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TECHNOECONOMIC ASSESSMENT OF THE CARBOTHERMIC REDUCTION PROCESS FOR ALUMINUM PRODUCTION

William Choate¹, John Green²

¹BCS Inc.; 5550 Sterrett Place Suite 306; Columbia, MD 21044, USA ²JASG Consulting, 3712 Tustin Road, Ellicott City, MD 21042, USA

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Abstract

In pursuit of the aluminum industry Vision and Roadmap goals, the Department of Energy has partially supported a consortium of Alcoa and Elkem in the development of the Aluminum Carbothermic Technology - Advanced Reduction Process (ACT-ARP), which promises significant energy and emission reductions. This report explores the progress of the ACT-ARP as a potential replacement for the Hall-Heroult process in the context of several evolving Hall-Hèroult development scenarios. Considerable progress has been made and demonstrated, including new furnace wall designs integral to successful operation of Stage 1 reactor, operational characteristics of vapor recovery reactor and aluminum decarbonization reactor, as well as significant modeling and simulation. Despite these considerable accomplishments, there are still formidable technical and economic challenges to overcome before the ACT-ARP can replace the conventional Hall-Hèroult process, such as slag and scale formation, metal and carbon quality issues, mini-mill operation, etc. All these and other issues will be discussed.

Introduction

The purpose of this report is to review the current iteration of the carbothermic production of aluminum and assess its future potential as replacement primary production technology. This summary report has been drawn from a larger internal report to the Program Manager of the Aluminum Industries of the Future program, an initiative of the Department of Energy (DOE) Industrial Technologies Program (ITP). The original report was submitted to DOE in May, 2005 [1].

The major goal of the DOE-ITP is to lower energy consumption and decrease environmental emissions in the manufacturing sector. To this end, DOE has sponsored projects with industry, on a cost-shared basis, to develop more energy-efficient production and manufacturing processes. Historically, the primary production of aluminum has been energy intense and several approaches are currently being pursued worldwide to reduce its energy intensity. In the United States, these projects have been guided and focused by industrial Vision and Roadmap documents that have set forth energy reduction and emission (sustainability) goals through the year 2020.

For the past 4 1/2 years, in the pursuit of the Vision and Roadmap goals, DOE has partially supported a consortium of Alcoa and Elkem in the development of the Aluminum Carbothermic Technology - Advanced Reduction Process (ACT-ARP) which promises significant energy and emission reductions. Recently, DOE program management requested a technoeconomic assessment of this process as part of its due diligence management procedures.

This report responds to the DOE's request and reviews the carbothermic technology progress, critical issues, competing aluminum production approaches and improvements, a proposed business model, and related issues. While this report does not have the advantage of access to proprietary information from the technology developers, efforts have been made to gain insights from a variety of industrial stakeholders, and from the literature. Also, attendance at TMS 2005 provided numerous opportunities for in-depth discussions related to aluminum industry trends, issues of metal and coke quality, carbothermic reduction procedures, the concept of minimill operations, the impact of China in the marketplace, and numerous related issues.

Background

In 1990, the total U.S. aluminum metal supply of 7,833 thousand tonnes was made up of 51.5% primary production with imports of 18.1%, and the balance being from secondary recovery. Following the energy shortfall in 2001, when all 10 smelters in the Pacific Northwest were shut down, the U.S. metal supply in 2002 was made up of only 28.5% primary production with 40.8% imports, and the balance from secondary recovery [2]. In addressing these unfavorable metal supply trends, the industry roadmap and vision documents established priorities in advancing both the conventional reduction process, through the development of inert anodes, wetted cathodes and drained cells, as well as through alternative reduction processes. Carbothermic reduction technology was considered to be the top priority in the category of alternative processes and while this process was considered to have high technical risk, it offers sizable benefits - reduced energy, capital and operating costs and a smaller plant footprint. Fig.1 shows the assessment of the carbothermic process from the industry roadmap published in 2003 [3].

This report explores the development of the Alcoa / Elkem Carbothermic Reduction (CR) process [4, 5] for aluminum reduction as a potential replacement to the traditional Hall-Hèroult (HH) process, in context of other developments. This is appropriate in that technology development never occurs in isolation. Incumbent technologies, when challenged, seem to be able to improve their energy efficiency. Also, the traditional HH process, with its roots in the United States and France, is now being significantly impacted by the interests of developing nations with more abundant low-cost energy and labor.

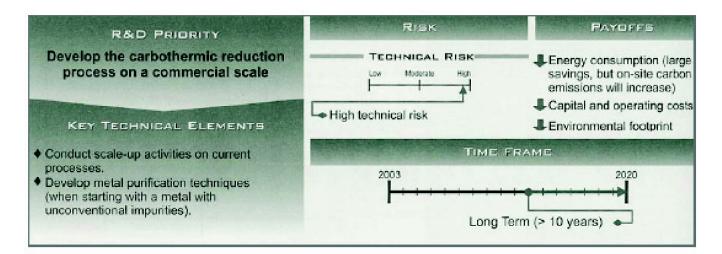


Figure 1. Roadmap assessment of carbothermic reduction process

At the project outset, (July 2000), the Alcoa / Elkem team claimed that the CR process offered several technical and economic advantages. For instance, it was claimed that the CR process has the potential to reduce energy consumption by 38%, environmental CO2 generation by 37% and capital costs by 70% [6]. In addition, at the 2004 Aluminum Project Review, it was proposed that the CR process could be operated as a "mini-mill" at a minimum level of 35,000 tonne/year. This would offer extensive benefits in plant flexibility, location and siting as compared to the traditional HH smelter, which requires large quantities of electricity and thus, is generally sited near a suitable source of hydroelectric power or other low-cost power. By contrast, the CR process relies on carbon (petroleum coke) at high temperatures to reduce alumina to aluminum.

Briefly, the CR process involves reactions of carbon with alumina at temperatures around 2000 °C. The current iteration (there have been several unsuccessful attempts over the past 40 years by the industry worldwide) by Alcoa and Elkem takes advantage of new high-intensity electric arc furnace technology, advanced thermodynamic and system modeling techniques, and an improved understanding of the process enabled by extensive modeling. A two-stage reactor concept is involved, together with a vapor recovery reactor (VRR), to ensure sufficient completion of the complex and reversible chemical reactions to guarantee that the process is both technically and economically viable [7]. Also, an aluminum product de-carbonization chamber is required to purify and cool the molten metal sufficiently prior to sale and application. The conceptual design of the Alcoa / Elkem Advanced reactor process is shown in Fig.2

The Alcoa / Elkem team indicated that the energy requirements for aluminum production could be as low as 8.5 kWh/kg, an improvement of 5.5 kWh/kg compared to the best U.S. HH cells, and also a net reduction 6.4 kgCO2 per kg Al (\sim 30%) would be possible.

Where's the Competition – Smelter Development Scenarios? Technology is rarely developed in isolation and accordingly it is instructive to examine developments in the established HH technology. In the history of technology development, incumbent processes, when challenged, always seem to be able to make efficiency improvements and modifications. Though the underlying principle of the HH electrolytic process has remained unchanged since 1886, the scale and sophistication of the present day electrolytic cell is hugely different from those at the outset of the industry. So how is the incumbent technology developing and how is it changing in response to the needs of the marketplace and the challenge of potential competitive technologies? Also, what are the likely scenarios for future developments in the primary production of aluminum?

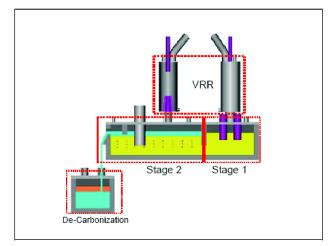


Figure 2. Alcoa/Elkem Advanced Reactor Process Conceptual Design

The conventional HH technology continues to make incremental advances in energy and environmental efficiency. According to the International Aluminum Institute (IAI) website [8], the industry set a goal to achieve a 10% reduction in smelter energy per unit of production over 1990-2010; by 2003, a 6% improvement had already been achieved, partly from a reduction in the frequency of cell anode effects (a process upset that decreases productivity and causes emission of perfluorocarbon (PFC) gases).

<u>Scenario 1: Incremental Improvements in Existing HH Plants.</u> The ongoing improvements incorporated in the IAI website data will undoubtedly continue where the modifications only need relatively minor capital expenditures. Some existing smelters may still add "point feeders", increase anode size, add insulation, adopt anode slots to improve the release of anode gases, or adopt wireless sensing technology to improve cell control and performance, etc. However, it is difficult to visualize older cells with such developments operating much below today's best cells at ~13kWh/kg.

<u>Scenario 2: HH plants with Horizontal Wetted Cathode</u>. The addition of titanium diboride (TiB₂) to the cathode, either as a thin layer or as a thicker coating in a carbonaceous binder has been shown to reduce the voltage drop across the ACD [9], generally by ~0.3V. This saving occurs through a better wetting of the cathode and a more uniform magnetic field within the ACD. The application of the TiB₂ also appears to reduce the penetration of sodium ions into the cathode and increase cathode life. The most significant factor slowing more widespread application of wetted cathode technology is the cost of TiB₂ particulate material.

Scenario 3: HH Plants with Wetted and Drained Cathodes. Here the cathode is slightly inclined and the metal drains off the TiB₂ wetted cathode to an adjacent sump. While the metal drains under the influence of gravity, the anodic gas bubbles, being less dense, ride up the anode slope and draw saturated bath into the ACD to maintain alumina supply and the cell reaction. The draining of the cathode is an extension of Scenario 2 in that the metal pad is reduced in thickness and its stability is further increased. By removing the molten metal to a metal sump, there is a more stable metal pad and less risk of shorting; consequently, the ACD can be further reduced, possibly down to ~ 2.5 cm, resulting in a corresponding energy saving. At TMS 2005, it was confirmed that Rio Tinto (Comalco) has restarted full-scale production from TiB2-drained cathodes at the Bell Bay smelter (this was first achieved during the 1980's), and was again producing metal from drained cells. The performance of the drained cells apparently was similar to that achieved previously at Bell Bay. Figure 3, taken from this reference [9], compares the performance of the drained cathode cells with the conventional side- or center-fed cells and with magnetically compensated point fed cells. It can be seen that compared to the magnetically compensated cells on the 5 cm ACD dotted line, the drained cathode cells close to the 2.5 cm line had a metal productivity that was about 25-40% greater at a specific value of kWh/kg metal produced, as indicated by the higher amperage.

It was also considered significant that the TiB_2 coated cathode blocks for the Bell Bay cells were being supplied by the Aluminum Corporation of China, CHALCO [10].

<u>Scenario 4: Inert Anodes</u> Ceramics, cermets and metals have all been researched as potential inert anode materials for the HH cell. A truly inert material would provide a couple of major energy efficiency advantages. First, the geometry of the ACD could be more precisely controlled and narrowed to give energy savings. Second, the gas emitted by the cell would be oxygen instead of the present CO_2 , thereby improving the climate change situation and perhaps generating a more usable off-gas. Unfortunately, the fact that carbon is not used means an additional 1 volt is needed to conduct the electrolysis process. Alcoa's recent development work on inert anodes was much discussed in 2000 [11], but since then, there has been a disquieting silence about the development. Problems seem to center around the robustness of the inert material and hence, its impact on metal quality (any erosion or wear of the anode material inevitably contaminates the metal quality), and the ability to make good electrical connections to it.

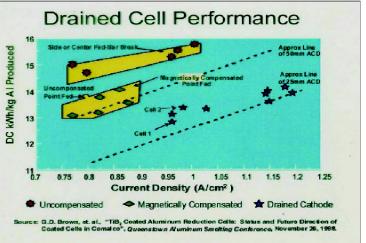


Figure 3. Comparison of Performance of Drained Cells with Regular Cells [9]

Scenario 5: Carbothermic Reduction Process details for this process can be gleaned from references [4, 5, 6]. Here, the alumina is reduced chemically, rather than electrochemically as in the HH process, and so plants potentially can be located closer to industrial centers. Also, the volumetric nature of the chemical reactor offers considerable production efficiencies as compared to the planar electrode geometry of the HH plants and gives it a much smaller plant footprint. Alcoa has committed to build the first unit in the U.S. and estimates the technology could be implemented by 2010-2012 [6].

Scenario 6: Slowing Down or Abandoning Technology Development and Investing Overseas. It is possible that the various materials, engineering and design difficulties associated with advanced HH cells or the extreme temperatures of the CR reactor may prove to be too intractable or uneconomic in the near term. In this scenario, industry could choose to grow by investing in additional modern HH smelting capacity overseas, where there is better energy availability at lower cost, lower labor costs, less stringent environmental regulations and possibly incentives as well. It is interesting to note that the industry leader, Alcoa, during 2004 has committed some ~\$2 billion to new smelter projects in Iceland (geothermal and hydropower) and Trinidad (natural gas). This scenario is not without precedent. In the mid 1970s and early 1980s, the Japanese with the tacit approval of MITI, the government department responsible for trade and industry, eliminated all their smelters and chose instead to focus their limited energy resources on the semi-fabricated and fabricated products aspects of the aluminum industry. Some 12 smelters closed during this time.

<u>Scenario 7: Multipolar Cells and Scenario 8: Ionic Liquids.</u> These scenarios are considered too embryonic to be discussed further in this context.

Carbothermic Reduction Process

The overall chemical reaction for CR is shown as:

$$Al_2O_3 + 3C \rightarrow 2Al + 3CO$$

However, in this scheme, CR of alumina to aluminum is proposed as a multi-step, high temperature, chemical reaction process. The necessity for this arises from the fact that when carbon (presumably petroleum coke) is mixed with alumina, formation of aluminum carbide (Al_4C_3) is favored over aluminum metal at lower temperatures [4]. Therefore, it is necessary to have two reaction stages with different temperatures, see Fig.2. In the first stage, alumina and carbon are heated to ~1900°C using two vertical electric arc electrodes, where the following reaction occurs:

 $Al_2O_3 + C \rightarrow (Al_4C_3 - Al_2O_3) (slag) + (CO-Al_2O-Al) (g)$

The high density slag underflows into the Stage 2 reactor, where it is further heated by pairs of horizontal electrodes, to a temperature necessary to produce aluminum (2100-2200°C). This stage of the process results in the formation of an aluminum-carbon alloy by reduction of alumina with aluminum carbide at temperatures above 2000°C.

$(Al_4C_3-Al_2O_3)$ $(slag) \rightarrow (Al-C)$ $(alloy) + (CO-Al_2O-Al)$ (g)

The aluminum metal produced in Stage 2, being lighter than the slag, floats to the top of the reactor (shown as blue in Fig.2) and passes through a weir to permit continuous removal to a decarbonization chamber. At this stage the metal is extremely hot and saturated with aluminum carbide. For process efficiency the heat needs to be captured, probably by remelting additional scrap. This has the additional benefit of lowering the carbide content of the impure metal. Further reduction of carbide is achieved by precipitation upon additional cooling and subsequent filtration; the Al_4C_3 is recycled to Stage 1. The Vapor Recovery Reactor (VRR) is probably the critical process step in that Al_2O and Al vapor must be recovered for efficient operation of the process. Only CO is intended to pass through the VRR, where the following chemical reactions occur:

$$2Al_{2}O(g) + 5C(s) \leftrightarrow Al_{4}C_{3}(s) + 2CO(g)$$
$$4Al(g) + 3C(s) \leftrightarrow Al_{4}C3(s)$$
$$2Al_{2}O_{3}(l) + 9C(s) \leftrightarrow Al_{4}C_{3}(s) + 6CO(g)$$

All these reactions produce both solid and liquid products and all reactions are reversible. To the extent the reactions are indeed reversible there will be proportionally greater need for additional carbon consumption. Accordingly careful process control is vital to minimize the generation of volatiles, avoid back reactions and recover the aluminum gas phase components efficiently.

Summary of Process Developments and Accomplishments

The Alcoa/Elkem team has indeed made several major advances in the CR technology [6]:

- Demonstrated Stage 1 of reactor at 1MW
- Demonstrated efficient use of oil-cooled copper sidewalls to control freeze linings critical to materials of construction issues
- Developed side-entering electrode seal assemblies and demonstrated successful operation, critical to Stage 2, to minimize Al vapor losses and avoid potential short circuiting
- Determined operating characteristics of vapor recovery reactor design concept for Stage 2 in 100 kW system
- Developed efficient de-carbonization process for removal of carbon to 0.15% or less at high recoveries
- Experimentally determined the mass transfer coefficients controlling vapor recovery reactions
- Experimentally measured the vapor pressures of Al and Al₂O species at operating temperature and utilized the results to verify the published thermodynamics for the Al₄C₃-Al₂O₃ system
- Developed a working model for simulation of a dynamic vapor recovery reactor

• Maintained economic estimates throughout the project. This represents a considerable list of accomplishments, though several major issues remain to be resolved as described in the next section.

Critical Process Development Issues

- Several keys issues still need to be addressed:
- Project metrics—in this area, the benefits of CR have been generally compared with a baseline HH value of 14.65 kWh/kg, which is now a dated value compared to the world's best magnetically-compensated cells operating in the range of ~13.0 kWh/kg [12]. The fact that the US smelters are relatively old unfortunately tends to skew the average data. Also, any comparison must take into account future improvements to the HH process that have been occurring at an annual rate of ~ 0.1kWh/kg. This corresponds to an additional 0.7 kWh/kg by the year 2012 when the CR potentially may be available.
- Metal quality will be critical since the industry requires the quality of the metal to be identical irrespective of its source, i.e. primary or secondary. On a theoretical basis, for each unit of metal production, the CR process consumes twice the amount of carbon as compared to the HH process. Already, the carbon source, presumably petroleum coke, is currently a major source of Fe, Si, Ni, V and S impurities in the HH process and the quality of the coke is continuing to deteriorate. For example, levels of V have doubled to 800ppm over the period 1983 -1998 and these trends are continuing. Ni and V also are known to be catalytic in nature and may complicate the reversible chemistry in the VRR step of the process. Another concern is that the gas stream from the VRR may be contaminated by impurity particulate and / or by SO₂ gas, thereby lowering its utility as a product. Further, it is possible that carbonyl sulfide (COS) and nickel carbonyl (Ni(CO)₄) may be formed; both these gases are hazardous and will require a mitigation plan to be in place.
- Carbon Reactivity—here the key issue is that as the CR proceeds in the VRR reactor, the formation of Al_2O_3 - Al_4C_3 slag occurs first at a temperature of ~1930°C, and then the needed carbide, Al_4C_3 , is formed at the higher temperature of

~1950°C. The key question is how the formation of slag will retard the kinetics of the subsequent reaction to form the needed Al_4C_3 for the overall CR process to proceed. The Alcoa /Elkem final report [13] did not provide reassurance on this issue of carbon reactivity.

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- Sensors and Controls—a robust control system will be essential especially of the VRR unit where the temperatures are extreme (2100-2200°C) and the chemistry is complex and reversible. The R&D team has been diligent in developing simulation models of the VRR unit. However, one issue will be how the formation of slag and scaling will compromise any control system. Reactor scaling will be aggravated by the presence of trace impurities of sodium and calcium in the feed alumina.
- Environmental Impacts—more recent estimates of environmental benefits of the CR process, as compared to the HH process, presented at TMS 2005, have narrowed somewhat. For example, the potential power consumption delta improvement has reduced to 21% or 26%, depending on whether the combustion of CO byproduct is included. This contrasts with a value of 38% at the outset of the project.

Mini-Mill Operation

Much has been made of the flexibility and marketing advantages conferred through a mini-mill operation. It would enable the industry to break the traditional connection between the smelting process and the need for nearby low-cost and abundant power supplies, and locate production operations adjacent to fabrication and assembly plants with potentially considerable energy savings. The term "mini-mill" was made popular by the steel industry and the aluminum industry has not adopted the jargon. However, in a sense some scrap based remelting operations (e.g. Aleris International) operating close to manufacturing centers like Detroit are already mini-mill operations. Thus, in a real sense, the economic competition for the CR process is not today's HH smelter but is probably the small remelt operation located adjacent to an industrial center. This is probably a tougher competitive situation for CR technology since, on a historical basis, the cost of secondary material to feed the remelter has generally been less, by several cents per pound, than the cost of primary sow or ingot metal. Of course, this is currently a debatable issue and depends on the availability of scrap and how much the price of scrap will be perturbed over a longer term by the activities of China. Assuming that a CR mini-mill is technically viable, Choate et. al. [14] undertook a survey of the US fabrication industry and concluded that 31 rolling mills, representing over 76% of the rolling market, have sufficient capacity to support the installation of a 40,000 tonne/yr mini-mill. Likewise, 19% of extrusion operations and 74% of casting plants could justify the installation of an aluminum mini-mill. The small footprint of a 40,000 tonne/yr CR mini-mill, estimated as having overall dimensions of 100x120 meters, would be helpful in siting plants near manufacturing centers.

Economic Comparison of Technologies

To assess the commercial viability of the CR process, Figure 4 illustrates the worldwide aluminum industry costs for all smelters for the years 1990, 1997 and 2003 [15]. This figure illustrates how aluminum producers have been able to increase efficiency and make productivity gains. From 1990 to 2003, the curves have moved to the right, indicating more capacity as well

as downwards, indicating reduced costs. This is consistent with the view that incremental improvements in HH technology will continue, and shows that CR technology will face increasingly difficult competition as time passes. As of 2003, Figure 4 indicates that the best plants produce aluminum at \sim \$850/tonne – a reduction of \sim 10 cents/lb over about 13 years for the newest plants. This is the cost that CR must compete with to succeed in the marketplace.

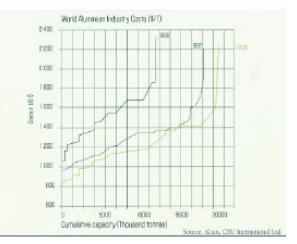


Figure 4. Cost Curves of World Smelters for 1990, 1997, & 2003 [15]

To continue the economic comparison of technologies, a more detailed economic analysis has been undertaken [16]. In this analysis, six specific smelter production technologies are compared: World Class Hall-Hèroult, North American Hall-Hèroult (both new and fully depreciated plants), secondary (remelt) plant in North America, carbothermic plant at proposed minimum size (35,000 tonne/yr) and a larger carbothermic plant (70,000 tonne/yr). The analysis considers both capital and operating costs, including materials and labor, and fixed costs as well as including a 10% profit. The estimated price/lb of aluminum produced by each route is shown below in Table 1.

Table 1. Comparison of Metal Costs by Plant Type	
Aluminum Production Path	US\$/lb
Hall-Hèroult, Non-American World Class	\$ 0.68
Hall-Hèroult, American "greenfield"	\$ 0.85
Hall-Hèroult, American "fully capitalized"	\$ 0.75
Secondary Metal, American	\$ 0.73
Carbothermic, Minimum Economic Size	\$ 0.66
Carbothermic, Twice Minimum Economic Size	\$ 0.63

Generating these data obviously involves making numerous assumptions for labor, energy and materials costs, etc. Many assumptions for world labor and materials costs were drawn from the recently published EAA Sustainability Report [15]. Despite the gross nature of the analysis, it appears to be sufficiently consistent to account for current global trends. For example, a new plant in the United States would deliver metal at some \$0.17/lb more than a new world class investment, explaining why there are no smelters being planned in our country. The existing smelters in the country have lower metal costs (\$0.75/lb) but still not as low as the world class investment. The secondary metal (recycling) plant has a lower metal cost than the HH plant, which is consistent with the more rapid growth of this portion of the industry. However, in both cases, the CR process potentially offers the lowest metal production costs. Doubling the proposed minimum economic size offers economies of scale and is estimated to provide metal at the lowest value of \$0.63/lb.

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Conclusions

All these data are consistent with the view that the CR process would be economically attractive and viable, provided it can be made to be technically feasible. Probably the biggest threat to the technical feasibility of CR comes from incremental developments and/or a retrofit to the HH technology of a drained, wetted cathode or inert anode system. Other formidable challenges stem from the extremely high temperatures of the CR process, in excess of 2000°C. In addition to materials and containment issues, one must anticipate difficulties due to slag and scale formation, complex and reversible chemical reactions, and process control. Issues of metal quality will become increasingly intractable as the levels of impurities such as Ni, V, and S increase in supplies of petroleum coke. These impurities will probably adversely impact VRR chemistry and the quality of byproduct gas streams. Lastly, the CR development will challenged economically by the evolution of the secondary aluminum industry itself, which has already developed the mini-mill concept by locating remelters close to major fabrication and assembly operations. In considering all the advantages and disadvantages of the CR process as it seeks to replace the incumbent Hall-Hèroult technology, which though mature still continues to evolve, it is considered unlikely that the CR process will be sufficiently mature and successful to replace the traditional HH process, at least before the year 2020.

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