5. Calcium sulfo aluminate Advantages

Division	Slump (cm)	Air(%)	Strength 4th (Kg/cm)	Strength 1 day(Kg/cm)
Commercial	22.5	4.8	254	303
Product development	23.0	4.8	244	311

pository components was estimated qualitatively and identified the performance parameters [19].

3.1. Coal Ash as a Barrier Materials

Annually larger quantities of radioactive waste were generated from the energy generating plants and mining sites. The waste must be stored safely for long terms. In general bentonite is used as a sealing material or absorbed material for the storage system. But the high quality bentonite sources are very limited so coal ash used an alternative material for bentonite. Recently, coal ash was utilized as an obstruction material for the radioactive waste disposal[20]. Coal ash has wide applications. In Japan, 75% of the total coal ash was used in cement industry and this is the major utilization in Japan. Clay, Limestone, and iron oxide are the major compositions of cement used for the construction.

Recently, a new approach to coal ash utilization was largely developed. Coal ash as a raw material for cement. However, the recent attention has been raised on developing coal ash utilization technologies as a backfill material, artificial aggregates, roadbed materials, and civil engineering issues.

4. Calcium Sulfo Aluminate from Coal Ash

Calcium sulfo aluminate from coal ash considered as a high functional cement with wide applications (multi stored building construction, multi complex constructions etc). The calcium sulfoaluminate also called

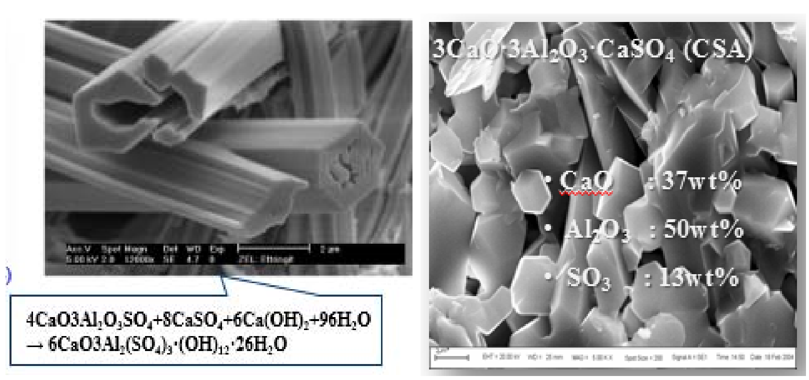

Fig. 4. Calcium sulfo aluminate from coal ash.

Table-4. Calcium sulfo aluminate preparation conditions

Conditions for low grade CSA clinker		
Sintering Temp (°C)	1200 [8 hr]	100 [8 hr]
Raw Materials (ton/hr)	100	
Heat (kcal/kg-cl)	620	500
Recycle rate (%)	25	10
CSA (%)	14-16	14-16

green cement with many benefits and advantages. The aluminate source is such as bauxite and coal ash is the main raw materials to manufacture calcium sulfoaluminate. The CSA cement has good hydraulic reaction properties, immense reaction with water, considerable expansion, good strength, and rapid hardening. It has good mechanical properties such as i. enlargement, ii. quick hardening, iii.durability (Fig.4).

Calcium Sulfo Aluminate (CSA) Crystal shows cubic shape composed of Al-O hexagonal group, form axial hole path, and a square column of Ca-O on the axis of the unit cell. The CSA preparation conditions and advantages are given in the Table -4 and 5.

In the case of products can be developed over medium heat isolation material properties are the same as commercially available product segregation will not occur. Excellent early strength characteristics of one day and three days of product development.

5. Conclusions

Cement and concrete materials are well known alternatives to bentonite for the radioactive disposal storage. Coal fly ash is main raw material in cement or aggregated that would be used in radioactive storage. Calcium sulfo aluminate called green cement manufactured from coal ash has wide advantages than commercial cement. The coal fly ash applications used as barrier layers/buffers for high level radioactive wastes are under investigations. We measured the mechanical properties and hydration tests, permeability etc showed significant properties. So, the CSA is highly beneficial and more feasible to use as an obstruction/barrier material for radioactive waste storage.

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References

1. Nuclear Energy Agency (NEA); 2012, Nuclear Energy Today, Nuclear Development, Second Edition, ISBN 978-92-64-99204-7, 1-123.
2. International Energy Agency (IEA); 2017, Electricity Information: Overview, Statistics, 1-8.
3. MZ Consulting, 2017, Europe roadmap 2050; As a Solution for Climate Change-Nuclear Power is Falling Behind., 1-12.
4. Nuclear Energy Agency (NEA); 2015, Technology Roadmap Nuclear Energy., 1-64.
5. Swedish National Council for Nuclear Waste. 2016, Yttrande över Svensk kärnbränslehantering AB:s ansökan om tillstånd enligtmiljöbalken iett sammahängande system för slutförvaring avanvänt kärnbränsleoch kärnavfall., (Dnr Komm2016 / 008 58 /M1992:A).
6. Swedish National Council for Nuclear Waste. 2016, SOU 2016:16 Nuclear Waste State-of-the-Art Report 2016. Risks, uncertainties and future challenges. Stockholm: Wolters Kluwer. 1-170.
7. Swedish National Council for Nuclear Waste. 2015, SOU 2015:11 Nuclear Waste State-of-the-Art Report 2015. Safeguards, recordkeeping and financing for increased safety. Stockholm: Fritzes. 1-165.
8. Swedish National Council for Nuclear Waste. 2014. SOU 2014:42 The Swedish National Council for Nuclear Waste's Review of the Swedish Nuclear Fuel and Waste Management Co's (SKB's) RD&D Programme 2013. Swedish National Council for Nuclear Waste. Stockholm: Fritzes. 1-101.
9. Swedish National Council for Nuclear Waste. 2012, SOU 2012:7 Nuclear Waste State-of-the-Art Report 2012 - long-term safety, accidents and global survey. Swedish National Council for Nuclear Waste. Stockholm: Fritzes. 1-104.
10. C. Pescatore; Long-term Records, 2016, Memory and Knowledge Preservation - Recent thinking

and progress in the field of geological disposal of radioactive waste, and further avenues of research., 1-18. www. Karnav falls ra det.se.
11. Status and Trends in Spent Fuel and Radioactive Waste Management, 2010, IAEA.
12. Radioactive Waste in the UK: A summary of the 2010 Inventory, Nuclear Decommissioning Authority 2010, 1-12.
13. J.T.Carter, A.J. Luptak, J. Gastelum, C. Stockman and A. Miller, 2012, Fuel Cycle Potential Waste Inventory for Disposition. FCRD-USED-2010-000031, Rev 5. Washington, DC: U.S. Department of Energy.
14. Haiyong Jung, 2013, Current Status, Experience and Planning of radioactive Waste disposal in Korea, Presentation, IAEA TM on the Disposal of Intermediate level Waste, vienna, 9-13,.
15. Michael I. Ojovan, 2015, IAEA Activities Related to Radioactive Waste Predisposal Management, Waste Technology Section, Department of Nuclear Energy, IAEA, 1-50.
16. UNITED NATIONS, 2000, Sources and Effects of Ionizing Radiation (Report to the General Assembly), Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), UN, New York.
17. Radioactive Waste Management Committee (RWMC), 2013, AEN, NEA, OCDE, Underground Research Laboratories (urls) and Geological Disposal of Radioactive Waste, 1-2.
18. ONDRAF/NIRAS, 2012, Cementitious Materials in Safety Cases for Geological Repositories for Radioactive Waste: Role, Evolution and Interactions, NEA/RWM/R(2012)3/REV, March.
19. Jan Tits, Xavier Gaona, Rainer Dähn, 2012, Dmitry Popov and Erich Wieland, Immobilization of np in Cementitious Repository Environments, Radioactive Waste Management, NEA/RWM/R(2012)3/REV, March
20. NEA and EC, 2010, The Joint EC/NEA Engineered Barrier System Project: Synthesis Report (EBSSYN), EUR XXXXXEN. Luxembourg: European Commission. Paris: OECD.
21. Takashi Sasaoka, Wahyudi Sugeng and Hideki Shimada, J.Geo. 2015, Env. Pro., 3, 31-43.